## SYSTEMS AND INDUSTRIAL ENGINEERING SERIES



# 4D Printing 2

Between Science and Technology

Frédéric Demoly Jean-Claude André





4D Printing 2

Series Editor Jean-Charles Pomerol

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### Foreword

Jean-Claude André, who has already provided us with three very insightful books on additive manufacturing, is taking it up a notch! Indeed, with the help of Frédéric Demoly, he now offers us an "encyclopedic" book on 4D. Let us recall that Jean-Claude André is the first Frenchman to have filed patents and to have believed in what we now call "additive manufacturing", which was just called "3D printing" at the time.

The fourth dimension is that of time, and the question we face is that of producing, in additive manufacturing, objects capable of changing shape or function over time. In old and simplified terms, we could say that it is a question of using shape memory materials or materials that can be deformed under the effect of external, thermal, photochemical, mechanical or electromagnetic disturbances in 3D printing. Biomaterials are also included in 4D, whether they are biocompatible or living materials, the latter being referred to as bio-printing. This important theme is hardly if at all developed considering the 4D field seems to be so important in its future achievements.

This book covers several aspects. First, it can be read as a classic science book that explains the state of the art and tells us everything there is to know and everything that is being done in 4D printing. Then, it goes through all the problems that arise for the designers of artifacts in the field: mechanical problems, material problems, choice of applications and industrial problems. Finally, this book can be read like an adventure novel. Indeed, it follows the research adventure step by step; the difficulties, the obstacles and the facilitators (or even the impediments to straight thinking) that researchers encounter on their way. It encourages reflection on creativity in research and the role of breakthrough innovations. The historical overview of inventions sheds light on the (sometimes impenetrable!) paths and methods of research, which lead to a profound epistemological reflection on research and innovation in the context of successive technological revolutions over several centuries.

In particular, our authors shed light, in a very original way, on the notions of rupture or "disruption" to use a fashionable term. Are 3D printing and now 4D printing disruptions or the intelligent use of known techniques? Will 3D end up imposing itself in manufacturing, thus opening the way to frequent use of 4D? These are some of the questions raised by the industrialization of additive manufacturing and its multiple applications.

Moreover, the authors do not avoid any question concerning the impact and social acceptance of new technologies and their applications. All the industrial applications that can already be envisaged are described, while taking care not to act as if the story is over (in fact, it has only just begun) and as if, consequently, the creative process at work in 4D research will hold no more surprises for us.

Those who have read Jean-Claude André's books on 3D know of his enormous erudition, which he once again makes available to us by peppering the book with numerous quotations, often delightful, always very instructive and inspiring. All of this is accompanied by an extensive bibliography, very useful for students and researchers alike.

From this book, the reader will not only learn everything there is to know about 4D, the state of the art, materials, applications, developments and problems, but will also understand how research progresses between incremental development, creativity and disruption. It is a whole world, full of reflections on science and its epistemology in relation to creativity and innovation, which is revealed to readers by two great researchers, pioneers of additive manufacturing in all its dimensions.

Jean-Charles POMEROL President of the AGORANOV incubator and of the scientific council of ISTE Group April 2022

## Getting Things Moving

Manufacturing, but not action or speech, always involves means and ends; in fact, the category of means and ends derives its legitimacy from the sphere of making and manufacturing, where a clearly reasonable end, the final product, determines and organizes everything that plays a role in the process – the material, the tools, the activity itself, even the people who participate in it; all of them become mere means to the end and are justified as such. (Arendt 1972)

#### Warning

In the 16th century, in his treatise on magic, Giordano Bruno had, among other ideas, a certain notion of reactivity and consequently of adaptive materials. Thus, he wrote:

If one resorts to the virtue of sympathy and antipathy of things, as when substances repel, transmute or attract other substances [...], one rightly speaks of extra-natural magic. [...] From these premises, it can be deduced that the magnet stone attracts by its very nature. Indeed, attraction is twofold: certain objects attract first of all by sympathy, as when parts move towards their whole, when what has a definite place joins its place... (Bruno 2020)

This chapter tries, however, to move away from any magical context by examining, as scientifically as possible, how by various stimulations a form can change spatially or in functionality, alone in a homogeneous way or by association with materials.

4D Printing 2: Between Science and Technology,

For a color version of all the figures in this chapter, see: www.iste.co.uk/demoly/4Dprinting2.zip.

First Edition. Frédéric Demoly and Jean-Claude André.

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#### 1.1. Introduction

The introduction of additive manufacturing in the 1980s transformed the manufacturing industry. This technology, despite its success, still has some drawbacks, such as printing speed, surface finish of the final product and lack of fully functional materials (Manikandan et al. 2021). 4D printing uses smart or active materials to achieve a change in form or functionality. Materials that are sensitive to certain stimuli, the stimuli themselves and the time required for the change in state to occur are key research areas in 4D printing.

This chapter summarizes the developments in 4D printing, with a focus on the materials and methods used for the manufacture of 4D printed objects and structures. Smart or active printed structures can be disrupted in a custom manner by external stimulus such as water, heat, electricity, light and solvents with different pH values. Although printed structures exhibit self-healing, self-diagnosing, self-acting and self-sensing capabilities, etc., this chapter illustrates attractive possibilities and also highlights some difficulties. The field is not yet mature and still needs further research.

Innovations, such as new tools, devices and processes, can bring significant benefits to society by providing elements of comfort, means of medical treatment, etc., all in conditions of frugality imposed by a crying environmental need. Only then will innovations become inventions that can help the world's citizens live better, longer, healthier, more productive lives, and provide new ways to build, move, communicate, heal, learn and play. Understanding and clearly communicating the value of innovation (not the promises of it) can help decision-makers understand the benefits of supporting initiatives for their development. With 4D printing, we are at the beginning of a new scientific and technological adventure and the reality principle must be taken into account.

The Rand Corporation (2021) highlights three lessons:

- The impact of innovations goes beyond the direct effect of the invention itself; it can serve as an inspirational image for other fields.

- Success stories, which must be seen by the public, can help solve challenges facing society.

- But, it is not enough to create an invention that must meet the challenges of industrial application and manufacturing to satisfy a potential public demand.

This is another book that all employees on the verge of burnout are snatching up and running to buy in their bookstores, like a crocodile rushing into a leather shop. (Vervisch 2020) However, before reaching this stage, we must go beyond simple proofs-ofconcepts (POCs). However, in Chapter 1 of Volume 1 we have shown a significant difficulty linked to the control of the deformation of complex shaped objects coming out of the basic origami. Methods of "successive approximations" have thus been proposed to reach a certain satisfaction in transformable devices, allowing for the adjustment of ends and means. If Lippmann (1937) wrote:

Technical progress, experimental in nature, requires much trial and error [...]. For these great centralized controls [...] are not suitable for a system of production which can only make a profit by new invention if it is flexible, experimental, and competitive,

this is only true if one has properly circumscribed the subject to avoid wasting time.

In order to do this, we felt that it was important to start from the most robust bases possible, in order to attack a market that still expects little or nothing from 4D printing. One of the elements that has already been widely discussed (because it is essential) is the effect of a stimulation to transform cohesive matter – thus materials – (we are obviously not talking about chemistry, which could claim this definition, nor about cooking) by providing a new functionality and/or a deformation. We know from Tahon (2003) that, despite (or because of) an increasingly unstable demand, unless we change the business model, there will continue to be pressure to produce as quickly as possible, and the diversity of products will increase. It will also be a matter of being ready on time in 4D printing.

Thus, the purpose of this chapter is to provide an overview of the means, of whatever nature, which make it possible to reach this objective. On the basis of the proposals, it will then be possible to evoke exploitation paths of these means for 4D printing applications. It is at this (partial) price that we will know if it is really a potentially attractive "disruptive technology". The concept of disruption was introduced and argued by Christensen (1997) because he recognized that few technologies are intrinsically of a disruptive or continuative nature. It is, according to Bessen et al. (2020), their use that manifests a disruptive effect.

In André (2017), a chapter was devoted to 4D printing and transformation processes under stimulation. These data and presentations are re-used here with (legitimate) enrichments, following recently published work, as the background from four years ago is considered robust.

Box 1.1 General note

It is true that the test of causality – the predictability of the effect, if all the causes are known – cannot be applied to the field [...]. But this practical unpredictability is not proof of freedom; it simply means that we are never in a position to know all the causes that come into play. (Darwin 2020)

#### 1.2. Actuators

2,000 years before our time, Sumer imagined that the gods created humans in the form of animated clay statues, so that they could work in their place (principle of intentionality). This is the active matter side. But this myth would be found, according to Couveinhes (2012), in the legend of the Golem: the clay creatures could be animated thanks to inscriptions, as well as by a scroll. There would thus be an association between active matter and information. The central question in this chapter is to know how these androids can move (and not why). According to Wikipedia (2021), an actuator is an object that transforms the energy supplied to it into a physical phenomenon that provides work, and modifies the behavior or the state of a system. To do this, the initial energy can be pneumatic, hydraulic, electromagnetic, mechanical, photochemical, electrical, thermal, chemical, biochemical, biological, etc., to finally transform into mechanical energy and provide work. The link sought is therefore the actuator or engine that connects matter to energy for practical actions.

Robots are platforms for the integration of advanced technologies: energy storage, support and active materials, information and acquisition, data processing and transmission, mechatronics, sensors, etc. These are autonomous systems for producing a service. "The convergence of advanced technologies and their increasing interoperability, resulting from dematerialization, make the robot a fertile ground for innovation, cost reduction, and new uses" (Roure and Arcier 2012).

#### Box 1.2. Remark on robots

Thus, an actuator is a device capable of producing (and reproducing) a physical phenomenon such as a displacement, heating, an emission of light, sounds and a change of functionality. It is a transformer of one form of energy into another.

#### 1.2.1. General information

Culture is nothing more than the transmission and sharing of representations that are temporal discretizations produced by the techniques of a given era. Telling is ultimately about organizing temporal discretizations in time. (Kaplan 2012)

While looking in the past for traces concerning actuators (before including them in additive manufacturing), we wished to give a quick overview on this aspect. While, a long time ago, industrial activity was focused on a centralized energy: windmill, water mill, etc., steam, the explosion engine and especially electricity allowed us to include or distribute engines and actuators in any object whatever its size, as a way to transform, at will, usable energy flows into spatiality. In classical technologies, discrete elements are integrated into each other to form a system that will operate a temporal change; with 4D printing, we hope to move from the "quantum" to the continuous, from an ancestral and robust technical know-how to a more innovative concept of a new mastery of matter. So, the possibility of repair in this new field imposes level maintenance integrated into the process itself of creating an object. We no longer replace elements: there is no need for skills, standardized modes of communication, disassembly, storage, maintenance, etc. This is a future that could, independently of the other performances associated with 4D printing, constitute a real disruptive leap in the technological functioning: we no longer repair, but we throw away! It would therefore be the "smart" use of the object that would paradoxically create value and not the object itself, which is becoming more or less invisible! Moreover, 4D objects are connected objects:

[They] do not pose any problems as such, what challenges us is the extent to which we have become, willingly or not, connected subjects, and therefore disconnected. (de Brabandère 2017)

But this is another story, for later.

Technical and formal innovation has involved and combined, in one way or another, a theoretical reflection that is now mature with a creative practice that links different energies together. We do not seek to find, reveal the various stages, methods and stakes which led the designers to push back the limits of the discipline, develop, as well as innovate, renew and create new materials, tools or uses through the production of these machines of transfer and transformation of energy in this chapter. We wish to make a state of affairs that will serve to initiate the reflection on this setting in shape- or property-changing objects.

Indeed, this prior knowledge can reveal how traditional systems of action and their cultural practices can inform and consolidate research regarding 4D printing. Like the tendency to focus on spectacular cases, anticipating a pre-foundational dogma, the possible exclusion of cultural knowledge and traditional practices can impoverish 4D printing development. It is thus a question here of re-examining the question of the links between external stabilized knowledge concerning what "moves" or what induces "movement" to consider possible ramifications. It is not a question of reconstructing the modes of thought, the socio-cultural and scientific markers, the impact on the social recognition of the designers and manufacturers of actuators, as well as the types of production and transmission that have led to the establishment of this approach within developed societies, but to examine whether this immense field of knowledge can serve as a medium for the applicative development of 4D printing.

An invention begins with the association of at least two existing techniques (Kaplan 2012) to lead, in the sense of Simondon (2001), to what he calls a "concretization": here, 3D printing (André 2017b) and active matter. Initially, it is classically a juxtaposition of knowledge, but in time one can imagine a mutualization, a more complete integration (which is only rarely envisaged).

Converging technologies (CTs) refer to the convergence towards a common goal of discoveries and techniques of fundamental science and technology: CTs are technologies and knowledge systems that complement each other in order to achieve a shared goal. Together or in isolation, [4D] technologies can contribute to this convergence. (Nordmann 2004)

Citton (2016) mentions hyper-objects in the field of complexity that can be applied to the 4D printing domain. He writes:

Hyper-objects are nowhere isolable like classical objects because their existence is inter-objective: their mode of existence is constituted by relations between the things we identify as objects (e.g. a glacier, a water level, a harvest, a thermometer, etc.). Whether we call them "systems", "attractors", "networks", "entanglements", it does not matter: they exist and operate as a web of relations conditioning the behavior of the objects on which our attention is focused. [...] Hyper-objects are viscous: they tend to stick to the beings whose existence is involved in theirs.

We will have to deal with it, like Captain Haddock in *Tintin...* if the theme keeps its promises. It is a thought of the adventure which,

by the moving and complex order of the knowledge that it gets, invites us to face the novelty and the unforeseen. This relationship of transformation, made of tinkering, of adaptation to the circumstances and to the unforeseen, of forgetting the pre-established models and of ingenuity obliges, in any case, invents a specific mode of transmission, which is not of the order of the teaching of an abstract knowledge but of the learning of an ingenious know-how. (Faucheux and Forest 2011)

It is far from this present and potential future, the objective of this chapter which only envisages examining the links between known actuators and their possible penetration into 4D space, with an essential concept of "printability" because it is necessary to accept a humiliating defeat (to varying degrees) in regard to additive manufacturing. It is also to examine other more autonomous sources of inspiration, and even to imagine others. It is by the yardstick of the mastery of these elements that we will know if this boundary object or this hyper-object that is the result of 3D printing (which belongs, whatever we think, to the old world) will introduce itself into our daily life in a punctual or radical way.

The only thing that counts is the process, for it is the process that lasts, not the goal, which is only an illusion of the traveler when he walks from peak to peak, as if the goal reached had a meaning. (Fourastié 1966)

But in relation to the theme of this chapter, the comments presented show a complementary partner, that of the sensor used to take information and which is also an energy interface like the actuator and the engine. So, if we combine these three elements, the scientific literature is particularly vast! Figure 1.1 illustrates the temporal evolution of the number of publications on the theme.



Figure 1.1. Evolution of the number of publications on the theme of "Actuators" (according to the CNRS database)

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Like most sciences and other scientific fields, the number of publications is increasing very rapidly with exponential evolutions. But, after having recalled that the 4D printing paradigm emerged around the year 2013, it seemed interesting to us to consider at the same time, in the same approach, "Actuators" and "4D Printing" themes to examine the existence of a particular "academic pressure" on this association. Figure 1.2, which represents this data in relation to that corresponding to the main theme "Actuators", illustrates the interest of the researchers on these associated themes in an obvious way.



Figure 1.2. Evolution of the number of publications on the themes of "Actuators" and "4D Printing" compared to the number of publications on the theme of "Actuators" (according to the CNRS database)

Obviously, even if this composite theme remains modest in number of publications, the subject is very quickly associated with the production of 4D objects (dotted line extrapolated in 2015). There is thus an almost consubstantial link between both fields (4D and actuators).

#### 1.2.2. Different types of actuator

Philosophy begins in wonder. And at the end, when philosophic thought has done its best, the wonder remains. There have been added, however, some grasp of the immensity of things, some purification of emotion by understanding. (Whitehead 1968)

By engineering definition, an adaptive structure is

a structure that can change its properties (mechanical, physical, electrical, etc.) or geometric form in response to changing environmental conditions. (Wagg et al. 2007)

In addition to the ability to move in multiple directions, some actuators have the ability to deform in any way. A concrete example comes from the bio-inspired actuator of a jellyfish's locomotion (Villanueva et al. 2010).

An actuator can be described as a mechatronic device that changes the mechanical state of a system by modulating an energy flow between the mechanical domain and another energy domain. Different types of actuators, according to Bréguet (1998), Vermes and Czigany (2020) and other more specific authors, are described in Table 1.1. To meet the specificities of 4D printing, the actuators, integrated or not, must satisfy the following: a resolution below a certain threshold, allowing a given stroke, depending on the application, a reasonable speed and a mechanical strength adapted to the objective, and the ability to be totally or partially integrable in the 4D device. Other performances may be of interest, such as active or passive control aspects, reproducibility, robustness, stability in a polluted environment and aging. A distinction is made between classical linear actuators (pneumatic, hydraulic and electric) (e.g. Joo and Sanders (2009)). Nonlinear aspects can come from several sources: large displacements (geometric nonlinearity), changes in contact status (contact nonlinearity) and nonlinear stress-strain relationships (material nonlinearity) (Lee 2012).

Nature	Comment
Hydraulic and pneumatic	In general, high energy density and very high stiffness
	Numerous on-off applications
Flexible fluidic	Application of a pressure on an anisotropic elastic structure
	Artificial muscles
	Origamis by elastomers
Electromechanical	High energy density linear or rotary engines
	High precision possible (step by step)
Electrostatic	Low power but can be used in parallel
	Dielectric elastomers (change in shape induced by electrical
	energy)
Deformation of materials	Ultrasonic actuators (piezoelectric vibration converted into motion), change in elasticity, change in electrical properties,
	etc.
	Inch-worm (displacement in a bore like a worm in its hole)
	Rotating inch-worm with bimetals
	Inertial actuators (a mass is moved away from another one

Deformation of materials (cont'd)	suddenly, then slowly brought closer)
	Stick and slip (one of the two masses is zero or negligible
	compared to the other)
	Electro-hydrostatic (avionics)
	Pyrotechnics (forcing by explosion of a membrane)
	Chemical reaction inducing a variation of volume or pressure
	Specific chemical absorption or adsorption
Modifications of 2D and 3D structures	Thermal expansion (bimetal and other amplifications)
	Structural reorganization; isomerization
	Electroactive polymers and electroactive ceramics, single or
	multilayer; resonances of piezoelectric elements
	Electrostriction, magnetostriction
	Shape memory alloys: reversible evolution between two
	crystalline states affecting the geometry
	Thermosensitive and photosensitive functional polymer
	actuators based on semi-crystalline polymers and/or liquid
	crystal polymers
Micro heat sources	Association of a heat-sensitive material with several heat
	sources
Integration of magnetic	Association of a magneto-sensitive material with several
micro-sources	magnetic sources
Modification of the bearing capacity	Drag and boundary layers: promote the laminar-turbulent
	transition of the boundary layer to delay separation and
	reduce recirculation zones; disorganization of sheared layers

#### Table 1.1. Different types of actuator (non-exhaustive list)

A liquid crystal is a state of matter that combines properties of an ordinary liquid and those of a crystallized solid. We express its state by the term "mesophase" or "mesomorphic state". Borshch et al. (2013) remind us that these are ordered structures (with an associated order parameter *S*). *S* goes from 0 for a completely absent structure to 1 for a crystal structure. The behavior of the system, at a macroscopic level, is given by a collective variable. The order parameter describes the behavior, that is the coordination of the various constituent elements of the organized system. If  $\theta$  is the angle between two elongated molecules, then:

$$S = \frac{3\cos^2(\theta) - 1}{2}$$

The effect of a stimulus (of the same type as those sought in 4D printing) can change the interaction potentials between the constituent elements of liquid crystals and thus *S*. Figure 1.3, from Ge (2019), shows the classical forms of liquid crystals.



Box 1.3. Liquid crystals

#### 1.2.3. Amplification

About the laws of nature: "To say this to a physicist is to say to a tiger in search of prey that all flesh is grass." (Weinberg 1997)

4D printing can reproduce physico-mechanical changes over time in a controlled manner, allowing the transition from static to dynamic. By combining the ability of 4D printing to create dynamic morphological changes under certain stimuli and the possibility of creating biocompatible micro- or nano-robots based on elastomeric materials, biomedical applications can be envisaged (Hann et al. 2020). However, among the current challenges and limitations of these devices, is the issue of deformation amplitude. As in any system where mechanical elements are involved, this question of the amplitudes of displacements is often raised. When these are considered too modest in relation to the objective, mechanics have already developed relays that allow the movements to be amplified. It is in this field that we will find, for example, the bilayer or constrained systems, presented in Figures 1.4 and 1.5, which could be used in 4D printing.



Figure 1.4. Bilayer

A bilayer, as its name indicates, is a device made of the juxtaposition of two cohesive blades of materials with different mechanical properties (e.g. different expansion coefficients). It is thanks to the different properties of these two materials that it will deform to achieve a mechanical action. Bilayer type elements allow for the generation of a bending moment or annular elements that will, using a shear mode, generate a displacement perpendicular to the element (Lambert et al. 2016). A 4D printing model is presented in Lee et al. (2019). For their part, Baran et al. (2017) propose multi-layer or multi-blade evolutionary systems.



Figure 1.5. Displacement amplification under stress

If  $\Delta l$  is the elongation, the height *h* reached is defined by  $h = \sqrt{2l\Delta l}$ , which corresponds to an amplified effect in spatial terms.

#### 1.2.4. Other modes of action from mechanics

The artist, taken in this highest sense, does not create only for the entertainment and instruction of his audience, he works solely and exclusively for the formation of his own existence. It is culture as Goethe understood it, as he opposed it to philosophy and science, it is this hard and rigorous task of knowledge that accompanied him all his life, forcing him to assimilate with a hunger never satisfied, all of the phenomena of life, and to metamorphose them to the true meaning of the word. (Broch 1985)

The discovery of new materials is an important part of our technological history. Manufacturers are increasingly using materials for high-tech sectors, as well as in more everyday areas; their objectives are to improve performance, lower the cost of manufacturing products and maintain or even improve reliability. The concept of "smart" materials and systems emerged in the early 1980s. They are capable of changing a characteristic such as shape, modulus, viscosity and transparency under stress (Elguesse 2015).

#### 1.2.4.1. Fluidic actuators

Before Koch, the bacillus had no real existence. (Latour 2020)

The first family of flexible fluidic actuators is based on the historical McKibben actuator, developed in the late 1950s. The basic principle of flexible fluidic actuators consists of a pressure applied on an anisotropic elastic structure. The basic configurations are shown in Figure 1.6.



Figure 1.6. Different types of fluidic actuator (see http://www.zpag.net/Tecnologie6s\_Indistrielles/Verin.htm)

A pneumatic actuator, as shown in this figure, consists of a cylinder containing a sliding piston with a fixed metal rod that transmits the mechanical energy generated by the pressure. For a double-acting cylinder, two compressed air supply ports are machined on either side of the piston. The commonly accepted advantages of these flexible fluidic actuators lie in high compliance (compliance is the opposite of stiffness), high efficiency of input pneumatic energy conversion, very low heating, low density and, most importantly, the absence of a magnetic part (Lambert et al. 2016). Since it involves the use of flexible materials, this type of actuator should advantageously be able to be used in 4D printing.

For Lambert et al. (2016), the characteristic elements to be considered are free elongation (without applied force), locking force (without displacement), and response time, developed stress (N/mm<sup>2</sup>) as a function of actuator volume (mm<sup>3</sup>), applied pressure (bar), dimensions, number of degrees of freedom, achieved performance, as well as indications on manufacturing choices (materials, processes), etc. (Huber et al. 1997; De Volder and Reynaerts 2010; Gorissen et al. 2011).

#### 1.2.4.2. Shape memory alloy (SMA)-based actuators

Time is the modification of space. (Fourastié 1966)

Shape memory alloys (SMAs) are exploited for their thermomechanical properties, such as their hyperelasticity in the austenite state, plastic behavior in the martensite state and the shape memory effect controlled by temperature change (Simoneau 2013; Yuan et al. 2020). In mechanical applications, SMAs have the advantages of being clean and developing high stresses for low weights (Lambert et al. 2016). The principle of these active systems is observed in large families of alloys including copper (CuAlZn), nickel-titanium (NiTiNb) or AuCd (discovered in 1932) based alloys. These materials can evolve between two crystalline states affecting their geometry: the austenite state at high temperature (cubic mesh) and the martensite state at low temperature, the latter has a significantly flattened mesh, as shown in Figure 1.7.

The transition from both crystalline states occurs through a threshold temperature. According to Lambert et al. (2016) and Boni and Royer-Carfagni (2021), the stress experienced can be a uni-axial elongation when the SMA is a tensioned wire or bar (Mavroidis 2002). But, for a larger stroke, it is possible to use an SMA material in the form of a coil spring (Velazquez et al. 2008). When this spring is stretched or compressed, the material works primarily in torsion and the internal stresses are of the shear type (Sheng and Desai 2015). The beam network form can be useful when looking for an optimal trade-off between force and output stroke (Abadie et al. 2009).



Figure 1.7. Shape memory alloys

Figure 1.8, from Lambert et al. (2016), represents the evolution of the martensite phase in the material as a function of temperature and for a given stress. This diagram accounts for the transformation hysteresis Y. The different transition temperatures are defined by considering that As and Af are respectively the thresholds of start and end of transformation from the martensite state to the austenite state, and Ms and Mf are those of the reverse transformation (Cianchetti 2013).



**Figure 1.8.** Evolution of the martensite rate in the material as a function of temperature and for a given stress

#### 1.2.4.3. Translation - rotation

Some ideas spread so well that, in different versions, they invade whole populations in a lasting way. Culture is made, in the first place, of these contagious ideas. It is also made up of all the behaviors [...] and all the products [...] whose presence in the shared environment of a human group allows ideas to spread. (Sperber 1995)

Ren et al. (2021) have taken up a system that transforms a stimulus-induced translation (electrochemical in their case) with a rotation (arm joint type). Figure 1.9 illustrates a working principle of such a biomimetic system.



Figure 1.9. Actuator transforming translation into rotation

Relative to what is proposed by work from homogeneous processes, the nanofiber-containing "muscles" provide an isometric stress of 10.8 MPa (about

30 times what is achieved with skeletal muscle). Using the accumulated isometric stress, the wire muscles achieve a high contraction rate of 36.3% s<sup>-1</sup>. The wire muscles can be grouped together to lift large masses and grasp real objects.

#### 1.2.4.4. Rotary engines

Most engines that output mechanical power in the form of torque (rotary output) use crank-crank (translation-rotation) foundations with sequentially excited actuators.

#### 1.2.5. Remarkable properties

Philosophy [...] must refuse the freedom indulged in by specialized thoughts that reject or exclude what is incompatible with their presupposition and that, if need be, boast of "scandalizing common sense". (Stengers 2020)

#### 1.2.5.1. General considerations

Time is the moving image of still eternity. (Morel 2020)

All happiness is essentially poetry, and poetry means action; one hardly likes a happiness that falls to you, one wants to have done it. (Alain 1985)

Recall that an actuator is an operating component of a functional system that transforms the energy supplied to it into a physical phenomenon that provides work, transfers energy or modifies the behavior or state of a system. An associated effector can finalize the work by producing the expected effect. Among the properties of the actuators are a number of important elements, such as the response time or the elastic modulus.

- Response time is a measure of performance defined as the time between the end of a request to an actuator and the beginning of the response (see Figure 1.10). Latency is also expressed as a creep (or delay) function.

– Young's modulus, the modulus of elasticity or tensile modulus is the constant that relates the tensile stress and the onset of deformation of an isotropic elastic material. The law of elasticity is Hooke's law of the form:  $\sigma = \varepsilon E$ , where  $\sigma$  is the stress (in units of pressure), *E* is Young's modulus (in units of pressure) and  $\varepsilon$  is the relative (dimensionless) elongation. The international unit is therefore the pascal (Pa). However, because of the high values this modulus takes, it is usually given in mega-pascals (MPa).



Figure 1.10. Concept of response time

– Other criteria related to the material are presented in Figure 1.11 (Troyano et al. 2021) and others can be searched for, such as a nominal power (Watt), an action frequency ( $s^{-1}$ ) and a torque to be transmitted (Newton.m or N.m). Additional elements resulting from their use in robotics concern aspects of impedances and mechanical admittances. These are stiffness or elasticity, mass or inertia, viscous friction or damping, dry or Coulomb friction and kinematic constraints (revolute, ball joint, etc.). The interested reader can find information on these aspects in Fauteux (2009). Indeed, the main elements such as the response time and the main mechanical properties will suffice to start a comparison between "traditional" actuators and 4D devices.



**Figure 1.11.** Material characterization (*E*:Young's modulus; *v*: Poisson's ratio; *G*: Shear modulus; *K*: volume modulus of compressibility)

COMMENT ON FIGURE 1.11.— The three moduli E, G and K allow for the characterization of the elastic behavior of an elastic material. These constants result from the existing relationship between a stress and the induced deformation:

 $-\sigma = \varepsilon E$ , where  $\sigma$  is the stress (in units of pressure), E is the Young's modulus (in units of pressure) and  $\varepsilon$  is the relative (dimensionless) elongation.

 $-\tau$ :  $\gamma G$ , where  $\tau$  is the shear stress (in units of pressure), G is the shear or Coulomb modulus (in units of pressure) and  $\gamma$  is the shear angle (dimensionless).

 $-p = -K \frac{\Delta V}{V}$ , where p is the hydrostatic pressure exerted on the volume (unit of pressure), K is the volume modulus of compressibility (unit of pressure) and  $\frac{\Delta V}{V}$  is the relative change in volume (dimensionless).

If the deformation is elastic, there is partial compensation of this increase in volume by lateral contraction of the specimen ( $\Delta y$  and  $\Delta z$ ) in directions perpendicular to the tension. The relative deformation in the y and z directions is then written (for a homogeneous material):

$$-\varepsilon_y = \frac{\Delta y}{y_0}$$
 with  $v$ , Poisson coefficient defined by  $v = \frac{\varepsilon_y}{\varepsilon_x} = \frac{\varepsilon_z}{\varepsilon_x}$   
 $-E, G, K \text{ and } v \text{ are related to each other by the relation } E = \frac{9KG}{(G+3K)} = 2G(1+v)$ 

- Temporal aspects must be taken into account with materials whose shape must evolve following a stress. Figure 1.12 (from Billon (2007)) represents the case of the deformation of a linear polymer after a first transition (not shown in the figure) from a completely disordered state, in thermodynamic equilibrium prior to the stimulation, to a change of order that favors the flow.



Figure 1.12. Orientation of polymeric chains in the direction of flow

Each "linear" chain has a free volume in which it can move by propagating local conformational changes. But, depending on the constraint exerted, the system that constitutes each polymer can be forced to adapt, for example by reducing the number of its Cis bonds relative to the Trans ones, thus prompting the transport phenomenon. These different movements have their own time scale. In polymers,

the elasticity energy represents the energy storage resulting from the rotation around the bonds, the variation of the bond angles and finally the variation of the equilibrium distances between atoms. The elasticity energy is a contribution from intramolecular bonds (within a macromolecule) rather than from intermolecular bonds.

The macroscopic properties of polymers depend on their local architecture: monomers, nature of the chains and their rotational abilities, length of the chains, etc. (flexibility of the skeleton, capacity of cooperativity, physico-chemical and physical interactions, entanglements and disentanglements). The formulation of the polymer material involves additives, such as fillers or plasticizers (inter-chain interaction), nucleating agents (semi-crystalline organization (rate and size)) and blends. All of these interactions lead to different relaxation times that can be considered in global 4D printing.

Thus, for a viscoelastic fluid, there are two modes of deformation characterizing the viscoelastic behavior: (1) stress relaxation which consists of imposing a deformation step on the material and observing the evolution of the stress as a function of time and (2) creep which consists of imposing a stress step and then observing the evolution of the deformation as a function of time. Under viscoelastic conditions, these different parameters become functions of time and space.

An elastomer is made up of long molecular chains assembled, at rest in "balls". These chains are linked together by cross-links, entanglements or polar bonds with mineral fillers. They form a 3D network. The mechanical properties of elastomers depend primarily on the length of the chains between the nodes of the network. The shorter this length (dense network), the more rigid the elastomer. When the material is extended or twisted, the molecular chains unfold and adopt a linear order which is achieved at the expense of the secondary bonds, the primary bonds remaining effective. The cessation of the extension-torsion stresses thus allows the molecular chains to return to their passive, folded and random configuration (elastic deformation), provided that the stresses have not broken the primary bonds, in which case the deformation becomes irreversible (passage into the plastic domain) (Mercier et al. 1999)

The deformation of a homogeneous polymer results from local changes of more or less cooperative conformations, which, from one to another, lead to global deformations.

These local conformational changes are constrained by interaction energy terms (van der Waals or others) between unbound atoms (not bound by a covalent bond) and by torsion potentials on the valence cones for bound atoms. The elementary processes of chain deformation. (Billon and Bouvard 2015)

The deformations of thermally activated macromolecules are time-dependent, dependent on the nature of the material (and its charges) and thermally dependent on their associated energies. "It also depends on the interchain organization through the conformational changes it favors and/or annihilates" (Billon and Bouvard 2015). Figure 1.13, from Pic (2009), recalls the effects of temperature on a polymer.



Figure 1.13. Effects of temperature on the elasticity of a macromolecule

These different parameters are defined under conditions where there is proportionality between the stress and the transformation. For too high stresses, the behavior may no longer be linear, with irreversibilities and performance losses (degraded processes and failures) (see Zhou et al. (2021)).

Evanescent memory: in viscoelastic materials, the deformations determined at the reference time depend on the entire history of mechanical stresses already experienced. According to Pic (2009), this type of material called "evanescent memory material" keeps the deformations undergone during its manufacture and during its use in its memory, but the effect of these constraints fades in the course of time (Lagache 2006).

Box 1.4. Conditions on materials

#### 1.2.5.2. Mechanical performance

He who finds what he is looking for generally does a good schoolboy's job; thinking about what he wants, he often overlooks the signs, sometimes minimal, that bring something other than the object of his predictions. The true researcher must know how to pay attention to the signs that will reveal the existence of a phenomenon that he does not expect. (Bourcier and Van Andel 2017)

Figure 1.14 (according to Liu et al. 2021a) depicting an Ashby diagram of compressive-density strength shows the range of possibilities depending on the inorganic materials used with very large dynamics.



**Figure 1.14.** Ashby diagram of inorganic materials (ceramics). 1: Hollow alumina nano-lattice; 2: SiOC micro-lattice; 3: SiOC honeycomb; 4: AIO/C<sub>23</sub> nano-lattice; 5: AIO/C<sub>23</sub> honeycomb polymer; 6: SiOC matrix and carbon nanotube matrix; 7: SiOC matrix and honeycomb carbon nanotube matrix; 8: AIO/C<sub>23</sub> nano-lattice polymer; 9: SiC foam

With respect to organic materials, Figure 1.15 shows that we are generally a long way away from the conditions achieved and illustrated in Figure 1.14. Most of the research efforts in the field of 4D printing are focused on smart (often soft) actuators with sensing and shape-changing capacities. They need to be observed through Key Performance Indicators (KPIs) and associated mechanical properties. Following the classification of actuators proposed by Zupan et al. (2002) and more specifically

their derived characteristics, some attributes can be given special attention by designers and engineers, such as actuation stress, actuation deformation, actuation modulus, work output, work capacity, actuator efficiency and stroke work coefficient, in order to define the industrial specifications. In the present analysis, to measure and monitor the performance of 4D printed actuators with active materials, these attributes can also be derived and expressed in terms of material-properties (Karothu et al. 2020), such as Young's modulus, response time under stimulation, force generated and actuation efficiency (i.e. the ratio of mechanical work produced to energy absorbed during a complete cycle). Thus, Figure 1.15(a) represents the KPIs ranges for the main categories of active materials as used in 4D printing (Shape Memory Polymer – SMP, Shape Memory Alloy – SMA, Piezoelectric Polymer – PZT polymer, hydrogel, Ionic Polymer-Metal composites – IPMC and Liquid Crystal Elastomer – LCE).

Many studies have focused on characterizing active materials to provide relevant evidence for material selection (Huber et al. 1997; Song et al. 2016; Boyraz et al. 2018; Kim et al. 2019a; Decroly et al. 2020; Halabi et al. 2021). However, large disparities can be reported between the calculated/measured indicators which make it difficult for non-experts to use them. This can be explained in particular by the different measurement scales highlighting different temporalities, the customization and distribution of materials, the size of actuators, as well as the different material shaping processes studied over the last two decades. However, it should be noted that the current KPIs (shown in Figure 1.15(a)) are still low when compared to the performance of existing actuators (e.g. hydraulic and pneumatic) as used in industry. Most 3D printed active materials have notable weaknesses in terms of mechanical stiffness, actuation forces, actuation/recovery responses (e.g. thermoactive materials with large heating/cooling times due to Fourier's law of thermal conduction), changeability and even functional degradation of the material over multiple cycles.



**Figure 1.15(a).** KPIs of the main categories of active materials used in 4D printing: a) Young's modulus, b) response time once stimulated and c) forces generated and actuation efficiencies (from Demoly et al. (2021))



Figure 1.15(b). Comparison of Young's moduli according to Adam et al. (2021)



**Figure 1.15(c).** Case of conventional polymers according to Lecomte-Beckers (2010). 1: Carbon fiber reinforced composites; 2: glass fiber reinforced polymers; 3: polypropylene; 4: polyethylene; 5 and 6: conventional polymers; 7: PTFE; 8: Butyl rubber and Isoprene; 9: rubber reinforced with carbon black; 10: silicone-based elastomers; 11: high-density rigid polymeric foams; 12: medium-density flexible polymeric foams; 13: low-density rigid polymeric foams; 14: low-density flexible polymeric foams

Figure 1.16 of Hofman (2021) shows other highly differentiating criteria between Young's modulus and coefficients of thermal expansion. The same is true for the items shown in Figure 1.17 corresponding to mass torque relative to actuator speed (Babaei et al. 2021).



Figure 1.17. Relationship between torque density and speed
Considering the considerable dynamics to be taken into account, going from an established field for which rules exist to a very recent field still "boiling", we can imagine the difficulty that a designer may have to find "good" materials adapted to their problem.

# 1.2.5.3. Actuator selection

The simulacrum is the identical copy of an original that never existed. (Baudrillard 1978)

Almost obviously, apart from the mechanical aspects (Young's modulus) and response time, other elements of choice can be considered. Depending on the use, the nature of the actuation (bending, elongation, deformation, etc.), the power in relation to the mass, the robustness, the durability, the frequency of use, etc., can be taken into account. In the same way (if only to prepare the next paragraph), the selection of materials can be a selection criterion: single-material system, multi-material, biocompatibility, biodegradability, etc.

Among some examples explaining this selection issue, Forget (2018) presents criteria for a pneumatic actuation with advantages: intrinsically compliant system; very dynamic movements; good control of the position/force of the joint, as well as its impedance/admittance; simplicity and lightness (good weight/power ratio); reduced price; pump relocation, etc. The disadvantages are the following: high pressures involved (risk of explosion); accurate modeling of the system difficult due to highly nonlinear behavior according to Alvarez Palacio (2020); friction losses; noise; each force/speed requirement requires different actuator sizing; pump sized for the "worst case", that is for the most powerful actuator; agonist-antagonist architecture imposing control of two variables per degree of freedom, making control more expensive in terms of computational power, etc.

In the case of an electric cable actuator also proposed by Forget (2018), using an electric engine and a ball screw/nut linkage used to drive a cable, the advantages would be as follows: well-documented and mastered electric engine control; reversible/transparent actuator, providing good perception of external forces from motor current, as well as good efficiency; use of a cable, providing flexibility that softens contact with the environment and providing an interface for estimating external forces (by measuring cable deflection); and the footprint is similar to a human muscle (exoskeletons and biomimetic systems). The disadvantages proposed by this author would be flexibility introduced by the cable, complicating the control of the system and reducing its bandwidth, and the presence of complex physical phenomena to identify and control.

For all that, these criteria need to be refined. We have chosen a single example from Abba (2012), related to the possible selection of engines for robotics.

Figure 1.18 shows that this selection based on efficiency and weight criteria is quite difficult to consider if it must interfere with the others presented above.



Figure 1.18. Relation between efficiency and weight of a motorization in robotics (knees)

# 1.3. Actuators and 4D printing

Power establishes an asymmetrical relationship between the one or those who command and the one or those who obey. If it is not reduced to domination, the power relationship can engender a consent to obedience, but also the possibility of disobedience, the impatience of freedom, the horizon of insubordination. (Spector 2018)

NAP (2019b) reminds us that "soft matter" (Modes and Warner 2016; Nagel 2017; Smalyukh 2018) can be described as a material with low or zero elastic modulus and that this parameter can govern the ability of a material to resist deformation. This domain concerns polymers, colloids, foams, emulsions, etc. It corresponds to a mesoscopic scale (intermediate between the atomic and millimeter scales). The energies taken into consideration are adapted to easily accessible energy inputs, energies involved in 4D printing. NAP (2019b) also writes:

Soft matter is often highly dissipative, disordered, entropy-dominated, far from equilibrium, and characterized by strong nonlinear and slow

dynamic responses. Although these properties have been extensively studied over the past decades, our understanding of them is still incomplete.

In fact, independently of 4D printing, this field is growing rapidly and it seems wise to follow the current developments to consider profitable applications for the manufacturing field.

## 1.3.1. General framework

To produce something new, one must certainly "doubt...". But also to be able to "doubt...", that is to say, to accept, at least temporarily, a badly supported, fragile and provisional knowledge. Beyond doubt, it is the risk of thought that we must assume. (Lévy-Leblond 2020)

In order to extend the application domain of the actuators presented in the previous section to 4D printing and then to partially unknown, dynamic or anthropogenic environments, the same or better, increased physical interaction capabilities are required. In this context, meeting the safety, robustness and versatility requirements of new 4D devices is a challenge.

If we had limited this book to homogeneous 4D printing, this volume of published information could have been greatly reduced by the addition of an essential discriminating criterion, that of "printability" (this will be mentioned later in this chapter). On the other hand, with heterogeneous 4D printing strategies, it is possible to introduce elements into a 4D device that are not necessarily realized by 3D printing (but with elements that are effectively mechanically linked together to constitute the object). For all that, we did not wish to widen the presentation too much by essentially limiting ourselves to the energetic material-stimulation relationships. Table 1.2 presents some works that may be of interest for combining additive manufacturing and actuators.

4D printing allows for the addition of active and reactive features in response to various external stimuli. The SMPs, which by selective absorption of light can produce a mechanical action, will often be mentioned (Hingoran et al. 2019; Zhang et al. 2019; Jeong et al. 2020). If, in principle, 4D printing can enable the fabrication of objects with complex geometries, the fine control of deformations with predefined responses is not yet optimal. We will later show that this difficulty is not unique to SMPs.

Energy	Subject and commentary	References	3D
Mechanical	Self-assembly of silk Pneumatic effects in elastomers	Elletro 2015 Di Natali et al. 2020; Lee and Bhattacharya 2021	N ?
Mechanical	Actuators made of SMA wires combined with a return element Valves	Morellon 2010	?
	Energy storage	Pal et al. 2020; Qian et al. 2020; Sweet et al. 2020	
Mechanical	Haptic devices and electroactive actuators	Biet 2007; Kaneto 2016	N
Acoustic	Polymers	Adam et al. 2021	Y
Mechanical engineering	Transfer in metamaterials; origami	Mukhopadhyay et al. 2020; Novelino et al. 2020; Vangelatos et al. 2020	Y
Fluid mechanics	Reduction of drag in flows	Onal et al. 2017; Ott 2020	?
Piezoelectricity	Microsystems	Cueff 2011	Ν
Electro-activity; electrothermics	Polymers; hydrogels	Choi et al. 2020 Garces and Ayranci 2020	? Y
Electric field	Ionic polymers	Carrico et al. 2019; Narayan et al. 2021; Tyagi et al. 2021	?
Various	Micro-actuators	Bréguet 1998; Vermes and Czigany 2020	?
Various	Electro-activity and flexible fluidics	Lambert et al. 2016	Y
Magnetic	Deformation under magnetic field; nanoparticles	Whip 2016; Song et al. 2020; Zhang et al. 2021e	?
Electromechanical	MEMS	Li et al. 2020b	
Magneto- mechanical	Nanoparticles	Glock et al. 2015; Golovin et al. 2015	N
	Elastomers	Sindersberger et al. 2018; Zhang et al. 2019; Mirvakili et al. 2020	?

Di-electricity	Active polymers	Chortos et al. 2019	N
Chemistry	Chemical reaction producing a change in phase, volume, etc.	Suzumori et al. 2013	?
Chemistry; color change	Color change associated with the chemical environment	Lu et al. 2020; Nie et al. 2021	?
Chemistry: pH	Oriented polymers	Sarikaya et al. 2021	Y
Heat, photons	Heat and/or light sensitive materials; SMAs	Ge 2019; Du et al. 2020; Lee et al. 2020	?
Thermal effect	Deformation of organized liquid crystal structures	Zhang et al. 2020b	Y
Thermomechanics	Bilames	Cao et al. 2010; Baran et al. 2017	?
Photochemical effect	Molecular rotor	Bhatti et al. 2020; Kuenstler et al. 2020; Lan et al. 2020; Li et al. 2020a; Lu et al. 2020	?
Microwave	Liquid crystals	Wang et al. 2020d	?
Moisture; water	Absorption/desorption inducing volume change	Le Duigou 2017; Dai et al. 2020; Li et al. 2020a; Zhao et al. 2020; Piotrowska et al. 2021	?
Biology	Bio-printing	Nie and Takeuchi 2020	Y
Electricity	Dielectric muscles	Haghiashtiani et al. 2018; Qiu et al. 2019; Ghosh and Basu 2021	?
Autonomy	Autonomous chemical reactor	Hu et al. 2020	Y

# **Table 1.2.** Examples of energy transformers (Y: publications that consider the use of 3D printing; N: do not consider it; ? do not mention it)

What this table shows is that in a top-down approach going from the main market (actuators) to the 4D aspect, the scientific field shows a weak interest in this last area, which remains quite confidential. Nevertheless, we find the classical effects presented in section 1.1 with originalities associated with active materials with stimulations presented in Figure 1.19 (Rastogi et al. 2019).



Figure 1.19. Main stimulation modes in 4D printing

For reasons associated with the resolution and/or the limited number of voxels, most authors have turned to the use of a transformable material to manufacture a polymer sensitive to a stimulus (heat, solvent, humidity, pH, light, mechanical, electromagnetic field, etc.) via an additive manufacturing process. Therefore, for the most part, there are classic additive manufacturing processes with so-called intelligent, active, informed or programmable materials. The underlying idea is to start from a given geometry so that it becomes a shape corresponding to an application through energy input. By playing on the spatial and amplitude distribution of a form of this energy (of these energies), there would also be the possibility of having an adaptive system like an actuator, a flexible robot, a structure evolving in time and perhaps in its functionalities (André 2017; Frenzel et al. 2017; Zarek et al. 2017; Kuang et al. 2019; Lehn 2002).

# 1.3.2. Specificities linked to the manufacturing process

There are still dead generations that make prudish books [...], books about the day, about hobbies, about travel. But not books that embed themselves in thought and speak about the black mourning of all life, the commonplace of all thought. (Duras 1993)

In the field of 4D printing, the main criterion is in the material itself, with processes that must "follow" to achieve the desired objective. We have recalled throughout this book one of the interests of additive manufacturing, which is that deposits of different materials located in the space constituting the 4D object are enabled. However, as shown in Figure 1.20, if at least two materials are used, several active associations can be retained (Du Plessis et al. 2021). Obviously, these structures have an effect on the behavior of 4D objects.



Figure 1.20. Possible arrangements of multi-materials

Natural structures are generally based on a limited number of repetitive structural design elements (Naleway et al. 2015), such as fibrous, helical, gradient, layered, tubular, cellular, sutured and tile. These structural design units are present in nature and can be used in 4D design, in connection with the search for the best performance (e.g. carbon fibers, already used for material reinforcement (Ahmed et al. 2020)). Helical structures improve fracture toughness by inducing curved crack paths. Cellular structures, tube, suture and tile designs can also improve performance (Du Plessis et al. 2021). What this figure basically shows is that the active component can be embedded in a supporting material, from a molecular homogeneity step to associations between objects of different natures and functionalities. One serves to support the action carried out by the other element. The "passive" component (effector) must have specific criteria in terms of weight, mechanical properties, etc. This is where metamaterials come into their own.

# 1.3.2.1. Metamaterials

Design is a problem-solving activity without a fixed, directly applicable procedure. (Malhotra et al. 1980)

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Metamaterials have origins dating back to the early 19th century (Veselago and Narimanov 2006). They are fabricated structures that can mimic the responses of known materials or possess new ones, some of which do not exist in nature (Engheta and Ziolkowski 2006). In particular, these organizations may have properties that are weakly related to the characteristics of their chemical constituents, but rather to the shapes given to these metamaterials (Liu and Zhang 2011). According to Saunders (2020), the origins of these organizations stem from the theoretical work of Veselago (1968) and later Pendry (2000) and Smith et al. (2000). Shelby et al. (2001) are said to have been the first to create such arrangements and demonstrate an interesting property such as a negative refractive index.

Furthermore, metamaterials have been developed and used for multiple purposes in many fields: morphing of aircraft structures, internal prostheses of implants, comfort of shoes and linings of helmets and cribs to reduce the risk of brain trauma in case of impact, etc. These new classes of metamaterials can be manufactured by additive manufacturing. At present, these materials are not stimulable (but this is being considered and is sometimes practiced) and can be used as effectors (Bristol University 2021).

Buckling, once considered the "ultimate" design failure, has been exploited in recent years to develop mechanical metamaterials with advanced features to create 3D structures with exploitable properties in 4D printing (Janbaz et al. 2019). The structural complexity of metamaterials is in principle unlimited, but most designs include periodic architectures that lead to materials with spatially homogeneous characteristics (Cui et al. 2010, 2014; Buriak et al. 2016; Coulais et al. 2016; Krödel et al. 2017; Jayashankar et al. 2019).

# 1.3.2.1.1. Metamaterials and their properties (based primarily on Kadic et al. 2019)

The provincial cities, quietly sitting on their heritage, are not often associated with audacity and willingly leave the effervescence of feverish creations and polemics to the capitals. This fixed representation of a cultural life where everything that moves and sometimes disturbs is necessarily concentrated in a single extreme of free creativity, corresponding to the place in which powers and wealth are concentrated, is a very French fact. (Francfort 2007)

- Metamaterials are rationally designed composites, made up of custom-made building blocks, with average properties superior to those of their constituents.

- Metamaterials can be designed by computers (to avoid tedious trial and error procedures and excessive experimentation).

- Additive manufacturing can advantageously be used to physically create complex shapes.

- Although many 1D and 2D model architectures have been considered due to their ease of manufacture and reduced design complexity, the potential of the metamaterial concept is fully open to 3D microstructures and nanostructures.

- For Salari-Sharif et al. (2018), it is possible to combine the stiffness of metals or ceramics with the damping capabilities of rubber (damping properties).

- In electromagnetism and optics, they have remarkable properties (diamagnetism, paramagnetism up to optical frequencies, impedance matching, negative refractive indices, maximum electromagnetic chirality, perfect optical absorption, non-reciprocal propagation of electromagnetic waves without static magnetic fields, etc.).

– In acoustics and mechanics, they have full flexibility in elastic parameter matching, chiral mechanical behavior, sign reversal of effective static compressibility, negative dynamic mass density, non-reciprocal sound propagation, perfect broadband sound absorption at the fundamental limit imposed by causality and highly nonlinear, multi-stable and programmable properties of linear elastic constituents (see Dubourg (2015)).

- Thermal conductance is highly anisotropic, such as sign reversal of absolute mobility and Hall coefficient, highly anisotropic Hall tensors, giant magnetoresistances and thermoelectric power factors that are increased by an order of magnitude.

- 3D printers could enable thousands of different metamaterial properties from a small number of input materials.

Therefore, it is not only mechanical and actuation properties that can be of interest to researchers invested in 4D printing, since many functionalities can be envisaged. The emerging areas presented above constitute a roadmap for future research on metamaterials. In particular, extending the study of the mechanical behavior of metamaterials to the nonlinear range of deformations and to flexible metamaterials should receive attention in the next few years, as well as developing the manufacture of metamaterials containing active materials, such as those used in homogeneous 4D printing. Many possible designs of nonlinear flexible meta-materials yet to be discovered could present opportunities for novel applications using, in particular, additive manufacturing.

Figures 1.21 (Ion et al. 2016, 2018a, 2018b; Novak et al. 2016) and 1.22 (Kamru et al. 2020) illustrate on these bases, the possibility of realizing flexible structures that can be set in motion using external mechanical actuators. These structures can

be made using additive manufacturing. The example in Figure 1.22 corresponds to a microstructure prevalent in the literature: originally realized in 1982 by Gibson and Ashby, according to Azoti (2012), in a 2D model, this simple shape is based on the hexagonal geometry of a conventional honeycomb structure. By defining an angular parameter, it is possible to reverse the orientation of the edges or struts of the geometry and thus define a "reentrant" geometry. Thus, contrary to the homogeneous 4D printing, we use wire structures or foams (and not a material in volume) with external actuations (let us recall however that the materials must be flexible to consider the deformation, but can only be inert).



B)

**Figure 1.21.** Deformation of heterogeneous structures under the action of an external force (the red disks correspond to the attachment points between the different structures: A: planar deformation; B: spatial deformation from a planar structure)



Figure 1.22. Reentrant structure

Other deformations can be considered, as shown in Figures 1.23 (Novak et al. 2016) and 1.24 (Kolken and Zadpoor 2017). The renewed interest in metamaterials is, in fact, associated with recent advances in additive manufacturing that allow for the manufacture of objects with complex nano/micro-architecture, potentially covering different spatial scales. Due to their rationally designed architecture, mechanical metamaterials exhibit unusual properties at the macro scale. These properties could be utilized for the development of materials with advanced functionalities, with applications in soft robotics (Jin et al. 2018), biomedicine, soft electronics, acoustic masking, etc. (Kolken and Zadpoor 2017).





**Figure 1.23.** Other examples of deformation: A): Multi-material system with a flexible joint; B): Flexible relationship between blocks; C): Bi-material (active, inert) system; D): Deformation of a chiral structure



**Figure 1.24.** Structural changes observed in a microporous PTFE (Poly-Tetra-Fluoro-Ethylene) foam subjected to tensile loading in the horizontal direction: A) Almost fully densified state; B) Tension in the fibrils causes displacement of the particles; C) Rotation of the particles into a disk shape (side view): D) Fully expanded state

However, it should be remembered that such a structurally complex implementation can lead to geometric frustration (whereby local constraints cannot be satisfied everywhere), which can prevent coherent operation and consequently hinder the desired functionality. Regardless of this aspect, the literature abounds with 2D and 3D lattice structures. The interested reader can advantageously consult the following references: Bodaghi et al. (2017b), Martinez et al. (2017, 2018), Liu et al. (2018a, 2018c), Alison et al. (2019), Kadic et al. (2020).

The shapes given to these metamaterials can be very complex (which is an area of excellence of additive manufacturing). In general, the mechanical properties of the metamaterials are programmed and fixed during the design and fabrication of the architecture, and do not change with changing environmental conditions or application requirements. However, according to Li et al. (2020c), while materials are of great research interest due to unusual mechanical responses and a wide range of deployment possibilities in plane and space, the cellular structure and nature of the constituent elements of materials that deform in bending or rotation generally still have relatively low stiffness. For these authors, this situation limits their applications when high stiffness, strength, hardness and energy absorption are desired simultaneously. They propose the use of auxiliary reticular structures as reinforcements, combining them with almost incompressible flexible materials to produce high-performance composites. However, in reality, as shown in Figure 1.25, since one does not seek to have a material suitable for additive and active manufacturing simultaneously, it is possible to cover a very wide range of densities and mechanical properties (Zadpoor 2016). But the debate remains open.



**Figure 1.25.** Different ultralight and ultrarigid metamaterials using various types of repeating unit cells (see also Zheng et al. (2014)) – 1: graphene-containing elastomers; 2: ultralight metal micro-meshes; 3: hollow alumina-based lattices; 4: foams containing carbon nano-tubes; 5: acrylic foam; 6: solid acrylic; 7: solid alumina; 8: elastomers; 9: solid composite materials; 10: metals; 11: "hard" metal alloys; 12: industrial ceramics

However, additive manufacturing technologies always play on an optimization between a precision or a resolution on the one hand and an object realization time on the other hand. On this observation, the definition of different metamaterial scales is limited. Yildizdag et al. (2019) recall that multiscale architectures are designed to achieve the desired macroscopic behavior by activating interactions between different length scales and coupling different physical mechanisms. Research perspectives are discussed by these authors in order to determine the most efficient methodology needed to design new metamaterials that optimally meet a functional specification. However, the number of spatial scales exploited is still modest.

Several examples of metamaterial structures can be found in the literature (Masters and Evans 1996; Schumacher et al. 2015; Jiang and Wang 2016; Boydston et al. 2018; Chen et al. 2018; Golaszewski et al. 2018; Ion et al. 2018a, 2018b; Jackson et al. 2018; Jiang et al. 2018; Mirzaali et al. 2018; Alison et al. 2019; Beli et al. 2019; Bodaghi and Liao 2019; Kadic et al. 2019; Lei et al. 2019; Yuan et al. 2019; Vangelatos et al. 2019; Zhao et al. 2019; Jing et al. 2020; Manna et al. 2020; Naify et al. 2020; Rafsanjani et al. 2020; Tao et al. 2020). Based on the

examples presented in this list of publications, there is, as can be seen, a solid foundation for moving forward.

# 1.3.2.1.2. Auxiliary materials (meta-materials)

The error committed by the scientist is the product of the haste of his desire, which decides to undertake an action when his understanding has not yet sufficiently weighed the parties involved. (Willmann 2010)

Lattice structures have specific geometric characteristics reported to have a negative Poisson's ratio (auxetic metamaterial), that is stretching induces expansion in the transverse direction, as illustrated in Figure 1.26 (Lim 2015; Saxena et al. 2016; Lei et al. 2019). Materials and structures with a negative Poisson's ratio obviously exhibit counterintuitive behavior (Ren et al. 2018).



Figure 1.26. Differences in the behavior of conventional and auxetic materials

The Poisson's ratio v is one of two mechanical properties used in the description of elastic and isotropic behavior of solid materials following the small strain approximation (Goldstein et al. 2009). In the case of tensile/compression mechanical tests, v is defined by the negative ratio of transverse strain to axial strain. Its effect is of fundamental importance in the behavior of materials so that a variation in its value leads to significant changes in mechanical performance. In the case of isotropic materials, the value of v is defined according to thermodynamic restrictions such as -1 < v < 0.5. Most materials have a positive v coefficient. Therefore, these materials deform by a contracted or thinned cross-section when they are longitudinally stressed.

By analyzing the deformation mechanism associated with auxetic behavior, it is possible to show that the macroscopic behavior results from a structure-related effect due to a network of architectural or periodic reinforcements. Foams remain the preferred domain of auxetic materials (Lakes 1987). Thermoplastic (polyester, urethane), thermosetting (rubber, silicone) and metallic (copper) foams have been reported by Friis et al. (1988) as auxetic. It is found that, in all cases, the value of the Poisson's ratio varies with strain, and values approaching the -1 limit for isotropic materials have been reported in polymeric and metallic foams (Choi and Lakes 1992). As a result of this work, it has been possible to design stiffer materials for a wider range of applications. For example, in 1989, Caddock and Evans discovered a form of microporous PTFE (Poly-Tetra-Fluoro-Ethylene) foam exhibiting strongly anisotropic and strain-dependent behavior, such that a Poisson's ratio value as low as -12 was observed in one direction.

## 1.3.2.1.3. Voronoi diagrams and 4D printing

The forms are passive insofar as they represent the actuality; they become active when they are organized in relation to the bottom, thus bringing the previous virtualities to the actuality. (Simondon 1997)

In mathematics, a Voronoi diagram or Dirichlet tessellation is a tessellation of the plane into cells (adjacent regions) from a discrete set of points called "seeds". Each cell encloses a single seed, and forms the set of points in the plane that are closer to this seed than to any other. The cell represents, as it were, the seed's "zone of influence". As shown in Figure 1.27, to generate a Voronoi diagram, only certain medians are used and not others: sites that are too far away are not taken into consideration; then the useful segments form a triangulation or partition of the plane known as the Delaunay diagram. In mechanics, a Delaunay triangulation structure is considered to minimize the squares of the areas of the triangles. Counting the outer boundaries, each site is inside a convex polygon. All points in this polygon are closer to the inner point than to all other points. This is an irregular honeycomb pattern. Voronoi diagrams can use more complex distance rules (power Voronoi network) (Delaunay 1934; Preparata and Shamos 1985; Okabe et al. 1992; Aurenhammer and Klein 2000; Baert 2003; Wormser 2008; Boissonnat 2010; Mitchell et al. 2018).

One design route is to spatially distribute materials with different properties within lattice structures to achieve desired mechanical properties. Advanced multi-material 3D printing techniques allow for the design and fabrication of multi-material cellular structures for which the elastic modulus and Poisson's ratio can be independently tuned in different directions (anisotropic) (Mirzaali et al. 2018). Seed distribution for additive manufacturing can be achieved by random Monte Carlo firing methods that allow for spatial coverage. In addition, it is possible

to define the oriented probabilities of the variable density type (case A in Figure 1.28) or preferred orientations in Figure 1.28 (case B).



Plot of the perpendicular bisectors of the segments formed by the close pairs. Each joins one of the vertices of the polygon (Voronoï points)

Figure 1.27. Basics of Voronoi diagram construction



**Figure 1.28.** Seed space coverage produced by random firing: A: variable density; B: variable orientation

Lee et al. (2018) used Voronoi diagrams for tessellation applied to additive manufacturing. But, to the authors' knowledge, it is mainly Sylvain Lefebvre's team at INRIA (Martinez et al. 2018; Ray et al. 2018; Bedel et al. 2021) that has applied this type of scanning to the realization of deformable objects. Figure 1.29 represents some examples from this team, illustrating the possibility of creating structures,

optimized relative to the desired effect, inhomogeneous of metamaterials with specific spatial functionalities.



Figure 1.29. Non-periodic metamaterials with optimized spatial functionality

Figure 1.30, taken from Martinez et al. (2017) from the same team, using the hand as a mechanical actuator, illustrates the potential of auxetic foams shaped from Voronoi diagrams using 3D printing.



Figure 1.30. "4D heterogeneous" application of auxetic systems

Publications envision the use of additive manufacturing technologies to create auxetic structures (Schaffner et al. 2018; Bodaghi and Liao 2019; Kanu et al. 2019; Sadeqi et al. 2019; Rafiee et al. 2020). Recall that recent advances in multi-material 3D and 4D printing should be able to be further exploited to extend the design space beyond the now conventional geometries of metamaterials. It is then conceivable to combine different materials and various additive manufacturing technologies to create multifunctional objects that better meet application needs (with, for example, multi-scale and multi-material structures; see Liu et al. (2021b)). Apart from the possibility of incorporating multiple materials. In their publication, they use seven scale changes, ranging from tens of nanometers to tens of centimeters. As a result,

these micro- and nano-array materials can support more than 16,000 times their own weight and exhibit a quasi-linear scaling between the structure's density and its Young's modulus. In addition, the aspect ratio is increased.

Zadpoor (2016) reports that fatigue behavior studies of mechanical metamaterials have focused on static, quasi-static or specific types of dynamic behaviors of mechanical metamaterials. In the real situation of structural use of mechanical metamaterials, fatigue behavior studies should be conducted.

# Box 1.5. Functional fatigue

# 1.3.2.1.4. Modeling of deformations

Withdraw into yourself and look. And if you do not find yourself beautiful yet, act as does the creator of a statue that is to be made beautiful: he cuts away here, he smoothes there, he makes this line lighter, this other purer, until a lovely face has grown upon his work. So do you also: cut away all that is excessive, straighten all that is crooked, bring light to all that is overcast, labour to make all one glow of beauty and never cease chiselling your statue (Plotinus 2017)

Each structure and material combination leads to unique structural properties such as stiffness, Poisson's ratio and overall elasticity. The ideal input or bypass force distribution mechanism could be suggested for the design of guards using different shapes, thicknesses and auxiliary materials to reduce the risk of damage (Yang et al. 2018). Related to homogeneous 4D printing, there is the possibility of modeling displacements as a function of actuation amplitude and directivity (Weeger et al. 2019; Liu et al. 2020c; Weeger 2021).



**Figure 1.31.** Possible degradation of metamaterials under mechanical stress

However, as it has already been specified, these deformations must remain within a linear behavior, in any case reversible. In the case of metal-organic material systems, Troyano et al. (2021) remind us (see Figure 1.31) of the possible irreversible fate of metamaterial structures, depending on the nature (and the amplitude) of the solicitation.

# 1.3.2.1.5. Some applications of metamaterials

Increasingly, metamaterials are associated in the literature with additive manufacturing and, in particular, with 4D printing. Table 1.3 gathers some recent references concerning these particular structures and applications.



Figure 1.32. Reconfigurable connector for medical applications



Figure 1.33. Relationships between the form of metamaterials and mechanical performance (Metamaterials – MM1: density 0.001; MM2: density 0.01; MM3: density 0.1)

Nature of the deformation	Comments	References
Reconfigurable connector	Stents (see Figure 1.32) Smart-robot (5 axes) Smart-sensor	Van Manen et al. 2021 Kim et al. 2019b; Zhang et al. 2021b Zhu et al. 2020
Active structure	Origami; kirigami; electromagnetic propulsion Ferromagnetic structures Electroactive polymers	Mousanezhad et al. 2017; Ning et al. 2018; Zhao and Kim 2019; Jin et al. 2020; Novelino et al. 2020; Chung et al. 2021; Guo et al. 2021; Li and Yin 2021; Ma et al. 2021; Zhang et al. 2021a Kim et al. 2018 Tyagi et al. 2021
Relationship between form and performance	Young's modulus (see Figure 1.33) Hierarchical architecture Compression-torsion Tensegrity Different configurations	Jenett 2020 Coulais et al. 2016; Ronellenfitsch et al. 2019; Surjadi et al. 2019 Wang and Liu 2020; Zhong et al. 2020; Goswami et al. 2021 Yin et al. 2020 Meng et al. 2020; Yang et al. 2020
Porous structures	Graphene-based structures	Zhang et al. 2018
Tunable performance	Shape memory polymers	Tao et al. 2020
Deformation and shape	Damping of acoustic waves, vibrations	Rocklin et al. 2017; Liu et al. 2020b; Wang et al. 2020; Xue et al. 2020
Pneumatics	Auxiliary structures	Zunker and Gonella 2021
Materials	Different organic materials Organometallic	Kuang et al. 2020 Hu et al. 2017b
Viscoelasticity	Viscoelastic materials	Dykstra et al. 2019

# Table 1.3. Metamaterials and applications

# 1.3.2.2. Notion of "printability"

I am convinced that half of what separates successful entrepreneurs from the non-successul ones is pure perseverance. (Jobs 2011)

Additive manufacturing enables complex parts that are true to their design to be manufactured in a single step. It aims to eliminate the need to assemble multiple components, train a new workforce or set up new equipment, while minimizing manufacturing time, wasted materials and energy. The available "library" of printable materials and the range of properties represented is growing, but needs to be expanded - for example, to include materials that have a variety of electrical and thermal conductivities, or that swell when they absorb a solvent, etc. (Overvelde 2019). But, even if at the beginning, the materials that are going to be used for the manufacture of a 4D object are compatible with each other, after their deposition or placement that is associated with an energy input, there is nothing to say that their behavior maintains cohesion between voxels of different origins - how then do we perform "tenacious" solidarizations (hence the interest or risk of working with heterogeneous 4D printing)? In the past, it was possible to use mechanical means such as rivets, weaving, felting, interlocking, glues and cements. We can push "open doors" with resins of the same family for 3D work using stereolithography (Katri et al. 2020), but there are very serious issues associated with this notion: problems related to the manufacturing process, the thicknesses involved with their internal tensions (Altenhofen et al. 2018), and also to the physico-chemical and mechanical interactions between the voxels. However, the consultation of craftsmen and industrialists in the trade reveals constants in the bonding operations: preparation of the surfaces to be assembled to promote the closeness between elements to be connected, using the largest possible contact surface (Wang et al. 2021b), playing as much as possible on the wettability of the adhesive so that it can spread in the area to be bonded, exploitation of the surface condition to promote adhesion (see Figure 1.34).

This technology has a weakness related to the bonding of the layers formed during the printing process. Gojzewski et al. (2020) defined a mapping of the contact Young's modulus across the layered structure with sub-micrometer resolution. They observed local depressions with values up to 30% of the maximum stiffness at the interface between consecutively deposited layers, indicating a local depletion of the macromolecular cross-linking density. Even with a single base material, differences in mechanical coefficients may exist, which may (or may not) favor voxel-to-voxel bonding and, consequently, printability...

# Box 1.6. Stereolithography

For Freund et al. (2019), the factors influencing the interface strength between different materials are not well understood, so this complexity is rarely utilized, which hinders innovation. A systematic approach to identifying and quantifying the adhesion phenomena that influence interface strength is a necessary step if one wishes to move beyond proofs of concept. For this reason, these authors propose experiments adapted to certain lines of action, such as roller peel tests and peel strength of rigid and flexible materials. Polarity, mechanical locking and surface roughness associated with the design of 4D structures have a great influence on the interface strength of multi-material parts manufactured by extrusion (FFF technique). In this example, criteria that mainly affect the interface strength are deduced and design recommendations for creating functional parts with mismatched material combinations can be formulated.

In the case of bonding, direct adhesion between materials occurs rarely, only when the materials to be joined have completely smooth and clean surfaces that allow intimate contact between the materials at a very small distance. It is therefore not possible between two materials with rough surfaces since the contact area is greatly reduced. The use of a glue (or adhesive), applied between the two materials, will therefore allow the creation of a film that will ensure an intermediate and intimate contact between the two surfaces to be joined (see Figure 1.34 inspired by ISPA (2015)). Amorphous polymers present a structure without any particular arrangement. The chains of molecules are intermingled in a disordered manner. They are characterized by a glass transition temperature Tg. Above the Tg, the energy is high enough for the molecular chains to become disordered. In this case, the cooling conditions have no influence on the mechanical characteristics of the material. The processing can therefore be done more easily compared to semi-crystalline polymers. Amorphous polymers have weaker mechanical properties than semi-crystalline ones. The reinforcing fibers, of which the most used in the traditional materials are glass, carbon and aramid fibers, have a role of optimizing the properties, especially mechanical, of the material (but also of surface). The choice of the reinforcement is made according to the application and the required characteristics.

On a practical level, positivism makes the programming of science assured, but horizontal: when an idea is truly new, one usually only has the most confused lights as to the means by which one may even conceive of verification by experiment. (D'Espagnat 2015)



# Figure 1.34. Roughness and mechanical adhesion

Alternatively, flexible composite 4D actuators can be manufactured by embedding SMA wires or fibers in flexible polymer matrices (Akbaria et al. 2021). This manufacturing method can be complex as it involves multi-material printing. Several solutions can be considered: deposition of a soft elastomeric layer on a more rigid layer of SMP, homogeneous or not (see Rodrigue et al. (2017)). Depending on the temperature, deformations of greater or lesser amplitudes can be achieved as preferred axes of action. If nonlinear finite element models can be used to predict the deformation of these actuators, nothing is said about the behavior in time of this type of hybrid actuator. Normally, a good adhesion between the passive and active elements is expected. However, if the expansion coefficients are very different (bimetal effect), nothing says that the two materials will keep a sufficient cohesion over time. This situation is even more critical when the two materials do not adhere to each other. It is in these conditions that the notion of "printability" can be introduced.

In the heterogeneous manufacturing processes discussed, we can have voxels of "reasonable" size that can be connected to each other by mechanical means (Demoly and André 2021b). However, this situation of classical (improved) mechanics no longer makes sense with voxels of sub-millimeter size and it is then necessary to examine whether the usable multi-materials are compatible with each other for fabrication and whether they will have a high number of actuations. Indeed, tensions between passive and active elements (which are responsible for the desired deformation) can lead to micro-fractures during use, which can be amplified during the loading/unloading time and cause the 4D performance to drop or even disappear.

Thus, this notion, which is not found in the work corresponding to "simple" actuators, seems here to be a primordial criterion in the definition of a good 4D actuator. One of the possible reasons is the small number of "multi-material" 3D machines (see Thomas (2021)).

Antoine Le Duigou (2019) reports on the advantage of using natural fibers such as those from flax, which he likens to nano-structured composites with very interesting mechanical properties for 4D applications. Flax fibers are composed of cellulose, hemicellulose and lignin for the most part. After suitable treatments, the moisture-active properties can be measured and lead to spectacular 4D demonstrations. Other natural fibers have similar properties (coconut, banana, wood, etc.).

# Box 1.7. Natural fibers

### 1.3.2.2.1. Multi-material 3D machines

When we are not in the grip of a theory, we are all capable of juggling multiple contextual, practical and semantic resources, according to the

demands of situations, and we are not seized with dismay at the idea that these situations may matter to others in a different mode. On the contrary, it interests us, it enters our ruminations and activates our imaginations. (Stengers 2020)

Rafiee et al. (2020) point out that a limited number of 3D machines are suitable for using multiple materials (see their tables of commercially available multimaterial 3D printers and their specifications for polymers and biomaterials). They present known results in a synthesis linking materials and processes without introducing adhesion criteria between voxels from different materials. However, at the end of their paper, they conclude with the following general points:

- Topological optimization can be used to propose new complex geometries that are easier to manufacture by 3D printing than by any other process. Although considerable progress has been made with multi-material 3D and 4D printing, the potential has not yet been fully explored.

- The mechanical performance of parts made by multi-material additive manufacturing is generally better than that of parts printed by single-material printing. The formation of voids between successive layers of printed parts can affect their mechanical performance due to a decrease in interfacial bonding between the printed layers. Different mechanical behavior under vertical tension or compression compared to that of the horizontal direction is another common challenge of 3D printing processes.

- The efficiency of the production of parts can be an area of research to explore with the mastery of post-processing methods adapted to the use of several materials.

- Most commercial multi-material 3D printers create parts from voxels of the same size. However, obviously, for a certain functionality and, in particular, mechanical strength (Young's modulus, elasticity), different sizes may be required.

- Future processes could tailor the microstructure properties of a 4D printed part to create more complex geometric transformations by strategically controlling the density and directionality of the deposited stimulus-sensitive materials. They can also improve the inter-material bonding of heterogeneous smart compositions, and even ignore active or non-active material properties.

In these senses, the development of multi-material 4D printing can be the object of decisive integrative research to accelerate the growth of the smart materials sector dedicated to this field. It is not just about serendipitous encounters or process modification (Muguruza et al. 2017; Toursangsaraki 2018).

# 1.3.2.2.2. Elements of "printability"

Better to be Socrates dissatisfied than a fool satisfied. (Mill 1988)

If you judge a fish on its ability to climb a tree, it will spend its life thinking it is stupid. (Vervisch 2020)

A literature review on this topic was conducted: out of more than 170,000 references on additive manufacturing, between 30 and 50 articles mention this problem without one or more generic paths being formulated. Obviously, the youth of 4D printing technology has not allowed this type of hindsight to generate all the hypotheses, validate them and propose practical solutions to designers (see, however, Mao and Anand (2018) and Ni et al. (2020)). Table 1.4 gathers the collected elements.

Elements of "printability"	Materials	References
Inter-diffusion between different voxels	Hydrogels	He et al. 2016
Shear effects	General framework	Peng et al. 2021
Polymer blend	Long and short chains	Zhang et al. 2021c
Linear polymers	Long chains	Zhang et al. 2020a
Surface condition	Interfaces between voxels	Freund et al. 2019
Bi-materials (improved electrical properties)	CNT and graphene polybutylene terephthalate (PBT) nano-composites	Gnanasekaran et al. 2017
Control of phase transitions	Liquid crystal elastomers (LCEs)	Saed et al. 2019
Temperature effects	Cracking of stressed polymers	Baek et al. 2021
3D CFD (Computational Fluid Dynamics) modeling	Liquid-liquid system	Urhal et al. 2020
Shear strength and thixotropic properties	Elastomer with magnetic field dependent rheology	Bastola et al. 2018
Damping effect	Foams	Montgomery et al. 2021
Viscosity and surface tension	UV resin loaded with metallic nanoparticles	Saleh et al. 2017
Collage	Binding between voxels	Frascio et al. 2020
The process itself	Metals and non-metals	Wang and Liu 2014
"Sandwich" structures	Various extruded polymers	Moises et al. 2020
Intermediate binder	Coated fibers	Bodkhe et al. 2020; Liu and Wei 2021
Fiber orientation Surface tension	Fiber filled polymers	Ma et al. 2019; Mantelli et al. 2019

Prioritization of deposits Time between layers	Polymers Cement	Teoh et al. 2017; Scalet 2020; Weng et al. 2021
Extrusion mode	Polymers	Baca and Ahmad 2020
Debinding and sintering	Metals, ceramics and polymers	Gonzalez-Gutierrez et al. 2018
Subtractive-additive coupling	Robotics	Mostafavi et al. 2019
Self-organization	General framework	Demoly and André 2021
Wave propagation	Crystals in a polymer matrix	Zhang and Wu 2021
Accession	Hydrogels	Li et al. 2021
Interfacial cohesion	Filaments	Lopes et al. 2018; Fernandez et al. 2019
Cohesion	Tensile tests	Kluczyński et al. 2021

Table 1.4. Some elements collected on the "printability" aspect

Professionals are suspicious of amateurs. They are wrong. The latter get it wrong with less method. (Lemire 2015)

The exceptional abilities of geckos to adhere to various surfaces are largely credited to the large actual contact areas of the fibrillar and hierarchical structures of their feet. These special features regulate the essential structural compliance for each attachment and thus provide robust but reversible adhesions (Wang et al. 2013). Figure 1.35 (taken from Wang et al. (2019b)) illustrates the principle of such a system. It can be a torsional (Kang et al. 2017) or shear (Wang et al. 2013) detachment process.



Figure 1.35. Mechanical adhesion and de-adhesion

Other methods have also been tested to change the adhesion values: temperature in Xu et al. (2018) and a magnetic field in Li et al. (2018). These biomimetic materials can therefore play an adhesion role and by different treatments see their bonding power transformed for the disassembly of the 3D object's constituent elements, an advantage relative to the case of using polymer adhesives and the maintenance of the adhesion material on the building blocks.

The adhesion of an interface could be activated and deactivated by the sole interaction between hydrophilic functional groups, introduced by surface oxidation, and the native  $AlO_{23}$  oxide on the aluminum surface (Khongtong and Ferguson 2004). This interaction also acts as a driving force (enthalpy) to stretch the chains to increase the entropy of the system, with functional groups being pulled away from the elastomer/aluminum interface, resulting in the weakening of adhesion. The basic science is developed in the paper, but practical solutions are not reached. The search for a follow-up to this work was not successful.

In the absence of cyclic processes, according to Zhang et al. (2020a), the use of inelastic dissipators often achieves strong adhesion (see Yang et al. (2019)). Good fatigue-resistant adhesion is proposed by Zhang et al. (2020a) using a particularly simple type of elastic heatsink consisting of long-chain polymers. Each polymer chain remains elastic before it breaks. When a single covalent bond in the chain breaks, the elastic energy stored in the entire chain dissipates, amplifying the adhesion energy by the number of links in the chain. They obtain adhesion and fatigue threshold energies that can reach 1,400 J/m<sup>2</sup> and 300 J/m<sup>2</sup>, respectively. This fatigue threshold would be linearly proportional to the square root of the chain length, in agreement with the Lake and Thomas (1967) model. This fatigue resistant design could be advantageously extended to the association of voxels of different origins as long as a suitable 3D process is available.

Freund et al. (2019) proposed a set of criteria to consider when thinking about the adhesion of voxels to each other. This set, building on the work of Awaja et al. (2009) and Mohamed et al. (2015) is the subject of Figure 1.36 (see also Nguyen et al. (2018), Alief et al. (2019) and Ando et al. (2021)). These are elements that will need to be examined on a case-by-case basis in combination with others yet to be considered.

On the other hand, Zhao et al. (2021) used hydrogels, composed of noncovalently cross-linking 3D polymer networks and water. The fatigue behavior of these conventional materials in 4D printing was studied. With poly(vinyl alcohol)/poly(acrylic acid) (PVA/PAA) gels with weak hydrogen bonding (called weak gels) and PVA/PAA-LiCl gels with strong hydrogen bonding (strong gels), they show that for fatigue damage of uncut samples, the weak and strong gels are amplified by fatigue for small deformations (rearrangement of the hydrogen bonds induced by stretching), but with fatigue damage effects in case of large deformations by the sliding of the polymeric chains, one over the other. For the fatigue failure of pre-cut samples, the weak and strong gels do not present any measurable fatigue threshold for these authors, but the strong gel can attenuate the growth of cracks. These results reveal that intrinsically weak non-covalent bonds cannot effectively increase the fatigue strength of physical hydrogels. More generally, Zhou et al. (2021) have practically investigated the effect of stresses on materials on fracture mechanisms. Zhang et al. (2015c) and Bai et al. (2019) had, on the other hand, studied the same phenomena of elastomeric materials splitting under stress.



**Figure 1.36.** Fishbone diagram of factors of influence concerning the interface strength of multi-material voxels

Regardless of these more or less predictable, but disparate aspects, Melanie (2020) reports that optimization involves a printability analysis corresponding to the evaluation of the part before printing. The AMFG enterprise reportedly includes a feature in its software package that ensures that a part to be manufactured is truly suitable for 3D printing, whether in terms of stability, shape or resilience.

# 1.3.2.2.3. 4D printing and functionality

Throughout this book, we have been talking about the form and functionality given to a material that must be thought of as a complex but structured system. However, a further analysis of the literature shows very few references where the active material (apart from materials dissolved in printable media) is concerned with anything other than changes in form. However, Table 1.5 gives some examples of changes in functionality.

Change in functionality	References
Electromagnetic properties	Wu et al. 2020
Piezoelectricity	Nkomo 2018; Tang et al. 2021
Nanotechnology, color	Zhang et al. 2021
4D and power supply	Teng et al. 2021
Temperature sensitive color	Chen et al. 2021a

## Table 1.5. 4D printing and feature changes

## 1.3.2.2.4. A return to aspects of self-organization

The truly good and wise man will bear all kinds of fortune in a seemly way, and will always act in the noblest manner that the circumstances allow. (Aristotle 2012)

An intentional artifact (related to the work of the engineer and/or designer) can be considered as a means of linking an "internal" environment, the substance, functioning and organization of the object itself, and an external environment, the surroundings in which it is implemented. If the two environments are in a compatible situation, the artifact meets the specifications. As Simon (2004) pointed out in another framework, knowledge of an artifact such as a machine:

has an advantage over knowledge of nature, because it is based on valid prior foundations, the purposes of which will be deviated from with a certain amount of intentional novelty to give the projects intelligibility and openings to society.

This can be true for 4D printing of single-materials, we have just shown that it is not quite the same for associations of materials.

The term "self-organization" refers to the spontaneous/stimulated formation of a dynamic nature leading to forms of organization resulting from a set of homogeneous or non-interacting units. Self-organization results from large-scale structures based on microscopic chaotic motions and small-scale random fluctuations (Lebedev 2012). After placing energy in the right place at the right time in a given environment, the initial system under stress organizes itself to achieve a form, if possible sought by the experimenter (Moreno 2004). From a thermodynamic point of view, this "shaping" supposes that the transport of matter and energy between the system undergoing transformation is functionally constrained in the absence of external energy input. As the object changes, there is an appearance of causality (universal laws of physics), which can be represented by a kind of selection between several possible alternatives (Campbell 1974).

Thermodynamics describes the change of a system from an initial state to a final equilibrium state, without taking into account the transitions. In fact, it is the path and "distance" of the system from its final equilibrium that generates and maintains the structural complexity found in living organisms and sought to be replicated with "non-living" matter (Monteiro 2018). Dissipative structures are processes in which a fluctuation stabilizes to form a dynamic macroscopic configuration enabled by the flow of energy through the system under construction. These macroscopic structures require a continuous supply of energy of an amplitude adapted to its coherence:

The term "self"-organization implies a spontaneous creation of order or organization rather than an agential identity capable of determining or constituting itself in a sense that implies functional plasticity. (Alvaro 2004)

It is basically to respect this comment by Bruter (Le Moigne 1994), who wrote: "The events shape with the time the object, it is above all to have pierced the secrets of its history, of the lineage of which it is, at the same time, the result and a projection".

Even if, by some extraordinary chance, you were to stumble upon her, how would you know it was her, since you have never known her? (Socrates 2021)

Order is only a random special case of disorder. (Schiffter 2019)

In Demoly and André (2021), we considered the possibilities of local manipulation of matter by different forms of energy, with this matter being activatable with the ultimate goal of realizing 3D shapes meeting industrial requirements. This field of "morphogenetic engineering", according to Doursat et al. (2013):

explores the design, implementation, and control (directly, through programming, and/or indirectly, through learning or evolution) of complex system agents capable of autonomously and reproducibly giving rise to large, heterogeneous architectures that will support a desired set of functionalities, without depending on central planning or external control.

Indeed, it is necessary to control the parameters presented in Figure 1.37 in order to achieve an operating efficiency that enables the precise realization of a 3D object.



Figure 1.37. Influence of self-organization parameters on additive manufacturing

For those who study the intrusion of temporal and functional aspects in additive manufacturing, systems have been fully determined for a long time. But, as "self-organized" phenomena, they can become sensitive, far from equilibrium, to factors considered negligible near equilibrium. It is the intrinsic activity of the increasingly complex system, with its increasingly nonlinear behavior, that would determine how it is possible to describe its relationship with the environment, and that would generate the type of intelligibility that would be relevant for understanding its possible histories. In this chapter, it was not only a field of application with its constraints, but also a theoretical field to be attacked in order to solve the equation finality/means to achieve it in a robust way. However, the inability to master the inverse problems associated with the aspects of morphogenesis presented in this reflection could (should?) have closed the debate. There is no realistic solution today for the realization of precise 3D objects by self-organization, starting from a certain disorder to reach a desired order!

Self-organization brought the idea that a stable and reproducible form can be the product of a balance of local rules and randomness. These are collective phenomena, since they are irreducible to one or more elements taken in isolation. (Petitgirard 2014)

On the other hand, in the situation considered here, we are trying to explain losses in performance that could originate from an order-disorder transition following a stimulation (which, obviously, is in line with the evolution of the entropy of a system). What we recalled is that in the case of systems with several degrees of freedom, not all attractors are equal: some aim for greater stability, others for greater growth potential. However, the dynamics involved do not necessarily lead to the most stable state: the system that follows the fastest path may reach local areas of stability. It is then possible that a certain reversibility of the processes is at work: the actuator does its job! But, after a certain number of actuations, this path will generally end with a global minimum of potential, and not with the local minimum (see Figure 1.38).



Figure 1.38. Considered energy degradation processes

When some change occurs in nature, the quantity of action necessary for this change is the smallest possible. (Moreau de Maupertuis, cited in Lemire (2015))

One way to get the system out of a local minimum is to add a degree of indeterminacy to the dynamics, that is to give the system the possibility to make transitions to states other than the one that is the most "adapted" locally. It is at this level that the concept of "printability" takes on its full importance by preventing this degradation. This can be seen as preventing the injection of "noise" or random disturbances into the system, allowing new trajectories. In physics, this is usually the effect of external disturbances (e.g. vibrations or shocks in the system) or intrinsic indeterminacy (e.g. thermal or quantum fluctuations, or simply unknown factors that have not been incorporated into the state description).

The deeper the "valley" representing free energy, the harder it will be for a disturbance to move a system out of that valley. Therefore, noise will generally move the system out of the shallower valleys and into the deeper valleys (a form of determinism). However, a system subjected to noise will never really settle into a local or global minimum, because no matter what energy level it reaches, it can always be perturbed with the risk of reaching more energetic metastable states. The actuator will be unstable and probably of modest interest!

By considering polymers of different chain lengths (as found, for example, in natural rubber), Gros (2016) considers both the thermodynamics and the inhomogeneous entangled characters of the polymeric network. Polymeric chains orient and crystallize as the system deforms under stimulation (in this case mechanical or thermal). Crystallization under stress would occur from successive states of matter whose characteristics and threshold extensions of crystallization and melting depend on the initial chain length distribution. A scenario for the evolution of the chain network organization, with varying degrees of cross-linking, involves the critical and entanglement molar masses and the stress rate. Qualitatively, it is possible to go from an unstressed state to a deformed state, but the return to the initial situation is never total (hence the fatigue problems).

These broadly outlined elements can serve as an explanatory basis for undesired phenomena. But, for the practitioner, it is necessary to go further than this global explanation which can make people understand, but not solve problems which are likely to be critical in the near future.

# 1.3.2.2.5. Fatigue and return to equilibrium of high entropy materials: aging

Top-down logic is not made to enrich knowledge and adapt it, but on the contrary to tighten and reduce it. Once the premises have been established, there is always one solution and one solution only, and this solution is imposed in the name of one principle and one principle only: consistency. At the end of the decision-making chain, the executors are then considered as a burden: they impede the limpid formal beauty of the process put in place. (Crozier and Tillette 2000)

In the spirit of what has been presented above, it is for example possible to deposit fibers of different materials included in oriented polymers in a shear field, as shown in Figure 1.39. The order parameter of these materials can then approach unity. The classical stimulation is the use of moisture to evolve the fibers in their medium (see Wang et al. (2018, 2020), Le Duigou et al. (2019, 2020a, 2020b), Zou et al. (2020), Armstrong et al. (2020), Cohades and Michaud (2020), Balla et al. (2020), Bernard et al. (2021) and Cera et al. (2021)). But, after several round trips, we can imagine that the fibers will slowly return to a more thermodynamically stable configuration, thus with an order parameter approaching zero. This phenomenon is probably associated with the creation of fractures/cracks in the support material... (Liu and Moran 2021). Internal shear occurs when the fibers slide over each other during deformation. To maximize strength and avoid failure through this creep process, the longest possible fibers with the highest realistic load density would probably be appropriate (NAP 2019a). Thus, 4D properties are likely a function of the number of cycles they are cycled through, without much citation of this in the literature, which is still in its infancy.



**Figure 1.39.** Degradation of the 4D properties of fibers induced by the degradation of the S order parameter during use

Zorzetto and Ruffoni (2019) remind us that helical fibers can form building blocks (already used by nature). These materials arranged in multiple layers lead to biological structures with good and durable mechanical properties. With synthetic structures combining fibers and fibrils, they show that the fracture strength can be improved if fibrils are oriented perpendicular to the applied load.

# 1.3.2.2.6. A hybrid "printable" 4D printing

The numerical discretization of all movements in individualization makes it possible to sum, process, calculate, and model them by producing categorical attractors. (Stiegler 2004)

Sertoglu (2021) reports a recent process of depositing in a given shape a layer of a filler, possibly consisting of fibers, with a high filler density on a flat surface. In a second step, a liquid resin is deposited on the entire object under construction. What is not mentioned by this author is that this material has good activation properties by energy stimuli. A layer of charge is then deposited, etc. The difference with what is found in the scientific literature is that we "work backwards", the charged areas constitute passive structures (because the charges present prevent any local deformation, while accepting global deformations), the areas without charges constitute the active areas. Moreover, it is possible to have free zones without elastomeric material, loaded or not. The principle of this 4D construction is shown in Figure 1.40.



Figure 1.40. 4D "reverse" hybrid with passive loads

# 1.3.2.2.7. A study path

Moore et al. (2020) proposed a stereolithography method to realize assemblies made of different materials. This technology, among others, can be exploited to realize composite prints allowing studies of aging, either under mechanical stimulation or under specific stimulation associated with at least one of the materials. This practical work has interest for real applications.

# 1.4. Stimulations of matter

Culture concerns objects and is a phenomenon of the world; leisure concerns people and is a phenomenon of life [...]. Culture is threatened when all objects and things in the world, whether produced by the present or by the past, are treated as pure functions of the vital process of society, as if they were only there to satisfy some need. (Arendt 1989)

We have tried to progress in this chapter from classical actuators to effectors, leaving behind the central question of energy-matter interactions which must manage the temporal evolutions of 4D objects in terms of form or functionality. We must be consistent with the title of the chapter: "Getting things moving"! However, these physical, physico-chemical and/or chemical interactions have already been approached, without being studied in depth. They will be only partially explored, so vast is this domain that they could constitute a whole library.

Box 1.8. Stimuli-materials interactions
4D printing involves the use of "smart" materials with possible changes in form or functionality via additive manufacturing processes. Stimulus-sensitive materials, stimuli and characteristic times required for the transformation to occur are key research areas (Manikandan et al. 2021). This chapter focuses on the materials and methods used to make adaptive structures. We will not revisit the most common processes used, which are stereolithography (SLA), fused filament fabrication (FFF), material jetting, selective laser melting and direct ink writing (DIW) (see for example Bodaghi et al. (2017a)).

One of the first goals is to try to get out of this opposition between the morphology of an artificial structure desired by the designer, designed digitally in advance and manufactured from one step to the next (current case of additive manufacturing) and a system that can develop over time thanks to the interactions between the different parts made up of informed materials of the structure, which we know generally leads to forms that are weakly structured enough to allow for evolutions. But is it possible? However, engrossed in the certainties of their scientific paradigms, a number of researchers are constantly ploughing the same furrow. What the authors wanted to do in this article was to approach the way of thinking like a detective or explorer by examining (no doubt with great naivety) whether it is possible to discover exceptions to the accepted laws, to reach the limit of the accepted theories (Lévy-Leblond 2020).

# 1.4.1. Programmable matter

Perhaps the simplest way to symbolize the permanent form of our life is as follows: it is determined by the pressure exerted on us by objects and circumstances, by nature and society, as well as by the reactions of our freedom, which abolish this pressure or allow themselves to be violated by it, fight it or avoid it [...]. This is perhaps the major complication of our life: what limits its spontaneity and oppresses its free upward aspiration is at the same time the indispensable condition for this aspiration to action to reach a visible expression, a creative activity of form. (Simmel 1990)

Molecular motors whose movement is induced by external energy inputs constitute an archetype of what can be programmable matter. As early as the 1950s, Richard Feynman predicted the development of molecular nano-machines, such as those made by Jean-Pierre Sauvage in 1983 when he invented a chemical method allowing two molecules in the form of rings to be intertwined, thus forming a chain named catenane (Collin et al. 2001; CNRS 2016). Through electrical or light stimulation it has been widely shown that it is possible to drive the rotation of a molecule in a controlled manner, similar to an engine. We can therefore consider

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these examples as already representative of what could be the programmable matter, which has been the object of interest of researchers for only a few years, without it being possible to identify a trend (linear or exponential) in the growing number of publications associated with the keyword "*programmable matter*" (see Figure 1.41). The total number of publications provided by the CNRS database is in the order of 1,000 (which should be compared to the 150,000–200,000 concerning additive manufacturing).



**Figure 1.41.** Evolution of the number of publications under the theme of "programmable matter" (according to the CNRS database)

By focusing on 4D printing, it seemed necessary to quickly examine this research field and then leave it to get closer to the concept of a connection between voxels, which effectively implies the notion of additivity. It is therefore necessary to have at least two molecules (or two voxels) in order to start talking about additive manufacturing. Indeed, the concept of programmable matter allows us to consider applications such as the remodeling of an object or to test different possibilities of actuation. According to Catry et al. (2020), this assembly can be realized in principle easily according to the needs of the designers. They propose several solutions based on self-reconfigurable modular robotics. A POC has been realized for a catome with links between voxels of electrostatic origin. However, if the shape is achieved, the mechanical strength of the realized structures can still be improved (see also Di Luna et al. (2017), Yue et al. (2018), Zakharov and Pismen (2019),

Almethen et al. (2019), Yigit et al. (2019), Liu et al. (2020), Gastineau et al. (2020), Piranda and Bourgeois (2020), Qi et al. (2020), Dolev et al. (2020) and Yang et al. (2021b).

The basic principle is to associate elements, as shown in Figure 1.42 from Catry et al. (2020).



Figure 1.42. Voxel after voxel association for realizing a possible evolving object

## 1.4.1.1. Starting from the material

Linking with others: "This question matters less for social endurance than for the partial interdependence between heterogeneous partners, the ontological choreography where each one needs not others in general, but some others, perhaps himself, where each time in his own mode, each one has to pass through others to accomplish himself". (Stengers 2020)

We have already mentioned Stéphane Leduc who, more than a century ago, wrote: "The elementary act of life is diffusion and osmosis" (see Thuillier (1980); see also Haudin et al. (2018)). The methods considered are often thought of as deterministic: the phenomena are controlled by laws, which must be discovered and applied as faithfully as possible. On this basis, it is possible to think that we will be able to control matter, in its most intimate elements, so that it transforms itself to achieve a given form and functionality. This is what was presented in the previous section.

Historically, DNA origami makes arbitrary shapes out of pieces of DNA. It was developed by Paul Rothemund, from Caltech, USA, in 2006. The chemical properties of the molecules that make up DNA, the nucleic bases, are well known. It is known that such and such a part binds to such and such another by "Watson-Crick" bonds, and from this principle, one can manage to model 2D or 3D structures. Rothemund's technique uses a long chain of DNA from a virus, with a

linear and relatively uncomplicated structure, whose sequence is known exactly. Then, by computer, the precise places where the DNA molecule must be folded to arrive at the desired shape are located. We then synthesize small "pieces" that will link themselves exactly where we want them to, to "pin" the viral genome.

Only these "staples" need to be synthesized, the basic material being borrowed from nature, and the operations are therefore both less costly and less time consuming. These small staples will then, by the play of chemical bonds, cling to the scaffolding chain. Moreover, as these staples will also attract other molecules, they will bend the central DNA strand and give it a particular shape. It is from this folding that the original name DNA origami is derived. In Rothemund's publication (2006), images of variously shaped constructions that he made were presented. The most original are probably the world map, the snowflake and the smiling man. Of course, there are not only drawings that can be reproduced with DNA. Hao Yan's team has managed to create closed 3D shapes (Han et al. 2011; Sawyer 2011, and Figure 1.43).



Figure 1.43. DNA origami (nanometer size)

Since then, nano-motors of a few nanometers (Dureuil 2013) with controlled direction of rotation have been developed:

The rotor is made by five ferrocenes connected to a central phenyl, while a ruthenium in the center acts as a rotation axis. Finally, the whole assembly is lifted by three feet and tiols to grip the molecule to the surface. (Dureuil 2013)

These spectacular molecular machines are animated by movements under the action of an external stimulus (nano-motors, nano-elevators, nano-pins, nano-transporters). Their use, due to their extremely small size, raises the question of the energy input (stimulus) and its mechanical recovery. However, applications in

drug delivery are currently being developed (Medina-Sanchez et al. 2018; Bandari et al. 2020). Other applications, shown in Figure 1.44, of these nanoscale entities are being considered by Julin et al. (2018), Woods et al. (2019) and Novotný et al. (2020).



Figure 1.44. Application areas of nano-machines

Once science begins to understand how nature has selected certain forms and not others, both in the physical and biological worlds, it can seek to apply/transpose the corresponding mechanisms to solve engineering problems that are of interest in the manufacture of artifacts, tools or final objects. The wide variety of applications given to a generic form such as the manufacture of a functional object is not inspired by what nature does, and it is interesting to examine whether knowledge about "morphogenesis" can be used to achieve the desired form by (rather stimulated) self-organization (Baquiast 2004). As early as the 1950s, Richard Feynman predicted the development of nanomolecular machines, such as those of Jean-Pierre Sauvage, a recent Nobel Prize winner, who invented a chemical method to intertwine two ring-shaped molecules (called catenan). Figure 1.45, taken from Sauvage (2017), shows some examples of such systems.



Figure 1.45. Examples of molecular machines and their movements

This is always a difficult problem in the case of linear systems which are generally ill-conditioned (see Mugnier (2008)), especially with noisy signals. This being the case, the stakes are high, as mastering the inverse problem can be an integral means of realizing an object from an easily realizable matrix whose deformation as a function of time or supplied energy could enable the realization of a complex-shaped object. Thus, according to Sussan (2011), it is now possible to create DNA Origami by computer. But it is already less satisfactory when we become interested in larger elements, since the number of chemical bonds to be considered increases with the power of three of the size (see, for example, Zentel (2020), Sanchez (2011) and Kotov (2017)). It does not yet seem possible to develop systems in which the object would be "entrusted" with a form of intentionality, thus, in the limit, leaving it the choice to look for what it needs to make itself, and thus pass self-organization with the selection of the final object.

However, these exciting aspects are still anecdotal (but more work needs to be done before we abandon this avenue). Another example is that of self-repairing cell growth, adapted to hydra, but not directly to humans. Turing (1952) showed how reactions between chemical substances coupled with a diffusion-reaction process of these substances could give rise to the appearance of specific structures or regularities (spots on a coat, shape of a shell, etc.). Will we ever be able to complete and experiment with his mathematical model of "reaction-diffusion equations" to create well-defined objects? The road does not exist yet, but... Besides, we have not, in Demoly and André (2021), found the right way!

Independently of this aspect linked to a spatial change induced by a stimulation, we find all the molecular chemistry which is largely out of the context of this work. However, and because the color change is mentioned in 4D applications, we have chosen to quickly present some photochemical reactions, including photochromic reactions. Photochromism corresponds to a reversible reaction of a chemical species between two forms, A and B, induced by light for the transition  $A \rightarrow B$  with a return  $B \rightarrow A$ , which can be photochemical or thermal. The transition between the two species can be manifested by various changes such as refractive index, solubility, viscosity, wettability of a surface or dielectric constants. This phenomenon can be observed in both organic and inorganic compounds (see, for example, Crano and Guglielmetti (1999), Dürr and Bouas-Laurent (2003), Irie et al. (2013) and Knyazhanskiy (2018)).

Moreover, nature, even if only via DNA or RNA knows how to dispose of information and is able to process it through chemical or biochemical pathways, within a reaction network (Dramé-Maigné et al. 2019). According to Ahmady (2017) "molecular computing" is defined as "the processing or storage of computer data using circuits or components made of living or non-living molecules, replacing

traditional materials such as copper or silicon". The idea is to use the properties of molecules: charge, structure, volume, polarity... to design new computing and storage models. Using these principles, it is in principle possible to:

find a solution to a combinatorial search problem (using all combinations of molecules, and their hybridization, which realizes, in a way, a parallel computer), but it is also possible to create "logic gates" ("logic switches", at the basis of computer science, such as AND, OR, XOR...) by relying on the links between DNA molecules. (VMF214 2017)

Many scientific studies explore the potential of molecular circuits to optimize modern technological tools. Examples of programming based on these principles have already been published (Pevzner 2010; Fujii and Rondelez 2013; Thubagere et al. 2017; Zadorin et al. 2017; Cherry and Qian 2018). Thus, with a small number of molecules or macromolecules in a network, these authors show the possibility of considering information processing through other routes than the current systems. If this field is clearly outside the authors' competence, this vision fits perfectly into the "4D doctrine" and should be followed up (monitoring), and there should be joint reflections with information science specialists to increase the applicative potential of the field, which, moreover, could lead to a large gain in legitimacy.

# 1.4.1.2. Starting from ordered voxels

Manufacturing differs from action in that it has a definite beginning and an end that can be fixed in advance. (Arendt 1989)

The current scientific context does not mean that the spontaneous complexity that was discussed above is abandoned, but that the engineer and/or designer is trying to regain control over autonomous phenomena that are difficult to control by accepting the manipulation of non-living matter and energy so that the initial forms adapt to their will. On the basis of KIS ("Keep It Simple"), publications either deal with deployable objects based on deformable origami, or with 3D objects containing matter that can be activated by different stimuli (pH, heat, light, electromagnetic field, etc.). We are therefore moving from a global approach to a more traditional methodology: creation of a 3D part using different methods, including additive manufacturing with active materials, followed by localized stimulation in space and time to change the 2D or 3D shape of an object.

#### 1.4.1.2.1. Nanometric aspect

A world transformed by the mind does not offer to the mind the same perspectives and the same directions as formerly; it imposes entirely new problems, innumerable enigmas. (Valery 1931) More than 25 years ago, the advent of near-field microscopy changed the landscape of condensed matter research. Based on the detection of a signal from a short-range interaction between a fine probe (a tip) and the object of study (surface, nano-objects, etc.), these microscopies have not only enabled the observation of the nanoworld, but also permitted the nanostructuring of surfaces.

Thus, in addition to their major role in surface physics and physicochemistry, these microscopies have become essential in many current research fields, such as molecular electronics, spintronics, superconductivity, nanostructure optics, nanoplasmonics, mesoscopic physics, etc. (INSP 2020; Tian et al. 2020)

But, for the most part, it is about moving atoms on the surface, rarely in volume.

## 1.4.1.2.2. Centimetric aspect

Intellectual judgments are regulated by emotions, not dispassionate analysis, and the goal is to minimize threats to the "self". (Western et al. 2006)

It has just been shown that it is possible to chemically produce nanometric structures whose shape changes in the presence of a stimulus. But to reach centimetric dimensions, we can imagine the difficulty of chemically manufacturing a molecular or supramolecular assembly that would contain about  $10^{22}$  atoms! So, it is possible to use elements of reasonable size that can be moved to achieve a given shape. These small robots are assembled and disassembled according to the programming that organizes their movements. This is how the cubes of Daniela Rus' team at MIT move to create real 3D structures, but still with a very limited number of voxels, as shown in Figure 1.46 (Gilpin et al. (2012); see also MIT (2019)).



Figure 1.46. Example of 4D printing using cubes (according to Sussan (2014))

The technology for the programmed assembly of homogeneous or heterogeneous modular robots is still in the research stage (Fischer et al. 2014; Thomas et al. 2014; Bakarich et al. 2015; Bückmann et al. 2015; Crawford 2015; Li et al. 2015; Abtan 2019, etc.). Indeed, as soon as the number of robots increases, the programming of

their movements must avoid collisions, while optimizing the travel times of each of the mobile entities (André 2017), whether for intra- or inter-reconfigurations (Tan et al. 2020).

# 1.4.1.3. Origami

Evolution, it is said, calls for mutations that allow us to survive and adapt to a new environment, now described as unstable, complex, and uncertain, and in relation to which our societies are constantly accused of falling behind. (Stiegler 2019)

If it is interesting to examine how to use this knowledge on Turing's "morphogenesis" to reach the desired form by self-organization. The resolution of the inverse problem, applicable to the direct manufacturing of objects, remains to be done... (Demoly and André 2021). In this field where complexity reigns supreme, we return to the pillars from which we can advance: additive manufacturing, or even more simply, origami (see Tenzeris et al. (2016)). In the latter case, the folding areas are printed (in 2D) using an active material that will fold and unfold the origami, turning it into an externally activated robot (see Figure 1.47).



Figure 1.47. Operating principle of a thermomechanical origami

Recall that one of the advantages of additive manufacturing is that "complexity is free" (André 2017). So, the deposition of an active material that will promote the spatial transformation of an originally planar structure may not be homogeneous, thus orienting the deformation in privileged directions. To come back to 4D printing itself, the "simple" solution of the engineer can also consist of the use of an active mono- or multi-material assembly with an additive manufacturing medium associated with a system allowing the movement: SMA, external actuation by pressurized gas, stimulated boiling of a liquid included in the material matrix, etc. From an "esthetic" point of view, this is basically improved 3D (but it works, as shown by the experiments carried out over 30 years ago on shape memory actuators!). So, to achieve "exciting scientific 4D printing", you just need to involve at least one active material in the additive manufacturing process. Any 3D technology can be used as long as (1) the active material does not lose its properties during the fabrication process and (2) they have the ability to print multiple materials.

For example, researchers at MIT (Hardesty 2015) have produced a small robot prototype capable of amazing feats. Indeed, this machine is capable of adopting different shapes, moving, carrying loads... capabilities that hint at many future applications. Halfway between an origami and an actuator, this small robot is indeed capable of folding in on itself in order to adopt different shapes (Ranosa 2015). But this amazing contraption is also able to walk, swim and carry loads heavier than itself before self-destructing in acetone (see Figure 1.48). Other origami inspired systems have been published recently (see Schulz et al. (2017), Abtan (2019), Callens et al. (2019) and Liu et al. (2019)).



Figure 1.48. Origami robot moving an object (from start to goal)

According to Hornyak (2019), robots without control by a CPU have been realized at MIT. The robots are connected by magnetic force and move by expanding or contracting. They use local measurement of light flux to decide when to start an expansion or contraction cycle, thus forcing the units to move in groups. This 2D extension can allow a "physical" simulation of the functioning of living cells.

#### Box 1.9. Small robots

## 1.4.2. Materials for 4D printing

It seems to me, then, that knowledge is nothing else than the perception of the connection and propriety, or of the opposition and disconvenience which lies between two of our ideas. (Locke 2019)

Several methods can be selected in terms of reversible movements with different stimuli external to the 3D object (e.g. Sossou et al. (2019a, 2019b) and André (2017)):

- Polymers sensitive to moisture, heat: either the polymer absorbs water or loses it, its density changes with a possible shrinkage (swelling/contraction). There are also reversible systems sensitive to heat (SMPs) with changes in density.

– Photochemical muscles: irradiation induces internal rotations in the material (classic example of azobenzene), which contribute to modifying the geometry of the irradiated area, leading to deformations that depend on the luminous flux received; these can be reversible if photochromic materials are available (reversible transformation).

- With multi-material systems, it is possible to play on the difference in behavior of materials with a stimulation to produce "bilayer effects".

- With charged materials, an electromagnetic field can induce a charge orientation leading to a desired deformation of the 3D object, etc.

## 1.4.2.1. Active materials

Unidirectional thinking is systematically favored by policy makers and their suppliers of mass information. Their discursive universe is full of assumptions which find their justification in themselves and which, repeated incessantly and exclusively, become hypnothic formulas, diktats. (Marcuse 1964)

We examine here how materials can be sensitive to a stimulus, independently of the additive manufacturing processes. Indeed, given the proposed manufacturing method, it is possible to introduce voxels in active materials into the object without them coming from additive manufacturing. The main information comes from: https://www.hisour.com/fr/electroactive-polymers-42852/.

Electroactive polymers, or EAPs, are polymers that exhibit a change in size or shape when stimulated by an electric field. The most common applications for this type of material are in actuators and sensors. A typical characteristic property of an EAP is that it will undergo a large amount of deformation while maintaining large forces (Bar-Cohen 2004; Lendlein 2010; Hu et al. 2012; Hager et al. 2015; Duduta et al. 2019; Lendlein and Gould 2019).

The majority of historical actuators are made from piezoelectric ceramic materials. Although these materials are capable of withstanding large forces, they typically deform only a fraction of a percent. In the late 1990s, it was demonstrated that some EAPs could exhibit up to 380% strain, which is far more than any ceramic actuator. One of the most common applications of EAPs is robotics in the development of artificial muscles (Maziz et al. 2017; Wang et al. 2017); thus, an EAP is often called an artificial muscle. EAP exhibiting a reversible transformation characteristic can have several configurations, but is generally divided into two main

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classes: dielectric (Du et al. 2015) and ionic (Carrico et al. 2019). Table 1.6 brings together the knowledge and properties of these materials. Figure 1.49, from Ganet-Mattei (2018), positions these compounds in a Young's Modulus-strain diagram.

Functionality	Comments
Electrostrictive polymers and dielectric elastomers	Actuation is caused by electrostatic forces between two electrodes that squeeze the polymer. Dielectric elastomers are capable of withstanding very high stresses and are basically capacitors that change capacitance when a voltage is applied, by allowing the polymer to compress in thickness and expand in surface due to the electric field.
Ferroelectric polymers and ferrofluids	They retain a permanent electrical polarization that can be reversed or switched in an external electric field. Ferrofluids are colloidal suspensions of ferromagnetic or ferrimagnetic nanoparticles of the order of 10 nm in size (Schärtl 2010; Kim et al. 2018).
Electrostrictive graft polymers	Electrostrictive graft polymers consist of flexible back chains with branched side chains. When an electric field is applied, a force is applied to each partial charge and causes the assembly to rotate. This rotation causes electrostrictive stress and deformation of the polymer.
LCEs	Synthesis of highly oriented elastomers leads to significant stress thermal actuation in the polymer chain direction, with temperature variation resulting in unique mechanical properties and potential applications as mechanical actuators.
Ionic polymers	The activation is caused by the displacement of ions inside the polymer (conducting polymers).
Electro-rheological fluid	Electro-rheological fluids change the viscosity of a solution with the application of an electric field. The fluid is a suspension of polymers in a liquid with low dielectric constant.
Ionic polymer-metal composite	Ionic polymer-metal composites act by electrostatic attraction between the cationic counter ions and the cathode of the applied electric field.
Stimuli sensitive gels	They are swellable polymer networks with volume phase transition behavior. These materials reversibly change their volume, optical, mechanical and other properties by small modifications of some physical (e.g. electric field, light and temperature) or chemical (concentrations) stimuli.
Stimulus sensitive materials	These are materials that are sensitive to light, humidity, temperature, etc. (see André (2017)). Produced in additive manufacturing, the objects are derived from so-called homogeneous 4D printing.

 Table 1.6. Stimulable materials



**Figure 1.49.** Young's modulus-strain diagram (1: piezoelectric ceramic; 2: piezoelectric polymer; 3: shape memory actuator; 4: EAP; 5: hydraulic actuator; 6: pneumatic actuator; 7: solenoids) – Energy densities: 1: 10 GJ.m<sup>-3</sup>; 2: 1 GJ.m<sup>-3</sup>; 3: 100 MJ.m<sup>-3</sup>; 4: 10 MJ.m<sup>-3</sup>; 5: 1 MJ.m<sup>-3</sup>; 6: 100 kJ.m<sup>-3</sup>; 7: 10 kJ.m<sup>-3</sup>; 8: 1 kJ.m<sup>-3</sup>; 9: 100 J.m<sup>-3</sup>; 10: 10 J.m<sup>-3</sup>

#### 1.4.2.2. Conventional materials used in homogeneous 4D printing

In 2007, when the iPhone came out, the CEO of Microsoft at the time said: this object has no industrial future. (Berry 2019)

4D printed materials must react to impose environmental changes: geometric deformations can be observed as soon as an external stimulus triggers the shape memory effect embedded in the material. Microscopic movements of the polymer chains then result in global changes of shape. Several modes of stimulation can be evoked to produce a 3D object by additive manufacturing and stimulation-induced shape changes (Chu et al. 2020).

#### 1.4.2.2.1. Active polymers – photochemical muscles

One approach concerns the effect of light on the structure of a polymer like photochemical muscles (Yu et al. 2003; Nakano 2010; Yoshino et al. 2010; Jochum and Theato 2013; Khoo et al. 2015; Yu et al. 2015; Fang et al. 2017; Roppolo et al. 2017; Jin et al. 2018; Jung et al. 2020; Yoon and Bae 2020), as shown in Figure 1.50, extracted from Kuksenok and Balzs (2013). Irradiation induces internal rotations in the material, which contributes to changing the geometry of the irradiated area, leading to deformations that depend on the received flux. These can be reversible if photochromic materials are available (temporary, reversible transformation).

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The full range of processes outside of photodegradation (Khim et al. 2014) can thus be exploited in 4D printing. Curvatures of polymeric fibers can be related to a non-homogeneous distribution of photo products (see Ge et al. (2016) and André (2017)). Different effects can be exploited (see Figure 1.51).



Figure 1.50. Light-induced deformation of a photoshrinkable polymer cylinder



Figure 1.51. Photomechanics of polymers

Examples include Mahimwalla's (2013) thesis showing significant deformation of polymers containing azobenzene groups (see Figure 1.52 and Mahimwalla et al. (2012)). Upon irradiation, these groups change from Trans to Cis configurations depending on the wavelength used (see Figure 1.51). The effects are notable, as can be seen in Figure 1.52 (see also De Simone et al. (2015), Rosales et al. (2015), Liu et al. (2017) and Hagaman et al. (2018)).



**Figure 1.52.** *a)* Azobenzene-based polymers and b) sizes of the two isomers (Deloncle 2007). The Trans -> Cis transformation takes place in the mid-UV (300–400 nm), the return in the visible (between 400 and 500 nm)

To alleviate this sense of cognitive dissonance, in many cases, instead of acknowledging an error in judgment, one seeks to reframe one's views in a new way that justifies the old views. (Livio 2017)

The azobenzene molecule is generally in the Trans conformation, which can be considered uniaxial. When this molecule is exposed to visible light, it can undergo photoisomerization and change to the Cis conformation (see Figure 1.53). The molecule can then return to the Trans conformation following a thermally or optically induced transformation. Once this cycle is completed, the molecule can be in a new orientation. This process can be used to affect the macroscopic orientation of the molecules in a film. When linearly polarized light is used to activate the Trans -> Cis photo-reorientation cycle, only molecules whose electronic absorption transition moment is in the direction (or in the vicinity) of the polarization absorb light and undergo an orientation change. In this way, it is possible to reduce the number of molecules oriented parallel to the direction of polarization and increase the number of molecules oriented perpendicular to that direction (Rochon 2015).

These authors present rather dramatic results from Yamada et al. (2009), corresponding to the deformation of a polymer film induced, both by UV irradiation (Trans -> Cis transformation) at 366 nm (240 mW.cm<sup>-2</sup>) and in the visible

 $(\lambda > 540 \text{ nm and } 120 \text{ mW.cm}^{-2})$  for the reverse transformation. Figure 1.54, kindly provided by Ge (2016), shows a reversible transformation of a polymer that deforms with temperature. Figure 1.56 by Cheng et al. (2010) complements the previous one: with a double layer of polymers, one of which contains azotolane that can undergo Cis -> Trans thermal and Trans -> Cis photochemical isomerization in the visible, it is possible to induce a temporary shrinkage of the irradiated film, followed by a return to the initial position after stopping the irradiation (see also Figure 1.56 by Zhang et al. (2015)). Other works may address this transformation principle (Ercole et al. 2010; Ilic-Slovanovic et al. 2011; Petr et al. 2015; Gao et al. 2016). As a side note, given the molecular structure of azobenzene and azotolane, it should be possible to take into account the orientation of the transition moments of these molecules placed in a polymeric system to make transformations in spatially resolved polarized light, to create for example programmable diffraction gratings (see above). Robotic applications are envisioned by Umedachi et al. (2013).



Figure 1.53. Effect of irradiation on a polymer film containing azobenzene groups: a) before irradiation, b) immediately after according to Mahimwalla et al. (2013)





Figure 1.54. Light-induced deformations of a polymer film (Ge 2016)



Figure 1.55. Polymeric bilayer under light irradiation

Such systems can be realized in additive manufacturing using various passive, yet highly elastic materials (e.g. with epoxy-based (monofunctional) and urethanebased (bifunctional) mixtures of aliphatic acrylic monomers published by Patel et al. in 2017).



**Figure 1.56.** Light-induced deformations of a polymer film according to Zhang et al. (2015)

Another example from the work of Chen et al. (2016): active polymers, ready to respond to blue light stimulation (from an LED), containing tri-thiocarbonate (TTC) moieties with an accordion structure can be activated by organic catalysts. In the presence of light, monomers are formed (reverse process of polymerization), stretching the macromolecule. As the monomers are uniformly distributed throughout the structure, they impart new properties to the material, which then allows the shape of the object to be changed. This complex technique requires specific conditions of use.

## 1.4.2.2.2. Active polymers - thermal effects on polymers

We no longer even understand how [objects] work, we have even become individually incapable of reproducing or repairing them. (Ariès 2010)

For Zhao et al. (2015), it is possible to have polymeric materials whose shape can depend either on temperature or on photochemical transformations. For these authors, it is a matter of finding reversible reactions affecting the space between molecules or macromolecules (see Figure 1.57) (see also Dutta and Cohn (2017), Zare et al. (2019), Zare et al. (2019), Yang et al. (2021a) and Aziz et al. (2021)).



Figure 1.57. Reversible systems with volume change

An explanatory example is given in Figure 1.58, illustrating the shape change. Other examples are given in this review article on different active materials.



Figure 1.58. Principle of thermally induced reversible deformations

Inks made from hydrogels are capable of moisture- and temperature-induced deformation (André 2017). Le Duigou et al. (2016) used plant particles as a filler in a polymer (see also Gauss et al. (2021)). Oriented in the extrusion shear of this meltable polymer, they retain the memory of their orientation below the gel point of the material and can see their shape change in the presence of moisture (or heating). Finally, from an optimization standpoint Zhang et al. (2011) added carbon nanotube fillers the heatand light-sensitive (hydrogel) to material. polv-(Nisopropylacrylamide) (pNIPAM), which facilitates heat transfer into the bulk of the material and light absorption on the surface. Han et al. (2012) exploited pH-induced transformations.

In Table 1.7, Rastogi et al. (2019) proposed liquid crystal transformations induced by photo-thermal effects (heating by light) (see also Rousseau (2008), Plucinsky et al. (2018), Roach et al. (2018) and Longo et al. (2019)).

The selection of materials is therefore very important when trying to define a target application. Pinho et al. (2020) consider that the chemistry responsible for the morphing effect of polymeric materials is not explicit enough, despite the growing number of articles published on this topic. They write: "For specific applications, only chemical modification of polymeric materials can achieve the characteristics required for 4D printing". For electro-thermal effects, Chen et al. (2021b) show that the presence of carbon fibers increases the strength of the 4D printed composite, and also improves the recovery strength of the active form.

Properties/nature	Polymer	Elastomer	Network
Cross-linking	No	Low	Important
Change after stimulation	No	Approximately 5%	>90%
Shape memory	No	Yes	Yes
Glass transition temperature	200°C	25°C	Between 60°C and 100°C
Modulus of elasticity	>2 GPa	<100 MPa	Intermediate

 Table 1.7. Performance comparison of liquid crystals usable in 4D printing

### 1.4.2.2.3. Active polymers – swelling effects in polymers

We take as true what is credible, what we want to believe. However, we must sometimes inhibit our automatic beliefs in order to develop more logical, rational thinking. (Houdé 2019)

Other physical phenomena can be exploited to see a 3D object made of an environmentally sensitive material change shape: pH, presence of a solvent, etc. (Bakar and Djaider 2007; Liu et al. 2017, Lv et al. 2018; Abdelmohsen et al. 2019). The swelling of cross-linked polymer networks in the presence of a solvent is a well-known phenomenon (Ancla 2010; Bounouira 2015). In the latter case, shown in Figure 1.59, very noticeable changes in the shape of a 3D object can be observed.



Figure 1.59. Shape changes in the presence of a solvent

With certain solvents, the chain sections between two cross-linking points unfold as in the case of linear chains, but this movement is limited by the existence of bridges. The phenomenon is therefore limited to swelling, which is more pronounced the lower the degree of cross-linking. The term hydrogel defines materials with a liquid and a solid component. They are composed of polymer chains assembled via the cross-linking process and can contain liquids such as water. Hydrogels can be used in additive manufacturing and have their shape change in the presence of a solvent (see also Oveissi et al. (2021)). Figure 1.60, from Curatolo et al. (2021), illustrates how a network can be deformed by the presence of micro-molecules or spheres.



**Figure 1.60.** Deformation of a network by the presence of active molecules or nanospheres

Table 1.8, taken in part from Gao et al. (2016), gives indications on processable materials and on the processing methods applied in this area.

Type of material	Mechanism and comments	References
Polyethylene glycol; lipid bilayers	Contact with water	Villar et al. 2011, 2013; Jamal et al. 2013
Low transition temperature polymers (PNIPAAm; PCL); ceramic alumina	Transition temperature	Peppas et al. 2000; Bakarich et al. 2015; Wang et al. 2015
Magnetic materials	Precise positioning by locating the magnetic field	Kokkinis et al. 2015; Zhu et al. 2018; Wu et al. 2020
Polymers containing light isomerizable groups	Photochromes	Zhang et al. 2015

Table 1.8. Different types of informed materials and associated mechanisms

# 1.4.2.2.4. Electrochemical effects

You don't know [...] that there is life only in differences, difference of temperature, difference of potential. And if the same heat or the

same cold reigns everywhere in the universe, it is necessary to shake them so that the fire, the explosion, the Gehenna are born. We will shake them. (Zamiatin 2020)

Figure 1.61 schematically represents the effect of ion transport on polymer conformation, according to Kaneto (2016). Either ions are made to act and we can engage in 4D printing, or we have a means of dosing the ions (actuator/4D printing reciprocity).



Figure 1.61. Conformation/ion relationship: Acrylic type polymers are "welded" or rather cross-linked and in the 3D structure thus generated, the presence of ions disturbs the shape of the structure

## 1.4.2.2.5. Biological and organic materials

Experience is the name everyone gives to their mistakes. (Wilde 2017)

Plants exhibit spontaneous curvature changes due to the orientation of cellulose, which undergoes anisotropic swelling in the presence of water. Gladman et al. (2016) mimicked this process with a hydrogel containing such rigid cellulose fibrils. They spontaneously align under shear as they flow through the printer's extruder nozzle, inducing anisotropies in swelling and elastic modulus. With positioning in bilayers, differential swellings can be achieved, resulting in a (calculable) curvature

of the object. Using anisotropy in the material, Qe et al. (2016) showed in Figure 1.62 how it is possible to develop the shape of an object depending on the stimulation it receives.



Figure 1.62. The anisotropy of the material creates a curvature when the temperature changes (the same phenomenon could have been obtained with the swelling of bi-layers of cellulose loaded threads). Complex morphologies (here a flower) are generated with changes in the shapes of the objects

Heating

**Temporary Shape** 

5 mm

III

mm

2 mm

11

2 mm

2 mm

IV

Lind et al. (2017) used heart muscle cells that contract under the influence of electrical stimulation as an activatable material. Figure 1.63 illustrates the compression/expansion phenomenon to bend a plastic material (see also Jackson (2016)).



Figure 1.63. Deformation induced by muscle cells

## 1.4.2.3. Elongation-Young's modulus relationship

Men are conscious of their desires and ignorant of the causes that determine them. (Spinoza 1955)

In his thesis, He (2021) relates this important aspect to design deformable assemblies in 4D printing. Figure 1.64 shows typical results with considerable variation depending on the materials and activation modes chosen.



**Figure 1.64.** Steady-state actuation performance of LCE-based vascular artificial muscle. Left: specific actuation stress as a function of strain of classically used materials and mammalian skeletal muscle. Right: working density as a function of strain (Abbreviations: PP: piezoelectric polymer; MA: magnetic actuator; EA: electrostatic actuator; MM: mammalian muscle; IPMC: ionic polymer-metal composite; MS: mammalian skeleton; CNT: carbon nanotube; DEA: dielectric elastomer actuator; PAM: pneumatic artificial muscle)

## 1.4.2.4. Some typical results

Dare I state here the greatest, most important, most useful rule of all education? It is not to save time, it is to lose time! (Rousseau 1995)

Kimionis et al. (2015) used both 3D printing and origami making for the manufacture of radio frequency (RF) antennas. An example of their demonstration that couples two emerging potentials in the same object can be found in Figure 1.65.



Figure 1.65. An origami-based RF antenna realized by multi-material additive manufacturing (silver ink patch; probe feedpoint: copper tape ground plane behind plastic)

Apart from origami, which can be temperature sensitive (Ge et al. 2014), more large-scale transformations can be achieved with light-sensitive polymeric materials. Another approach involves the effect of light on the structure of a polymer (Nakano 2010; Yoshino et al. 2010; Kuksenok and Balzs 2013; Gao et al. 2015; Petr et al. 2015; Patel et al. 2017; Shao et al. 2018; Jayashankar et al. 2019; Kriegman et al. 2019; Goo et al. 2020; MIT 2020): Irradiation induces internal rotations in the material, which contributes to changing the geometry of the irradiated area inducing deformations; these can be reversible if photochromic materials are available. Polymeric fiber bends can be related to the inhomogeneous distribution of photoproducts following either surface resolved illumination or tailored deposition, as illustrated in Figure 1.66 (Sossou et al. 2019a; see also Baker et al. (2016), Zhang et al. (2017b), Zolfagharian et al. (2018), Carrell et al. (2020) and Yuan et al. (2021)).

Another example is the significant deformation of polymers containing azobenzene groups (Mahimwalla et al. 2012; Mahimwalla 2013; De Simone et al. 2015; Rosales et al. 2015), that upon irradiation, change from Trans to Cis configurations. The effects are notable, with reversible shrinkages of over 20%. Using anisotropy in the material, Qe et al. (2016) showed in Figure 1.57 how the shape of an object can be made to change with the stimulation it receives.



**Figure 1.66.** Optimization of an active material deposit for a given deformation (Sossous et al. 2019a)

Chen et al. (2019) performed 4D metal printing using a specialized machine. This is one of the first demonstrations of metal use with the publication by Lohmuller et al. (2019). Kim (2014), however, had already made prints with piezoelectric nanoparticles. But a reverse approach has already been attempted by implementing piezoelectric materials in structures to avoid deformations of various origins (Bent and Hagood 1997). With piezoelectric fibers in an epoxy matrix, orthotropic activation can be achieved using interdigitated electrodes, producing an electric field in the plane of the structure (in the direction of the fibers). If these advances in the field of smart materials have opened up new possibilities for active control of composite structures (Gakwaya 2006), it is conceivable to use these systems to perform heterogeneous 4D printing.

However, according to Reiser et al. (2019), the synthesis of industrially good inorganic materials remains a challenge. They show that current additive microfabrication techniques lead to a wide range of microstructures and mechanical, elastic and plastic properties. While this work provides practical guidelines for users of metal additive manufacturing methods, a high level of specialized materials expertise is required.

Dufaud et al. (2002) studied the effect of electric current on composites containing PZT ceramics with piezoelectric properties aimed at the realization of actuators. More recently, Martin et al. (2015) combined magnetic particles with photo-polymerizable resins that can be specifically oriented by changing the field amplitude and orientation. This realization where two forms of energy are coupled allows the realization of structures with singular magnetic properties, along with a possible reinforcement of the material by increasing the order of the elongated particles in the polymeric matrix. Catheters for neonatal applications have been realized on this principle.

Box 1.10. Additional remarks

# 1.4.2.5. Transition between activation modes

Objectivity, associated with knowledge, relegates everything that does not belong to it, values and principles, to the side of subjectivity, belief, opinion, etc. (Roux 2011)

In André (2017a), dealing with emerging niches in 3D printing, different themes useful for the development of 4D printing were presented. These themes are presented in Table 1.9.

Theme	Comments
Doing chemistry	By chemical means, we can modify the structure of a macromolecule which, by reaction, can be deformed to create an object. However, given the number of links to be considered, these objects are limited to nanometric sizes (3D DNA; nano-machines, catenanes, etc.)
Self-organization	The example of the crystallization can be retained: if it was possible to guide the shape of the structure, it would be possible to reach in the time of the transfer of matter shapes answering the specifications
	Another example is bio-printing
Displacement of atoms	The development of near-field systems allows the displacement of atoms and the realization of 3D sub-nanometric structures
Nano-manufacturing	Additive-subtractive couplings
Swimming robots	Directed particles that gather to form an object (as long as the cohesion between these particles is assured). The number of possible particles remains modest because of the slowness of the movements
Robotic voxels	Programming the movement of voxels (autonomous robots) to create a 3D object whose shape can evolve over time. The size of the voxels and their number are factors that limit the development of this construction principle
4D origami	Foldable 2D structures can be generated by placing stimulable adaptive materials in the folds, allowing the fabrication of externally controlled centimeter-sized robots by external actuation

# Table 1.9. Other processes that may be of interest in 4D printing

In this transition, it is possible to mention associations between electrical potential and physical chemistry. Figure 1.67 illustrates the principle of deformation (Narayan et al. 2021; see also Coltelli et al. (2021) with additional microfluidic effects).



Figure 1.67. Diagram of deformation under tension due to migration of hydrated cations

Without returning to the construction of volumes atom by atom, the following sections deal with other ways than those related to chemical interactions exploiting the assistance of different types of energy sources. Furthermore, materials subjected to mechanical stress can undergo a change in their structures, properties and/or functions (Troyano et al. 2021). This mechano-reactivity can be exploited to trigger chemical transformations (Do and Friščić 2017). Poro-mechanics is devoted to modeling and predicting the deformation of porous materials in response to external loads. It could also be investigated in 4D printing.

# 1.4.3. Activations by physical pathway

Within a limited rationality, the terms of reason become disjointed: losing their unity, they lose their meaning. It seems that rationality and finality do not pre-exist the facts which carry them and which one supposes that they bring (or peddle). (Lefebvre 1971)

We have already discussed the modes of activation of materials. While most of the work deals with the chemical methods just mentioned, there are other ways of physical origin, which can make them evolve in form or in functionality or, more simply, help them to move. These are the different elements that are recalled below.

# 1.4.3.1. Mechanical effects

## 1.4.3.1.1. Internal effects

Grossweiler et al. (2015) use polymers containing spiropyrans that are sensitive to mechanical stress, these deform and change color with a relaxation time of several minutes (see Figure 1.68). This type of material has enabled micro-clamps to be made.



**Figure 1.68.** Effect of mechanical stress on the chemical transformation of a spyropyran-based polymer

Recall further that Vatankhah-Varnosfaderani et al. (2018) synthesized an elastomer composed of a central block, which has side chains extended by two terminal blocks grafted onto it (see Figure 1.69). The end blocks of these structures gather into nanoscale spheres, distributed in a matrix formed by the "pin-like" structures. The light interferes with this architecture according to the distance between the ends; a stretching of the material thus results in a change of color (chameleon effect). It is therefore possible to encode mechanical properties (elastomer) and optical properties in a single polymer, with modulation possibilities. By imposing a mechanical deformation on an object, it is therefore possible, in principle, to show where the areas of mechanical stress are located.

Qian et al. (2021) teach us that precise mechanochemical transformations in polymeric materials are poorly explored. Before the advent of mechanophores (synthetic force-sensitive molecules) few mechanosensitive functions were known in polymers (Nixon and De Bo 2020). Mechano-actuation has a directional parameter that, in turn, is coupled with the geometric attributes of the mechanophore through its points of attraction:

thereby tuning the force responsiveness. In other words, by modulating the attachment points of a mechanophore in polymer chains, the force-induced reactivity of the mechanophore and the reactivity of the material vary significantly. (Church et al. 2014; Gossweiler et al. 2015; Stevenson and De Bo 2017)



Figure 1.69. Active structure of the chameleon type

### 1.4.3.1.2. External effects

Even in its most fortuitous forms, chance does not make the discovery: it is only the occasion. (Picard 1928)

These can be, as shown in Figure 1.70(a), simple and fairly traditional solutions of actuators made by additive manufacturing (Peele et al. 2015; see also Walters et al. (2010) with a pneumatic actuator principle using origami in Martinez et al. (2012)). In this figure, the left or right displacement depends on the difference in gas pressure between the two compartments of the actuator. Foldable systems whose fast internal swelling (compared to response times obtained with solid materials) leads to interesting shape changes like those in Figure 1.70(b) of Yuk et al. (2017) correspond to an object made in additive manufacturing, whose shape can be modified by the effect of hydraulic pressure. Recently, Sparrman et al. (2021) produced a study in this same field with flexible fluidic actuators (whose principle is based on the historical McKibben actuator) made with silicone polymers in 3D printing, with the possibility of operating them sequentially for complex deformations (application in soft robotics - see, for example, Franco et al. (2020)). Other (simultaneous) actuation devices are proposed by Shao et al. (2021) with magnetic actuation. In this last case, a change of scale is sought for applications in micro-robotics (millimeter size).



Figure 1.70a. Principle of a pneumatic 3D actuator (André 2017)



Slow osmotic actuation (actuation frequency =  $1.4 \times 10^{-5}$  Hz)

Figure 1.70b. Hydraulic actuator (Yuk et al. 2017)

# 1.4.3.2. Physical changes

Any action is risky. But inaction is almost always more risky. There is no progress without risk. (Comte-Sponville 2015)

#### 1.4.3.2.1. Macroscopic effects

MIT (2013) has shown that on simple examples, it is possible to "order" the material if only by visual recognition. This idea, illustrated in Figure 1.71, is based on the displacement of cylinders as used in two dimensions by carpenters to take the impression of a cornice, or as some toys to achieve three dimensions. If a numerical control is added to the displacement of each cylinder, it is possible to make the material "programmable" directly or indirectly. However, in these two examples that serve as an introduction to the subject, the material is inert. The idea is to go a little further in the process by "informing" the matter directly, at finer scales than the one in this figure.



Figure 1.71. Reversible sculpture toy (Cdiscount 2017)

Figure 1.72 with the idea from MIT (2013) shows the intrinsic complexity of such a system that will not be able to scale down due to the very large number of actuators that would need to be put together in a finite volume (voxel size problem to consider).



Figure 1.72. MIT experimental setup (2013)

#### 1.4.3.2.2. Microscopic effects

The necessity that all sciences be directed towards a useful goal, and that the point of coincidence of all their discoveries be the physical and moral prosperity of the French Republic. (Abbé Grégoire, quoted by Lévy-Leblond (2006))

A surfactant is a molecule with a hydrophilic head and a hydrophobic tail, the hydrophilic head being preferentially oriented toward the aqueous phase, unlike the hydrophobic tail. Thus, surfactants are generally positioned preferentially at the water/air or water/oil interfaces.

Their presence stabilizes the interfaces, which implies a local decrease of the surface tension. As the presence of surfactants on a surface decreases the surface tension locally, it is possible to create a surface tension gradient. This results in a movement of fluids toward areas of higher surface tension. (Marques-Serra 2014), as illustrated in Figure 1.73

The addition of a surfactant (orange) causes a decrease in surface tension from  $\gamma_0$  to  $\gamma$ , which induces to a deformation of the surface that leads to a flow of fluids (blue arrows) toward the areas of higher surface tensions  $\gamma_0$  (André 2017).



Surfactant or soap

Figure 1.73. Surfactant-induced deformation of a fluid

If an active surfactant (e.g. containing azobenzene) is used, Trans-Cis isomerization is likely to change the surface tension, which may allow material flow. Under UV irradiation the concentration of Trans isomer decreases, which increases the surface tension. Depending on the light distribution, a surface tension gradient can be obtained with the possibility of having a photo-induced Marangoni effect. Thus, if a drop is deposited on a surface, it will be able to move on it by playing, as shown in Figure 1.74, on the changes of the surface tension induced by light (Shin and Abbott 1999).



Figure 1.74. Possibility of guiding the movement of a drop on a surface

Considering the energies involved, if this technology enables droplets to move on a flat horizontal surface, it can only be considered with immiscible fluids of the same density (see swimming robots in section 1.4.4) for an application in additive manufacturing.

With Trans-BTHA, Shin and Abbott (1999) showed that the irradiation of hanging drops could cause their fall. From there, to consider a principle of rapid deposition of material by this process, there is only one step... (see Figure 1.75 representing the structural formula of the surfactant considered).



Box 1.11. Influence of the surfactant

### 1.4.3.3. Deformation of metal parts

Scenario method: "[Kahn and Wiener's] method turns out to be fundamentally heuristic: they do not seek to know whether the hypotheses put forward are true or false, but rather to explore progressively, through them, problems that may arise". (Durance 2014)

## 1.4.3.3.1. Drops

As an initial example, the 3ders.org paper (2014) announced the possibility of rotating and moving droplets of a Ga/In/Se alloy melting at 10°C, floating (due to surface tension) on water via a magnetic field. Sheng et al. (2014) from the Chinese Academy of Sciences in Beijing were able to change the shapes, as well as the sizes of metal droplets. It will be interesting to see how we can access the third dimension, and then the fourth. Figure 1.76 illustrates the effect of an electric field (New Scientist 2015).



Figure 1.76. Effect of an electric field (left) on the shape of alloy droplets

### 1.4.3.3.2. Oriented magnetic architectures

Martin et al. (2015) have combined magnetic particles with photopolymerizable resins that can be specifically oriented by changing the field amplitude and orientation. This realization where two forms of energy are coupled enables the realization of structures with singular magnetic properties, along with a possible reinforcement of the material by increasing the order of the particles arrayed in the polymeric matrix. Catheters for neonatal applications have been realized on this principle.

## 1.4.3.3.3. Shape memory alloys

Don't kid yourself, don't abuse yourself, with the understanding that you yourself are the easiest person to abuse. (Feynman 2000)

SMAs can be used (Ballandras et al. 1996) to realize actuators by heterogeneous 4D printing. These alloys have several properties that are new among metallic materials: the ability to keep an initial shape in memory and to return to it even after a deformation; the possibility to alternate between two previously memorized shapes when its temperature varies around a critical temperature; and a superelastic behavior allowing elongations without permanent deformation that are higher than those of other metals. Among the major SMAs are a variety of alloys with nickel and titanium as primary constituents in nearly equal proportions (Meier et al. 2012; Dadbakhsh et al. 2014). Although the name "nitinol" is actually only the name of one of these "quasi-equi-atomic nickel-titanium alloys", this name has become commonly used in the literature to refer to all of these alloys, which have very similar properties. To a lesser extent, some copper-aluminum-based alloys also possess shape memory properties (Wikipedia 2015). While these materials have not really been used in additive manufacturing, their application areas now span medicine, aerospace, automotive, women's clothing, eyewear, security and research... (Nimesis 2017). This development has long been limited by poor durability in their ability to return to their original shape (Chluba et al. 2015). However, everyone remembers a now classic example, that of Uri Geller's demonstration, in front of an astonished audience of the deformation of spoons made with such alloys by thermal effect (L'arrêt public gouverne MENT 2010). Several applications in additive manufacturing can be considered (keeping in mind small volume variations, but changes in shape that can be reasonable). In particular, as shown in Figure 1.77, SMAs have a specific advantage (relative to polymers), which is their good weight/power ratio, increasing their potential use as actuators by 4D printing.



Figure 1.77. Power/mass ratio of SMA linkages versus motors according to Patoor (2006)
Example: It is possible to 4D print various structures made of composite filaments, for example polylactic acid (PLA) and PLA/FeO<sub>34</sub>. The shape memory behaviors of 4D printed structures triggered by a magnetic field have been studied by Zhang et al. (2019). The printed structures can quickly recover their original shape. Furthermore, the structures activated by a magnetic field at 27.5 kHz have a uniform surface temperature around 40°C, which is consistent with a physiological operating temperature.

## 1.4.3.4. Intermediate conclusion

For if I wish to speak the truth, I will have to talk in the way I've described. Whether it's the business of a philosopher to tell lies, I don't know, but it certainly isn't mine. (More 1997)

This section is partly taken from André (2017) with a modest update. Indeed, this domain stemming from physico-chemical interactions in matter seem to be of less concern to researchers than the more large-scale interactions between different forms of energy and matter (NAP 2019b). The original component of this recently emerged 4D domain to open up additive manufacturing to other application niches lies in the principle of informing matter to extract spatial functionality (rather than being part of spontaneous self-assembly). At this stage, while the photochemical muscles have already been the subject of work for many years, to the knowledge of the authors, the idea which consists of starting from a simple form, easily realizable by traditional techniques (i.e. molding and traditional machining) for after treatment by light (or any other form of energy localized in space and time), to obtain a complex object has not been emitted. It raises the question of having materials with high expansibility (remember that for SMAs, the difference between the two shapes is generally of the order of a few %), to be able to treat an inverse problem. Starting from a shape to be reached, what could be the initial shape?

In any case, while this path seems interesting, it would be advisable to begin studies on very simple objects to deduce laws. Obviously, it would be necessary to work on surfaces rather than volumes to simplify the situation. However, the number of influencing parameters to be taken into account may be high, leading not to a unique solution for a given spatial resolution, but to a set of solutions (and at the same time to the absence of solutions for too complex shapes). Thus, starting from a continuous material, the formation of a percolating medium, for example, could be problematic.

### 1.4.4. A transition to 4D printing: swimming robots

Breakpoints reveal situations where the existence of unstable conditions makes it impossible to predict future events because our

knowledge of the present state is only approximate and far from reality. (Goux 2014)

There is a possibility to move a droplet of inert matter by bringing an external energy onto it (see Sheng et al. (2014)). Based on this principle, tiny swimming robots containing iron oxide particles, bound together by chemical bonds, can move by magnetic force (Cheang and Kim 2016). In this paper, these particles are not intended to assemble to create an object, but, with a size on the order of a nanometer, this object must be able to move through the vasculature from an external magnetic field (see Rao et al. (2015), and Figure 1.78). Indeed, the swimming of robots is well controlled, as they move at low Reynolds number:

Indeed, the Reynolds number of the flow is very small, on the order of 0.00001, whereas in usual flows (that we have to consider for a human swimming in water, for example) this number is typically on the order of 100,000. (Alouges 2012)

A swimming robot consisting of three spheres joined in a line has been studied by Alouges et al. (2008), Naja and Golestanian (2004) and Garcia (2013), where it is shown that it can indeed swim (slowly) in a 3D flow with low Reynolds number.



**Figure 1.78.** Movement principle of swimming robots (different types: translation (T); rotation (R); circular movement (CM))

Even if swimming robots can be considered as solid structures, they have their own energy for their movements or have a movement stimulated by an external energy. They are therefore systems exploring both space and time.

#### Box 1.12. Swimming robots

If the vision of the researchers is to transport drugs (cargo effect) or to use the swimming robots to clear blocked vessels (ice-breaking effect), it is possible to use, in a voxel, transformable matter which, in weightlessness, can be guided on an object under construction. The principle of this construction requires precise guidance of each robot considered as isolated from other robot-swimmers (otherwise, apart from the feasibility of precise control, these entities have density-and flow-dependent behavior, as shown by Lushia and Peskin (2013) and Liebchen et al. (2016)). In groups of swimming robots, using particles of varying sizes, it is possible to consider the construction of nanoscale or larger objects, via a "multi-voxel" principle, or even multi-material with voxels of various sizes (see Figure 1.79).



**Figure 1.79.** Application of swimming robots to additive manufacturing, for example in light-induced polymerization

Different guidance techniques can be considered, and they are presented in Table 1.10, widely inspired by Rao et al. (2015).

Principle of locomotion	Materials	Comments	References	
Electrophoresis	Microspheres, bimetallic rods, fibers	Known mechanisms, self-generated electric field, sensitivity to the conductivity of the medium, high velocity > 100 μm.s <sup>-1</sup>	Paxton et al. 2005; Wheat et al. 2010; Dou et al. 2016; Katuri et al. 2017; Brooks et al. 2019	
Diffusio- phoresis	Heterogeneous voxel	Self-generated concentration gradient with enrichment of the carrier fluid with dissolved substances during the process (difficult to apply in additive manufacturing)	Howse et al. 2007; Ibele et al. 2009; Hong et al. 2010; Clement et al. 2016; Clement 2016	
Thermo- phoresis	Nanoparticles	Gallium heated by NIR laser	Wang et al. 2021a	
Separation	Colloid/ surfactant	Elimination of water molecules at the interface	Youssef et al. 2016	
Chemo-taxicity	Gold nanoparticles	Glucose oxidase in the presence of glucose	He et al. 2019	
Bubble propulsion	Elongated shapes	Catalysis of the decomposition of a reagent producing bubbles, but uncertain trajectory	Gibbs and Zhao 2009; Solovev et al. 2009; Wilson et al. 2012; Zhang et al. 2015a; Hu et al. 2017a; Patino et al. 2018	
Electric field (1)	Micro- particles	Localized electrolysis of water producing bubbles, difficult precision and slow movements	Calvo-Marzal 2010; Loget and Kuhn 2011; Katuri et al. 2017; Rajonson et al. 2020	
Electric field (2)	EAPS	Robot weighing about 100 g	Tang et al. 2017; Wang et al. 2019	
Magnetic rotation	Flexible elongated shapes	Rotation of the swimmer in a magnetic field inducing locomotion, possible precision but slow speed	tation of the swimmer magnetic field inducing ocomotion, possible ecision but slow speed Ghosh and Fischer 2009; Gao et al. 2010, 2011; Peyer et al. 2012; Tottori et al. 2012; Li et al. 2016a; Shi et al. 2019; Wu et al. 2021	
Ultrasound and acoustic waves	Metal particles Particles	Conversion of energy into motion, uncontrolled behavior, high speed Focus of the waves	Wang et al. 2012; Ahmed et al. 2016, 2017; Kong et al. 2019 Shapiro et al. 2021	
Photochromy	notochromy Elongated shape Spiropyrans or azobenzene are susceptible to light-induced Trans-Cis transitions resulting in flagellar movement Simmch		Huang et al. 2015; Stanton et al. 2015; Li et al. 2016b; Zhang et al. 2017a; Richard et al. 2018; Heckel and Simmchen 2019	

 Table 1.10. Different forms of propulsion for swimming robots

Nature of the magnetic excitation	Speed (µm.s <sup>-1</sup> )	Comments
Internal oscillation of the particle	30.9	Fish-like movement
Oscillation	6-14.4	Sculling movement
Rotary feeder	18-40-180-250	Screw movement

For magnetic propellants, Li et al. (2016a, 2016b) give some indication of travel speeds by mode of action. These data are presented in Table 1.11.

## Table 1.11. Travelspeeds obtained by Li et al. (2016a)

In the case of the Trans-Cis transformation of azobenzene related to a light-induced transformation, the engine is related to the voltage induced by the change in shape of the azobenzene in a sandwich structure, as shown in Figure 1.80. There is, with this type of device, a way to control the partial rotation of a flagellum attached to this engine, as shown in Figure 1.81 (Huang et al. 2015; see also Egunov et al. (2016)). If the stresses are not homogeneous in the different layers, a moment of force arises and the thin layers can then bend spontaneously under the action of external forces. Thus, by exploiting these stresses, a new "self-curling" material made with cross-linked Polydimethylsiloxane (PDMS), a transparent and biocompatible elastomer and silicone oil, can be realized.





Figure 1.80. Operating principle of the photochemical engine



**Figure 1.81.** *Relationship between a photochemical engine and its flagellum* 

In their publication, Khoo et al. (2015) performed a synthesis of applicable processes in terms of informed matter. A summary of their synthesis is shown in Table 1.12.

Materials and structures	Approximate resolution	Comments
Piezoelectric nanocomposites	5 µm	Production of electrical charges when stress is applied (and vice versa); the support material is a conventional acrylic polymer such as PEGDA
SMAs (NiTi)	/	Temperature-induced shape change (hysteresis problem to be taken into account)
SMPs	20 µm	Deformation of a bi-component system with different properties (bilayer effect) induced by temperature or light (heating)
Bimaterial system including a dielectric elastomer	30 +/- 10 µm	Electrical activation and bilayer effect
Polymeric composite and origami	45 +/- 15 μm	Temperature-induced movements

 
 Table 1.12. Some examples of informed materials applied in additive manufacturing

# 1.4.5. Current scientific offer and application specifications

We will simply note that the nature and size of the object studied, or the confrontation between adjacent objects, determine different levels of structuring of interdisciplinarity. (De Beaulieu 2006)

The current scientific context presents a particularly rich set of means to make shapes evolve or modify the functionalities of structures realized at least partially by additive manufacturing. It is therefore possible to think that the engineer and/or the designer have a large offer allowing them to regain control over the phenomena of manipulation of matter and energy so that the initial forms adapt, in the course of time, to their will(s). On the basis of KIS, publications deal either with objects that can unfold based on deformable origami – 2D printing followed by folding (Gillman et al. 2019), or with 3D objects containing massive amounts of matter that can be activated by different stimuli (pH, heat, light, electromagnetic field, mechanical, etc.). We are therefore moving, in principle, from a global approach to a more traditional methodology: creation of a 2D/3D part using different methods, including

additive manufacturing with active materials, followed by localized stimulation in space and time to change the 2D or 3D shape of an object. Several methods can be selected in terms of reversible movements with different stimuli external to the 3D object (e.g. Ge et al. (2013), Sossou et al. (2019a, 2019b) and André (2017a)):

- Polymers sensitive to moisture, heat: either the polymer absorbs water or loses it, its density changes with a possible shrinkage (swelling/deflating). There are also reversible systems sensitive to heat (shape memory polymers) with changes in density.

- Photochemical "muscles": irradiation induces internal rotations in the material (classic example of azobenzene and derivatives), which contribute to modifying the geometry of the irradiated area, leading to deformations that depend on the luminous flux received; these can be reversible if photochromic materials are available (reversible transformation; bistability).

- With multi-material systems, it is possible to play on the difference in behavior of materials with a stimulation to produce "bimetal" or bimetal effects.

- With charged materials, an electromagnetic field can induce a charge orientation leading to a desired deformation of the 3D object, etc.

I believe in evidence. I believe in observation, measurement, and reason when confirmed by independent observation. I can believe anything, no matter how strange and ridiculous, if there is proof. However, the stranger and more ridiculous the thing, the firmer and more solid the proof must be. (Asimov 1983)

Table 1.13 from the considerations presented in André (2017a) recalls the interests and limitations of homogeneous 4D printing. The knowledge of the synthetic results presented will then be used to consider prospective aspects regarding the scientific and technological developments of 4D printing.

While the offers of temporal evolutions are multiple, the following sections can, by the knowledge of the real and the possible, be the origin of certain difficulties, even frustrations because it is not always possible to satisfy the demand or the desire. This hiatus, which is quite general in this work, will be highlighted again with spectacular POC and applications that are not accessible (failure to meet "reasonable" industrial requirements). It is in this role of "shipwreckers" presented hereafter that we commit ourselves, defining by the conclusions to which it leads, a form of roadmap to follow if we do not wish to see a loss of interest from the socio-economy for this original technology.

4D printing	Comments		
Advantages	- Sequential 2D process using a localized deposition of active material on a surface followed by folding leading to spectacular effects		
	- Additive manufacturing process allowing an object to be manufactured <i>in a single step</i> and then to have more or less homogeneous actuation modes		
	- Use of external stimulation modes to change the shape (see Figure 1.4) and/or the functionality of the object		
	- Large number of stimulation modes (mechanical, heating, humidity, pH, light and other electromagnetic fields)		
	- Large application domains (see Figure 1.5 for the use of 4D printing in terms of functionality)		
	- Obvious spectacular aspect		
	- Limited choice of active materials, with very high deformation coefficients, mainly polymers that must be sufficiently elastic to allow deformation and, consequently, with modest mechanical properties		
	- Difficulty in locating the stimulation on the object		
	- Anisotropy of the stimulation		
Limitations	- Size of the stimulation device required to actuate the 4D object		
Linitations	- Long response time after stimulation, incompatible with industrial uses		
	- Stimulation energy that can damage the object		
	- Anisotropy related to layer-by-layer fabrication mode		
	- Durability		
	- Modeling of volume deformations is difficult		

## Table 1.13. Current advantages and limitations of 4D printing

# 1.4.6. Some constraints

Science discovers these laws of power, of motion, of transformation: industry applies them to the raw method, which the Earth provides us with in abundance, but which acquires its value only through knowledge. (Priestley 2015)

It is clear that the range of deformable systems is wide, without a specific path being favored today. In fact, reference is made to the POC in an emerging field which hides a certain number of scientific and technological problems to be solved. While the volume of the literature is rapidly increasing, the number of methods for deforming a macromolecule due to stimulation is limited to a few classical principles. Moreover, as the object is realized, the stimulus must reach the areas to be deformed, which for actuators may pose some space problems. This may be less constraining if the choice is turned to the use of light as a deformation stimulator (André 2018, 2018a; Li et al. 2019), as it is possible to selectively excite certain areas of the 3D object with great precision and distance. However, several problems, not mentioned in most of the literature on the topic, have emerged (and are mostly valid for other stimulation modes) (see André (2020a)).

### 1.4.6.1. Mechanical strength (E: Young's modulus)

In order to make an active object out of a single material, it is necessary that the material has a certain mechanical strength and also elastomeric properties to achieve a shape change. With Young's modulus of a few tens of MPa, classical in additive manufacturing, the forces necessary for a deformation of 1 mm of a bar of 1 cm, 0.1 mm thick and 1 mm wide are of the order of 2.10<sup>-4</sup> N. Conventional polymer materials are therefore not suitable for deformation to realize an efficient actuator. Moreover, in the literature deformations are just shown, without too much mechanical or energetic potential associated with them (see, for example, Listek (2019)).

*Young's modulus*: Young's modulus, modulus of elasticity or tensile modulus is the constant that relates the tensile stress and the onset of deformation of an isotropic elastic material (linearity). Figure 1.82 (see: https://www.xr6805.fr/school/sti\_web/rdm/deforme/deforme.htm) represents a beam resting on two supports with a concentrated load in the middle (see Thorin and Forêt (2013)).



Figure 1.82. Deformation of a beam as a function of its Young's modulus

With *F* as the force exerted at the center, *L* as the length of the beam and *E* as the Young's modulus, the deflection  $\Delta y$  is expressed by:  $\Delta y = \frac{FL^3}{48E}$ . With conventional elastomers, a deformation  $\Delta y$  of 0.1 cm for a beam of size 1 cm (thickness and width: 0.1 cm) corresponds to an approximate load of  $5 \times 10^{-3}$  N. As for metal, the corresponding load is approximately 100–500 N.

# 1.4.6.2. Quantum efficiency

I am just having fun. I was determined to do physics for fun, as I liked to do it. (Feynman 2000)

If we want to cause the rotation of a carbon-carbon or nitrogen-nitrogen double bond (case of compounds of the azobenzene family), it is necessary to absorb a photon of suitable energy (energies of the order of 400 kJ per mole or about 1 Megajoule/kg). But this process occurs with a quantum yield sometimes much lower than the unit. Regardless of the possibility of absorption on small elements (Beer-Lambert absorption law for a one-photon process), it is necessary that a large number of bonds are activated, which with a quantum yield of 0.5, for example, can lead to a very significant rise in local temperature. With a heat capacity of 2 J/g/°C, in the absence of energy transfer (conduction, convection, radiation), a temperature of 250°C can be reached! This critical situation raises the question of the choice of the light source's power, because the time associated with heat transfers can be a parameter to be considered.



with light stimulation

With one-photon light absorptions, the absorbed intensity follows a law of decreasing exponential in the simple cases, with more significant effects in the vicinity of the arrival of photons. It is then a complex nonlinear process whose

modeling of the deformations remains difficult. Moreover, in the chemical system generally retained (which must be reversible) the compound Cis produced also absorbs (but less than the most stable compound, which is the Trans (see Figure 1.83)). It is the Trans-Cis passage which induces the deformation which is reversible by return of the Cis to the Trans, but the sharing of the incident photons is a factor of thermal rise of the medium. In reality, it is necessary to place ourselves in conditions where the thermal aspects can be neglected (small size actuators).

# 1.4.6.3. Heat transfer (Fourier law)

The event is inseparable from the options to which it has given rise; it is that place constituted by often surprising choices that have modified customary distributions, groups, collectives, parties and communities, according to an unexpected division. (de Certeau 1994)

The materials selected in the literature are essentially polymers with low thermal conductivity. If heat is used to induce physical transformations of the object, the temperature must change in the mass of the material (which explains the choice of a stimulation by light). From an applicative point of view, the average characteristic time of heat transfer  $\tau$ , inside the material assumed to be homogeneous, for a given distance *d* and a diffusion coefficient  $D_t$ , is defined by the following approximate law:

$$d = \sqrt{D_t \tau}$$

So with  $D_t = 10^{-3}$  cm/s<sup>2</sup> for d = 1 cm, a heat transfer time of about 15 minutes!

Fourier's law is a phenomenological law similar to Fick's law for the diffusion of matter or Ohm's law for electrical conduction. These three laws can be interpreted in the same way: the inhomogeneity of an intensive parameter (temperature, number of particles per unit volume, electric potential) causes a transport phenomenon that tends to fill the imbalance (thermal flow, diffusion current, electric current).

#### Box 1.13. Fourier's law

Lantada (2017) conducted an analysis of the response times of different active systems. Figure 1.84 from his synthesis confirms the considerations presented above. Given heat transfer coefficients about 100 times higher than for material transfers, strongly amplified effects are expected.



**Figure 1.84.** Force-time response relationships in actuators: 1: magneto-resistive system; 2: biological muscles; 3: high-performance EAPs; 4: McKibben-type muscles; 5: SMP structures; 6: ceramic and polymer electroactive multilayers; 7: electroactive polymers; 8: hydroactive polymers; 9: shape memory foams; 10: SMPs; 11: ditto with fillers; 12: SMP + McKibben; 13: bi-metal; 14: tri-metal; 15: SMA; 16: SMA + Peltier effect; 17: MEMS; 18: magnet-MEMS; 19: NEMS

# 1.4.6.4. Material transfer (Fick's law)

Material transfers are generally between 10 and 100 times slower than heat transfers. The possible effects will therefore be very slow. This is what happens, for example, when a deformation is induced by the pH, requiring the diffusion of H+ atoms in the solid material, or the transfer of a solvent inside a polymeric matrix. Therefore, with  $D_m = 10^{-6}$  cm<sup>2</sup>/s for d = 1 cm, a material transfer time is of the order of 10 days (but only 0.01 second for a distance of 1 µm). Apart from these "average time" aspects, which already illustrate a problem that is difficult to deal with, there are aspects related to the thicknesses crossed: for sensitive materials, the effect of humidity, for example, close to the arrival of water molecules, will start much earlier than for materials placed deeper. Moreover, delay phenomena can appear with active materials which have space-dependent material diffusion coefficients.

Moreover, over the course of time, micro-fractures can appear within the material(s), significantly modifying the transport processes or the diffusion coefficients.

Within these two time-consuming areas are internal elements that are largely related to materials. These consume internal energy or extract energy from stimuli to move/deform and generate forces for spatial change of passive structures. Non-equilibrium effects are related to creep as in linear polymers or filaments.

The interplay between activity and conformational degrees of freedom gives rise to new structural and dynamic features of individual polymers, as well as interacting assemblies. (Winkler and Gompper 2020)

The properties of active materials depend on the coupling of the stimulated process with the conformational degrees of freedom of macromolecular structures associated with the coupled presence of local steric and hydrodynamic interactions that result in "time-consuming" conformational and dynamic properties. These two antagonistic effects probably play a role in the response time of the active structures and in the maintenance in time of their mechanical actuation properties; for the moment, the literature is, on this problem, relatively silent. It would undoubtedly be useful to examine the importance of the flexible modes corresponding to conformational fluctuations in connection with intra-macromolecular interactions.

# 1.4.6.5. Manufacturing anisotropy

Disruption is not a fad, not a trend, it is our new paradigm. (Mallard 2018)

If an object is made by stereolithography (for the moment with 1 photon), the light arriving on the surface to be polymerized is more absorbed at the surface than at depth (André 2017). The part present toward the surface is thus more polymerized (or even rigidified) than the areas located deeper. Under these conditions, the elasticity of materials, active or not, depends on spatial parameters. If we try to use actuation modes exploiting light (case of azobenzene type compounds, for example), the photons can contribute simultaneously to a hardening of the resins and to the desired deformation. This situation is thus not, a priori, the most recommendable! In other modes (molten wire in particular), it is the anisotropy of the deposit that leads to the desired deformation effect.

It is therefore not possible to separate the manufacturing process from the desired 4D effect. Moreover, these examples show the difficulty of a good programming of the relations amplitude of the stimulation – an effect on the changes of form and/or the functionality.

#### 1.4.6.6. Other problems

One must be willing to scuttle in order to comply with the requirement imposed by the disruptors [...]. But to scuttle, you need a vision. (Mallard 2018)

Not mentioned here are other problems that would have to be considered: that of the mathematical control of deformations (considered for massive materials as very difficult by Weeger et al. (2019) and Ding et al. (2019)), of the optimal design, nor of the response time and local amplitudes of the stimulations in the active material, nor of the footprint of the stimulus generating system. Figure 1.80 illustrates one of the difficulties of exploiting 4D printing: knowing the properties of the materials under stimulation, how and where to position the active voxels in an inert supporting material, how to calculate the local flows in time and space of energetic stimulation (advantageously using light?) for a 3D object to move from one shape to another, from one functionality to another? This work is beginning to be approached in simple model situations. At best, it only concerns grid structures, as shown in Figure 1.85.



**Figure 1.85.** The issue of modeling shape change by stimulation: the "possible" case of a structure composed of segments

These elements define real limits to the development of 4D printing. For example, for low response times, it would be necessary to produce very small objects, or perhaps to use optical (stereolithography technique) or electromagnetic (wire fusion process) stimulations, for which we know how to have time bases adapted to the demand. In any case, these technological constraints are areas of research to be conducted to push 4D printing toward an industrial technology.

#### 1.5. And tomorrow?

Scientists founded, beyond the production of knowledge, systems of action that influence nature and society, systems of value that inspire norms and ideals, systems of representation on which discourses and postures of authority are based. (Rasmussen 2015)

The most common liquid crystals are formed by amphiphilic molecules. The specific forces involved in these crystals are thought by NAP (2019b) to be atypical (Davidson et al. 2015; Zhou 2017). This results in unusual conformations and functionalities (Jeong et al. 2015). For example, the ferro-nematic liquid crystal phase (a liquid exhibiting long-range ferromagnetic order coexisting with nematic order) was created by dispersing ferromagnetic nano-disks with magnetic moments normal to their flat planes in a conventional nematic. After further treatments, the magnetic excitations are overdamped and would not exhibit the dispersion observed in the traditional way. It is with such modes of action that new ways of using liquid crystals have opened up, in particular for chemical and gas sensors. These advances are based on the disruption of liquid crystal orientations at the liquid crystal/solid or liquid crystal/air interface, caused by the product to be measured. If this type of behavior has not, to the authors' knowledge, been directly exploited for 4D printing, it is not the case for elastomeric crystals.

In another domain, granular materials are aggregates of individually solid grains that interact with their neighbors via contact forces. Their properties do not depend significantly on temperature and they do not explore the many internal configurations of systems in thermal equilibrium (they are non-ergodic). One of the most remarkable properties of granular materials is their ability to change from a fluid to a solid state by simply applying pressure or, equivalently, by increasing the volume fraction of the constituent particles. Moreover, according to their radial distribution function (for mono-dispersed hard spheres) the space can be filled with a variable number of spheres, as illustrated in Figure 1.86 (in statistical mechanics, the radial distribution function, classically noted g(r), describes, in a particle system, how the density varies as a function of the distance from a reference particle). In fact, between materials and their forms, there are, independently of the modes of stimulation, an infinite number of possibilities. Let us recall Friedman (2018) who wrote: "I do not know reality, but it seems to me that it cannot be faced otherwise than by the image". 4D printing makes us see, so it is an excellent laboratory to try.

NAP (2019b) shows that non-spherical and same-sized particles with more exotic shapes can jam under their own weight. This area falls under random architecture (Keller and Jaeger 2016), but, to the authors' knowledge, has not yet been applied in 4D printing.



Figure 1.86. Density related to regular stacks of spheres (according to http://villemin.gerard.free.fr/Wwwgvmm/Geometri/SpheEmpi.htm)

Thus, regardless of the form (e.g. metamaterials) the exploration of systems that can evolve under the influence of stimulation is probably not complete. It would be interesting to examine whether transitions linked to regular voxel stacks (heterogeneous or hybrid 4D) can be at the origin of original 4D printing, provided that we have first solved the questions of printability.

How do we know that we will find the right threesomes between materials, local and global forms (3D printing) and stimuli that meet the specifications in this almost infinite (and largely unexplored) world? Will we have to develop anticipatory thinking on new possibilities of action, using a complex analysis of weak signals? Or will we have to work with trial and error methods? Perhaps the future will tell us.

According to Schoemaker et al. (2012): a weak signal is "isolated information that, at first glance, is akin to background noise, but when studied through another analytical framework, or linked to other information, can ultimately be understood as part of an important informational pattern for the organization".

### Box 1.14. Weak signals

[Universities] do the basic research that large companies no longer do and assume the risks and part of the cost of that work. (Lécuyer 2015)

Engineering is also, in its own way, about working your way through the space of all possible solutions to a problem, climbing from solution to solution, each better than the last, until you reach the optimal solution. (Benyus 2017) Science is in fact entering a transitional phase. It was historically anchored in Cartesian principles of duality and reductionism defined by empirical facts that can be apprehended by the senses. (Vinardi 1980)

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2

## Energy Stimulation: The Abandoned Child?

The Scientific Revolution was not a revolution of knowledge but primarily a revolution of ignorance. (Harari 2015)

We can be ignorant of a great many things without it affecting our daily lives. Still, it is undeniable that low literacy decreases our understanding of our environment especially when coupled with meta-ignorance (ignorance of one's ignorance). (Larivée and Sénéchal 2019)

### 2.1. Introduction

The introduction of additive manufacturing from a privileged relationship between digital and physical realization is starting to transform the manufacturing industry. This attractive technology still has some drawbacks such as printing speed, surface finish of the final product and lack of fully functional materials. 4D printing is the additive manufacturing of active or smart materials with sensitivity to energy stimuli (e.g. moisture, pH, heat, light, solvent, electric or magnetic field). The previous chapters have focused on material–stimulus relationships, additive manufacturing strategies and the time required for shape- and/or property change to occur.

Thus, from a perceptive system to a stimulation associated with a more or less complex structure, a 4D object is able to make the equivalent of a decision (form and/or functionality) which may be revisited in future energetic interactions depending on the effects produced. This last learning mechanism has been considered: the contingencies at time t are then reversed at time t + 1 and require

re-evaluation and new encoding. It is in this context that the notion of a "perceptual" system partially decoupled from a spatio-temporal evolutionary system could be considered.

Since the beginning of this book, it has been said that 4D printing has three roots: additive manufacturing (active and passive) materials and their stimulations. Significantly, these first two areas have been the subject of further study. Figure 2.1 of this short chapter brings together the essence of the knowledge asserted about the different stimulations (Shakibania et al. 2021).



Figure 2.1. Classical stimulation methods applied to 4D printing

We have shown that other energy inputs can also be used to achieve objects with shape-changing capacities. In particular, the spatial localization of active voxels has been discussed, while the same has hardly been done for the energy inputs at the origin of the 4D effect! One of the major challenges today in the 4D printing field is being able to explain how, from a set of voxels encoding or representing certain properties (shape and functionality), we obtain:

- A resultant as decided by the designer, transient or stable for a certain time for a fine internal structure under stimulation.

- How this resultant can modify the activity of the dynamic system (fatigue, degradation, feedback, learning, etc.)?

- Among the continuous flow of stimuli (of variable amplitudes and natures) that reach the 4D printed object and that influence the active voxels, first of all it is important to detect and extract the value of an evolution linked to each relevant stimulus (linearity of the effects? nonlinearities?) on each active element, as well as to evaluate the potential consequences that it can generate for the object taken as a whole. - How do these stimuli reach the deeper or shallower areas of the object? How, when its shape or functionality changes, does the local amplitude of stimulation change?

- Can we robustly find an analogy between spatially distributed active voxels and equally distributed energy sources?

– With 4D printing, we use materials that may be out of thermodynamic equilibrium with scientific knowledge that is still fragmentary (bi-stability, quenching, polymer creep, etc.). Thermodynamics and statistical mechanics provide rules for averaging the well-known dynamics of classical or quantum states to obtain the macroscopic properties of 4D materials and systems in thermal equilibrium with their surrounding environments. But, according to The National Academies of Sciences, Engineering and Medicine (2019), the universal guiding principles are not stabilized. This report concludes: "Nowhere is the need for a deeper understanding of non-equilibrium phenomena greater than in materials science". Yet in this field, temporal aspects are important and should be studied in depth.

- What are the interdependencies between active and support zones?

- A degree of complexity to which the active system is subjected according to the energy inputs distributed in time and space with the question of whether we are able to reconstruct the stimuli differently for the same desired 4D effect (effect of pattern recognition using artificial intelligence for example).

– Etc.

It is true that the bibliography is remarkably poor in this field and one would have appreciated works using spatial and temporal resolutions of 4D deformations with a single energy source or several, which can have many forms. Ignorance, according to Larivée and Sénéchal (2019), gives those who know how to take advantage of it – including politicians who are certainly among the most immediate beneficiaries – power over the population (Firestein 2012; Girel 2013). Processes can be taken advantage of by promoters of false beliefs, but in the domain of 4D printing, it is difficult to find such a manipulator (and, in particular, their motivation(s))!

Kruger and Dunning (1999) have indeed shown that the less competent we are in a domain, the more we tend to overestimate our competence, as if this made us incapable of imagining what precisely we do not know. On the other hand, highly competent individuals tend to underestimate themselves, thinking that if a task is easy for them, it must also be easy for others. Perhaps this is the direction of this scientific and technical weakness that can only be detrimental to this technology!

#### 2.2. To go a little further

The process of knowledge creation can be represented as an unstable non-linear dynamic system, not predictable in practice in the long term and based on the Second Principle of Thermodynamics. The induced chaotic reasoning sees as the starting point of creativity a dissatisfaction, a tension between the desire to evolve toward a higher form of organization (negentropy) and an observation of evolution towards a lower form of organization (entropy). (Saulais and Ermine 2016)

Figure 2.2 from the Royal Academy of Engineering (2021) recalls the set of interacting elements that should be taken into consideration to move from the research stage to societal utility applications.



Figure 2.2. Interaction activities to be carried out after the R&D stages

These action lines are what companies do to move from mock-ups or prototypes to commercial applications. They cover the risks involved in successfully completing the testing, demonstration and manufacturing of new products, services and processes in order to achieve commercial use, generate value, both socially and economically, and remain competitive. But, by "forgetting" a part of the research activity on the most appropriate stimulation processes, one increases from the beginning of an action, risk taking, to an industrial voluntarist action.

On the other hand, the focus on incremental innovation is based on a demand for significant improvement. The stimulus-action framework in 4D printing is not a

simple change and probably requires creativity. The latter aims to "modify knowledge by building on it, reworking it, recombining it and giving it other meanings and values" (Saulais and Ermine 2016). These authors recall that classical creativity methods are based on a principle of "divergence-convergence" subjectivity, (divergence: analogy, imagination, etc., and convergence: transformation of ideas into solutions). They assume the initial existence of a question. Once considered we can expect some acceleration in innovation processes, especially if they require some funding, but "also, privileges and positions, status and recognition, friends and lovers, etc. and [after] this allocation takes the form of a competition" (Lemire 2015).

For their part, Lin et al. (2021) have shown the influence of citations on the short-term development of a scientific theme (not the long-term). In the absence of anticipation, they show that citation dynamics remain directly related to productivity on the topic under consideration (going to the rescue of victory?) (see Wang et al. (2013)). In science and technology, transformative ideas diffuse slowly at first, but accelerate after a tipping point. Lin et al. (2021) call this the "sleeping beauty" phenomenon (see Ke et al. (2015)). In technology, this is referred to as the "J-curve", which suggests that disruptive technologies take time to emerge because they require and develop complementary technologies. Once a supportive environment develops, they see an increase in productivity (Brynjolfsson et al. 2018).

Focusing on the technoscience of the future, the slow accumulation of innovative and disruptive research contributions reveals an enduring resistance to radically new ideas (which sometimes struggle to get published). This underscores the history of how many breakthroughs in recent science were initially rejected or ignored, sometimes for decades (André 2015, 2016; Lin et al. 2021).

Considering these cases, successful in retrospect, some may question whether it is possible to formulate science policies that accelerate the exploration, dissemination, and application of transformative scientific ideas. We argue that designing, testing, and implementing metrics that allow us to quantify and evaluate new failures along the long path to transformational success will reduce the tyranny of short-term rewards that have unwittingly encouraged narrow, incremental, and redundant research. (Lin et al. 2021)

If 4D printing is ever to be moved out of its role as a spectacular academic toy and into profitable application areas, this simple problem (but one whose answer is certainly not) will have to be addressed. Where, when, and how do I transmit a particular form of energy to optimally achieve a 4D effect? Once again, it is a "reverse problem" type of treatment that must be engaged and, as has already been pointed out, it is outside the logic of academic offerings and certain practices. But, this question alone is a huge thorn in our sides!

Divergent and convergent reasoning: Guilford's distinction between convergent (conventional) and divergent (unconventional) thinking has influenced cognitive research on creativity; the former is oriented toward producing the single, best, or correct answer to a given question, while referring to information already available; the latter involves producing new, multiple, and different answers from the data provided. (Masmoudi 2010)

I [...] followed a golden rule, namely that whenever published fact, a new observation of thought came across me, which was opposed to my general results, to make a memorandum of it without fail and at once; for I had found by experience that such facts and thoughts were far more apt to escape from the memory than favourable ones. (Darwin 2002)

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# Material-Process Duality in Industrial 4D Printing

We are in a world of "Sunday drivers", as George Friedmann writes, "of Men who have never looked at their engine, and for whom things have not only but for mystery to function". (Baudrillard 2001)

The famous pure/applied dichotomy has an assignable origin: it dates from the 1750s and was invented by a Swedish chemist, Johan Gottschalk Wallerius (1709-1785). [...] The notions of pure and applied allow for a strategic reversal [...] in favor of a logical dependence of the arts on science. [...] The purity of science is a purely ideological notion. (Bensaude-Vincent 2009)

#### 3.1. Introduction

Henry Le Chatelier (1850–1936) wanted to organize scientific research on the model of an industrial enterprise (Le Chatelier 1925). He envisaged a cost/benefit evaluation of scientific production according to the innovations that it could allow for the national industry, with essentially a certain obligation of results (this is not exactly what the authors have observed in their present analysis of publications concerning 4D printing). It is against this model that Jean Perrin elaborated the concept of "pure science" because science must remain a creative, disinterested activity, which has its own internal purpose. It is on this basis that the CNRS was created in France before the beginning of the Second World War. It is not a question here of returning to debates on "research that finds" but rather of examining how one can go from an idea (supposedly favorable) to an application (which must be

4D Printing 2: Between Science and Technology,

For a color version of all the figures in this chapter, see: www.iste.co.uk/demoly/4Dprinting2.zip.

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just as favorable). However, in all that has been shown in this book, it is the material-process duality that we wish to discuss further in this chapter. This chapter could have been entitled: "From matter to form; from its form to its function" or the opposite, hence this less catchy title. Indeed, it is not because we can observe spectacular physical phenomena with attractive proofs-of-concept that we have finished our work; on the contrary, other rules are to be followed with their potential rejections.

A common issue in product design and object creation concerns the transmission and physical translation of an idea. This aspect associated with creativity has been widely discussed in the different chapters of this book. This question is crucial because an idea, generally individual, must be shared by several people, in and by different professions, between the demonstration (proof-of-concept) and the industrial development which will use its own design methods.

The context of simultaneous/collaborative engineering and project platforms encourages the meeting of different design actors who never speak the same language, and who use similar words with different meanings depending on the job and discipline. This is one of the first paradoxes of simultaneous engineering, Babel's paradox [it represents a risk of project stalemate, analogous to the causes of the stoppage of the tower of the same name], born from a partial response to a complex problem. It stems from design projects that, even if simple, need to be studied in their diverse attributes through an interdisciplinary approach. (Bassereau et al. 2015)

These authors remind us that the final object intended for users must go through intermediate trial and error processes (see also Leroi-Gourhan (1950)). This observation, which is almost obvious to the human involved in the art, makes the hypothesis of time-consuming practical consequences, which is indispensable for successful industrialization (even with growth in terms of publications of more than 40% per year). These are indeed intentional representations, by design, of situations in which actors intervene on the classical register: seeking to model in order to understand and to understand in order to do (Le Moigne 2013). However, everyone knows that it is not enough to identify the disciplines and businesses necessary for design in order to put them to work (Minel 2003; Carlile 2004; Hatchuel 2006). This problem will be discussed in more detail in the next chapter.

#### 3.2. From research to innovation

The world is not vicious as tortured chastity imagines. The world is at risk. It is enough to make the wise ones whose morality is that of
bursts of laughter, but if they do not risk anything themselves, they survive on the risk of others... The world is at risk. The world will be tomorrow who will risk the most, will take his risk more firmly. (Bernanos 1947)

We talk a lot about understanding each other and communicating [...]. We believe that there are great secrets to be discovered. We hope that Science will show us the way and, if not, religion. We dream of a life in the distant future that will be radically different from the one we know today; we attribute to ourselves unspeakable powers... (Miller 1957)

An inherent dynamic of research (interdisciplinary or not) is to let oneself be carried away by one's curiosity or to engage in spontaneous or stimulated requests toward new questions or new objects. This is all the more true in science, where innovation should be a major criterion of success. This process sometimes leads to objects or questions that go beyond those defined by the cognitive and institutional structures that organize research, while at the same time imposing continuities, that is to say disciplines. The previous chapters have shown a great richness (certainly not totally stabilized) in the more divergent scientific works concerning the field of 4D printing, in particular at the stage of academic research. They try to give reason to Miller, but for all that, the essential remains to be done before reaching the final user.

## 3.2.1. Research

Imagination consists of representing absent objects and combining images of these objects with each other. It virtually brings new possibilities into existence. In this sense, it frees itself from the frameworks and leads to seeing the world from another angle and to leaving the dominant logic. It allows us to experiment with many possibilities in our minds to come up with new ideas. (Parmentier 2020)

According to the classical vision of creativity (put forward by Guilford (1950, 1956, 1959, 1967)) on the dichotomous principle of divergence/convergence, the creative process begins with the recognition or the making of a problem. From there, a process of divergence begins, and finally ends, by convergence, in a new solution of the problem (when it exists). So where does this new buzzword come in: disruption? It is the ability of the creative or divergent to bring together seemingly distinct worlds, to find a link where none existed before, to go and see what is happening outside a scientific discipline, a profession or a specialization, to be open to everything, without taboo. It is a thought of adventure. It invites, as in the case of

4D printing, to face the new and the unexpected. This relationship of transformation, made of tinkering, of adapting to circumstances and the unexpected, of forgetting pre-established models and of ingenuity, obliges us, in any case, to invent a specific mode of transmission, which is not of the order of the teaching of abstract knowledge. For all that, the achievement of an acceptable robustness of this technology is still to be sought.

If, for the authors, there is no question of leaving the link with disciplinary research, it is important to continue to set up techno-scientific research operations that can be used for direct or indirect application, for a time, thanks to the specificity and genericity of the themes and projects developed and open to the future needs of society, enabled by science. This proposal should avoid the significant remark of Janicaud (2002) who wrote:

Technicism is a [...] utopia, apparently neutral, but in the long run even more dangerous perhaps, because it never ceases to renew its attractions and to disguise its stakes under immediate satisfactions whose real consequences are masked.

This opinion is supported by Stiegler (2008) who considers that:

the problem of contemporary science [...] is to know if science has become a pure efficient cause that produces possibilities within which an extrascientific causality, and therefore extra-rational, should be able to choose. In fact, today, such an extra-rational criteriology is nothing other than the market, that is to say more exactly marketing.

Engineering sciences are defined as sciences at least partially distinct from traditional disciplines. Their objective is to innovate, to study and understand the new properties of artifacts designed and therefore built by/for humans with the aim of benefiting society (acting to understand and understanding to act). In the 1990s, at the CNRS, they became the science of artifacts, the science of objects and systems where knowledge of Nature and intervention of human genius are combined in order to solve, by abstract or concrete means, problems born indirectly and remotely, of functional concerns.

More and more, artifacts or objects created for humans spread throughout society – at a speed never seen before – leading us to re-examine the cultural foundations defining "relevance" and "performance", even the legitimacy of technosciences and their applications.

In fact, the sciences of "artifacts" (artifact means "made of art", implied to be human art according to Micaëlli and Forest (2003), any entity, tangible or not, conceived to answer needs) that are attached to what could be an object (Murray 1923; Simon 1973; Demailly 1994) or engineering sciences constitute "a space between the natural and the cultural that we don't know how to attribute to one or other of these fundamental orders. The in-between of the artificial and the cultural evokes the illegitimacy of the incest" (Laufer and Paradeise 1982). According to Vinck (1995), the artifact emerges in the course of the action as a contingent intrusion, even if a supported imaginary may be at the origin of the research activity. It is not necessarily a question of going along with Habermas (1968) who wrote: "Technoscience has become a social subsystem where rationality dominates. This subsystem has progressively colonized the rest of the social structure, soon confusing rational hierarchies with the social hierarchy."

Either the researcher treats the object in a partial way, ignoring or concealing all the links that inevitably exist between this partial view and the global biases that he cannot fail to have on the object. Or else, he puts his global biases at the heart of scientific questioning: not as interesting elements to look at, but as the driving forces of a dynamic that then takes the inevitable desire for unity as a stepping stone towards what becomes a requirement for unity. (Alvarez-Pereyre 2003)

This desire to get even closer to real use, with all its difficulties (Nicolescu 2002), is based on a cultural background, that of modeling and integration (systemic approach), that of experimentation (technology centers, e.g.) as well as an indispensable opening to other scientific and technical fields, by "borrowing" their concepts, their methodologies (principles of permanent learning and diversity), their instruments and by "bringing" subjects of study to them. All disciplines can be concerned, from the "hard" sciences to the human and social sciences.

So, a concrete technical object is a system in which interdependencies are exercised according to all the laws of science (if necessary). But, without this being easily perceptible, the artifacts, by feedback, act on the practices and on the culture of the society as a whole to such an extent that Luis de Miranda (2010) writes: "By acting on the matter, Man acts simultaneously on himself. This makes us not only external users, but theoretically responsible creators, insofar as technology is a part of ourselves." In this recursive framework, Attali (2007) assigns to the world in the making a certain number of missions; he writes:

Freedom in the sense of less pain and less effort, more free time, despite the eminently illusory character of this freedom "enclosed between four walls which are those of death and birth". This freedom,

a desire to live better within these four walls, a will to remove these walls by making life longer and better, and then a will to pass on to those who will later live in this same room better living conditions.

This ethics on how to "live well together" in the sense of Ricoeur (1990) ("aiming at a good life, with and for others in just institutions") leads the technosciences to help produce innumerable goods for technical progress. In fact, it appears today that our technological know-how increasingly exceeds human knowledge, disturbed by the very fast dynamics of change. In any case, Puech (2008) writes that "technique is neutral only if it is considered as a means only, the difficulty being in the 'only". It cannot indeed be considered as neutral by the instigation which is associated with the production of new consumer goods, and as Ellul (1954) points out because of the "principle of irreversibility of the effectiveness".

As we have shown, the emerging 4D communities where a common disciplinary passion is shared are innovative, but tend to close in on themselves. We need to find ways to develop interdisciplinary, heuristic, temporary and open spaces in research, allowing a more collective functioning of the creation and exploitation of new concepts. In this way, discontinuities would emerge, leading to scientific recompositions and adaptations. This is why it will be necessary to account for the way in which 4D-printed objects return to the social world. This is a central question that strongly links research to the economic, the social and the political, and puts 4D printing in direct link with society in a responsible way. In a few sentences, these are questions that could be debated, but which still await robust answers. Basically, paraphrasing Henri Guaino (cited in Salmon (2017)), can we not write that 4D printing is preparing to write a history shared by those who make it and those for whom it will be intended? One does not transform a context without being able to write and tell stories. It is on this basis that it is possible to build forms of collective imagination that allow for action.

# 3.2.2. Innovation

Everything simple is false, everything complex is unusable. (Valery 1991)

As reason makes no demands contrary to nature, it demands that every man should love himself, should seek that which is useful to him – I mean, that which is really useful to him, should desire everything which really brings man to greater perfection, and should, each for himself, endeavour as far as he can to preserve his own being. (Spinoza 2005) For Durand (2004):

corporate strategy [...] attempts to resolve the paradox posed by the relationship between freedom of creation and initiative and the influence of economic selection. It thus consists in shifting, through particular choices, the selective constraint on competitors thanks to a control of master assets, resources, and strategic aptitudes. As such, companies do not innovate only to satisfy their customers, but to impose pressure on their rivals and push them to the fault or to abandon.

This situation, in which the dynamic image of the company is important, makes it possible to reinforce, in principle, the links between production activity and techno-scientific research (provided that it has been correctly anticipated). Indeed, "every organization is an immune system whose goal, like any organism, is to preserve a certain status quo. But, for decision-makers, the adaptive value of change has become greater than that of stability" (Blanquart et al. 2006). Thus, at the same time, the company is becoming more international: exchanges, personnel, legislation, ISO standardization, etc., and is largely modifying its social organization: subcontracting, specialization, individualization of performance, aging, etc., in a context of rather lifeless access to work. However, several phenomena (at least) are at work:

- The gigantism of industrial research centers and design offices leading, as in academic research, to a subdivision of activities. Gaudin (1978) writes that "the object is cut up into pieces, and this division alone guides production towards reproduction"; in fact, out of "fearful conformism" (Gaudin, 1978), one also looks where one has already found.

- Research is rarely the source of innovation; it is the "result of heterogeneous and improbable initiatives" (Gaudin 1978).

- Innovation depends on specific events, results of history, leading to an absence of precise determinism and predictability over the long term (butterfly wing stereotype) (Bak 1996).

- The technological systems we use are not isolated from each other; they refer to each other (Puech 2008).

- The development of technological artifacts is regulated by their uses (appropriation); Puech (2008) speaks of the adaptation of technology to use and the adaptation of citizens to technology (idea of coevolution).

- The high cost of investment in new installations is likely to be an obstacle to innovation; "capitalism itself organizes its regulation of technology by choosing

what should be developed and by favoring profitable investments. Capitalism moderates the insatiability of technology by its own [financial] insatiability" (Chiapello 2007).

- Production managers are not interested in research whose effects are envisaged in the medium term, whereas they are asked to produce financial results in the very short term. This phenomenon is amplified by the short duration (a few years) of these managers on the same site. We leave it to the next person to enter a risky universe.

Matter reveals the mind that needs matter to be revealed, mind reveals matter that needs mind to be revealed. (Vergely 2017)

However, in spite of these reservations, the technological movement, as a means to an attractive financial result, must be a creator of new wealth accessible by the citizen. At the same time, the citizen is asking for more regulation of technical change through regulation, with legislation aiming globally at their protection as a consumer. Moreover, does Kaufmann (2008) not remind us that "the less mandatory the standards, the more people's main activity is to produce new ones, to the point of obsession with normality"? But, as Landes (1975) expresses it:

the industrialization of the world continues for better or for worse, and if there are people in advanced countries to whom this worship of material achievements makes them gag, it is because they can afford a critical attitude, while the vast majority of the planet's inhabitants dream of accessing this material ease.

In this dual context, technology has become both a guarantee of material prosperity and of constantly renewed social, political and moral concerns. There is thus, in fact, a demand for a social evaluation of technological developments, associated with a certain will to control technical and organizational changes. Recent mediatizations increasingly imply the consent of the actors of the transformation of the society to a public debate (see: setting up in France of the national commission of public debate (commission nationale du débat public) on September 4, 1997). Under these conditions, society is condemned to adapt to change by striving to bridge the gap between the complex and ambivalent aspirations of the social body and the estimated effects of the transformations induced by the innovations. In principle, there should be a debate between the two technocratic and democratic points of view, aiming at a consensus defined in terms of control, protection, regulation, etc. This debate, however, remains open but provisional, because a pending question, widely put forward in the cases of asbestos and mad cow disease, concerns the anticipation of the long-term consequences of technical change. Indeed, how can we anticipate the perverse, unexpected and unintended effects, in order to

retain only the positive consequences of industrial transformations? It is in essence an attempt to resolve the dilemma between attractiveness and the desire to possess on the one hand, and risks for others and especially for oneself on the other hand.

Another way of thinking exists, centered on innovation, which is "innocent" only in the eyes of the engineer and the researcher in technosciences who produce it or the economist who accounts for it (Salomon 1992). What can we say then for the researcher if we follow the writings of Zweiacker (2007):

In itself, the scientific method does not include criteria for selfregulation, and this is indeed the case: the erroneous idea that the researcher should impose an ethics on himself is tantamount to leaving it to the scientists alone to set their limits; whereas in reality, it is up to society as a whole to have such prerogatives!

In fact, and this is what this book is all about, innovation disturbs the person who undergoes it (even if they have somewhat wished it), upsetting the social organization, perhaps much more than just a factor of financial gain. Where is the real challenge of change? To what does the competition between human and machine lead? (Boy 1999). The contemporary critique of technical progress can thus be expressed by the following elements:

- process of the machine de-skilling skills, but likely to save human labor;

- traumatic uprooting (rural to urban, migrants);

- scientific organization of work (time management);

- scientific progress that can be experienced by some as a perversion of the status of the human in nature;

- the machine is the means of work that threatens work, that enslaves the employee to its rhythm;

- automation creates unemployment; it can lead to wage cuts and desocialization in society;

- the over-concentration of the sources of transformation and globalization create new risks, etc.

First of all, for more than 150 years, we have seen the transition from an agricultural society to an industrial society and now to a service or knowledge society. In 2010, in the West, 80% of work concerned this field (Jeanneney and Barbier-Jeanneney 1985). In this transformation of the world, the share of manufacturing in the value of the final product is constantly declining (45% in 1960, less than 20% today, how much tomorrow?) The decline is partly linked to the relocation, when advantageous, of a certain number of "labor" companies and to the

development of automation and mass production processes. This context highlights the need for reflection on the act of designing manufacturing processes for new products and materials, and also for a thorough examination of how to connect the product to the future consumer. The association between "on the spot" production and service creates a new logic called servo-industrial. It must enable the reinjection of pleasure into purchasing practices, either through novelty or at a modest cost, thanks to the proposal of a mass production of products adapted to demand (almost tailor-made), with additional choices of services (credit, e.g.). It must also increasingly produce usage (we no longer only want an object but also its maintenance...). Thus, the main concerns of companies require:

- better identification, anticipation and direct demand through innovation;
- better understanding and mastering of the phenomena related to matter;
- bringing new features to promote real or fake attractiveness on new artifacts;
- managing technological, economic, human complexity, etc.;
- improving the productivity of intangible investment;
- maintaining their competitiveness.

In these conditions, the industrialist builds the "business" of their company with the help of a plurality of specialized individual businesses, aiming at "customer" satisfaction through an integrated or collective organizational approach. To achieve this, work organizations, which are becoming increasingly complex, are highly dependent on the quality of the links that exist within the company. Such an observation highlights the fact that education alone through deductive methods is not enough to give training that is totally professionally valuable, as Blang (1982) points out: "most employers, whether in the public or private sector, are less interested in what potential workers know than in how they will behave." How does the researcher, enrolled in their scientific quest, position themselves in relation to a company that considers technological research not as a goal, but as one of the means of its performance, with different missions and methodologies (Maret and Pinion 1997)? Moreover, do they think that the search for the functionality of an artifact is oriented toward an applied goal or more cynically toward an order and integration into a whole? For the object, it is, according to Baudrillard (1968), "the possibility to exceed precisely its function towards a second function, to become an element of play, of combination, of calculation in a universal system of signs".

If we can consider that there is still a core of stable jobs, associated with the "heart" of the industry's competence, a certain number of more precarious jobs come to participate in the economic development at the same time as a certain number of jobs are outsourced (subsidiaries, subcontracting, consultancy, certain areas of research, etc.). The development of new information and communication technologies with all the sciences and digital technologies is likely, with the strengthening of the service activity, to amplify this trend. Where will be, what will be the heart of the company in the implementation of a network of actors too separated? What will be the socialization of such a system that will significantly disrupt all interactions? How will the trust between all the partners be defined? It is, in fact, possible to imagine a strong evolution targeting the pooling of knowledge through the interaction induced by this new know-how, associated with a redefinition of the status of operators, with a search for elitist optimization of the cost/benefit ratio in the case of services. In André (2017a, 2017b, 2017c), the emergence of 3D technologies has resulted in a certain continuity of industrial production exploiting the new manufacturing methods on the one hand, and also in the emergence of FabLabs on the other hand. The rise of immaterial work is likely to lead to a certain deregulation and a more individualized wage relationship. If we look at the general innovation market, several principles are at work (Guzzetti 1998):

- A first element corresponds to the principle of assimilation based on the success of a new product, or identified as such (launch of new products in traditional markets, attacking new markets with confirmed products, just readapted). Success can lead to the stabilization of modes of thought and action.

- The second is linked to the principle of plurality, which assumes that a new element (product, process, etc.) can only be assimilated if it can be integrated into an existing group (concept of technological disruption).

- The principle of alignment stipulates that the technical qualifications induced by a technical or organizational change must not exceed (training, hiring, etc.) the maximum cost already accepted in the company in other situations.

- The principle of transferring to others aims at making the initial research and development effort profitable.

- The principle of expectation, linked to a response to demand without the company trying to propose an offer... In fact, the creative offer is often only understood because it results from a technological innovation.

These observations, already analyzed in part in other chapters, highlight the accelerated production of new products using well-mastered methodologies; under these conditions, there can be a connection between the temporalities of research and that of the company, provided that the scientific expertise of the laboratory is effectively present. However, it could be important at this stage to think about the training methods of people, and therefore of researchers, in order to build a facilitated interaction between science and business. With the advent of 4D printing, as with all other novelties, a pattern of "trust/defiance" is created and relies on the fact that social exchanges and their interactions are established in a determined psychological context, within a system that will impose sanctions in case of

breaking the agreement and specific supports in case of respecting it. It is naturally based on a form of knowledge. It is therefore at the origin of communities of practice (languages, routines, shared artifactual meanings) based on (or using in its own way) scientific knowledge and its experimental demonstrations.

At the same time, the development of well-being allowed by the production of goods authorized by new 4D technologies should continue and lead to irreversible consequences: can we live today without our cell phone? How can we stay young? How can we not live with our computer and its immense possibilities of connection with the world? How can we not remain indifferent to a personalized evolutionary environment? Thus, without realizing it, citizens are transforming themselves and could not survive if they had to live in 1900. As an example, Michalon (1992) writes: "Rural culture, all centered on an ethics of the relation was consciously immolated to the profit of a city culture, centered on the individual competition in the exercise of the functions leading to a society where the anonymity leaves to each one all its chances of blooming." How then can we return to the old ways of life? Is this desirable or desired? It is this transformation, with a weak or unperceived gradient, that should be analyzed in a logic of technological innovation; it is more and more associated with ethical reflections.

Ethics is by definition the reflection on the principles that allow us to live well together. Research ethics is the reflection on the principles that allow us to live well with the practice and the results of research. This is a very broad definition that often leads to the inclusion in research ethics of a large number of questions related to the impact of research on society. (Bordé 2008)

#### 3.2.3. Inclusion of 4D printing in future projects

Science advances only one burial at a time. (Planck, cited in Harari (2017))

I am amazed at how an immortal and elusive idea can act on matter. (Lallemand 1974)

In general, it is not only the field of additive manufacturing that is subject to new proposals. Everything is moving around us with psychological dynamics acting on the social matters at work, seeking a better control of the "de-collectivization" of the company and the development of individualism at all levels. In this context, it is undoubtedly still possible to consider this sentence by Huxley (2013) relevant: "Such is the goal of all conditioning: to make people like the social destination from which they cannot escape."

The propaganda of the commodity serves a double function: firstly, it affirms consumption as an alternative to protest and rebellion [...], secondly, the propaganda of the commodity, or of the consumption of the commodity, turns alienation itself into a commodity. It turns to the spiritual desolation of the modern world and proposes consumption as a remedy. (Lasch 2000)

This society is linked to the organizations of the production of material or nonmaterial goods allowing the search for a performance (at least financial for the capitalists) that exploits the possibilities of use of a set of scientific, organizational and technical knowledge represented by innovation in general. It is the same in another framework, but associated with this offer, that of uses (and their detour), at all levels. Thus, complexity explodes in a framework of dematerialization induced by the exploration of new information and communication technologies, a real space of freedom as well as control.

After having considered innovations resulting from the new possibilities offered by 4D printing, it seemed necessary to us to explore the importance of the current trends on the most adapted forms that innovations should take. But, at the upstream design stage, can/should the researcher develop their responsible "quest" alone or in consultation with "enlightened authorities" who define the framework, or finally with the society as a whole? It is therefore easy to imagine the difficult and risky path that the application of the scientific imagination can take (but which is beyond the scope of this reflection). This one must pass from the discipline to interdisciplinarity, to the integration with aspects of attractiveness, of supply, of questioning for the performance and to think of the possible risks as well as of the recycling, of the productions which it helps to make emerge. Is it not already at this stage a hopeless adventure or in any case strewn with (too many) pitfalls finally innumerable? Especially if one can be satisfied with good publications contextualized and expertly reviewed by peers.

Foresight techniques have been used in the case of emerging fields. Based on knowledge of the real and the possible, and with a little imagination, it is the subject of the synthesis presented in Table 3.1 (see André (2009), de Kerorguen and Leroy (2008), de Riedmatten (2005), Attali and de Boissieu (2007) and Lebeau (2005)). In this table, we looked for examples of questions that could be asked of science. Working in engineering schools, the authors have tried to use these futuristic proposals to transform them into lines of action. In this way, a reflection from the global societal context appears to fall back into more local aspects, close to the discipline or in a general way to the scientific context. At the same time, this table allows us to envisage a place for 4D printing in this future. However, as a reminder, the realization of an artifact requires the convergence of results for a "happy" outcome.

Theme	Targets	4D comments	References
Robotics and virtual spaces	Augmented reality; Internet via glasses and implants in the ear; speech recognition and voice control; simultaneous translation; decision support; active safety in transport; human–machine communication, etc.	From flexible robotics to programmable material	Yeoman 2008; Rus and Tolley 2015; Russell et al. 2015; Kopacek 2016; Hamet and Tremblay 2017; Lesort et al. 2018; Osaba et al. 2020; Schüppstuhl et al. 2020; Vu et al. 2020
Education, culture, virtual meetings	Artificial intelligence; sensors; microphones; simultaneous translation	4D little concerned	CES 2007; Kaplan et al. 2007; Mysiak et al. 2014; Rezaei and Behnamian 2021
Food and climate change	Resistant strains; water needs; smart home automation; automated supply; food safety; genetic modification; green chemistry, etc.	4D weakly concerned	Griffon 2007; This 2007; Puech 2008; Li 2016; Subramaniam 2016; Ryazanceva et al. 2019; Demoly and André 2021a
Getting around and living in the city	Telecommuting; work organization; energy; lighter materials; artificial intelligence; transportation; communication; adaptive architecture; self- cleaning	4D relationships with transportation modes and living spaces	Mangin 2005; Ferrier 2007; Widloecher 2008; Zartarian 2008; Nodesign 2009; Yian et al. 2017
Factory of the future	Artificial genomes; atomic and macroscopic additive manufacturing; robotics; self- assembly; nano-industry; human-machine communication; new energies; ITER; Industry 4.0; Society 5.0	Strong links with additive manufacturing robotics and the Industry 4.0 concept	Joachim 2008; Kim et al. 2016; Zhang et al. 2016; Tanaka 2018; André 2019; Komaki et al. 2019; Huang et al. 2020
Development of the body	Exoskeletons; killing death; active implants; artificial muscles; bionics; assistive devices; RFID and biometrics; NBIC convergence; smart clothing; reproduction	Assistive devices such as smart clothes are part of the 4D domain	Kaplan et al. 2007; Le Douarin 2007; Andrieu 2008; Wang et al. 2020
Medicine and health	Nano drugs; telemedicine; aging; computer-assisted surgery; artificial/augmented brain; predictive and precision medicine; active prostheses	Drugs delivery, stents, organoids and prostheses in 4D	Eynard and Salon 2007; Guerreiro et al. 2014; Wicks et al. 2014; Fatehullah et al. 2016; Kelava and Lancaster 2016; Takebe and Wells 2019; Ozturk et al. 2020; Tatullo et al. 2020
Soldiers of the future	Augmented reality; war without soldiers; emotional control; adaptive clothing; active protection; space	Active wear and protection	Schwartz 2006; Seligman and Fowler 2011; Fazal 2014; Drury 2015; Zheng and Walsch 2016

 Table 3.1. Artifacts envisaged in 15–20 years when associating 4D printing

Many areas are expected to develop, leading to significant changes in technology. What this table suggests is that it is reasonable to think that these emerging technologies will, as they develop, impose criteria that are specific to them, criteria that will have to be respected by the 4D printing processes that could contribute to their development (e.g. the need for biocompatibility for certain medical applications such as the delivery of drugs). These constraints (to be identified) should be exercised in the industrial development processes. In this context, and this is very important to remember, there is a significant gap between proofs-of-concept (which avoid bad avenues) and industrial prototypes that must meet all the expectations and constraints identified for possible economic development (Rubio et al. 2019; Battaglia et al. 2021).

#### 3.2.4. Weaknesses between research and profitable applications

[We] may say roundly: "If you try to further the progress of science as quickly as possible, you will end by destroying it as quickly as possible; just as the hen is worn out which you force to lay too many eggs." The progress of science has been amazingly rapid in the last decade; but consider the savants, those exhausted hens. They are certainly not "harmonious" natures: they can can merely cackle more than before, because they lay eggs oftener: but the eggs are always smaller. (Nietzche 2012)

The complexity that has been present since the beginning of this work is a situation in which a variety of elements, of different forms and natures, are dynamically interacting. Based on scientific knowledge and proofs-of-concept, innovation moves the complexity (without necessarily exploring or exploiting it) of a situation by modifying a relationship between interacting elements in order to offer citizens robust objects that meet a set of specifications. It can be of disruptive or incremental origin, with characteristics of interaction between multiple actors, interdisciplinarity, uncertainty, unpredictability, co-evolution of the project and its context, etc. It is therefore also part of a complexity issue. However, between proof-of-concept and industrial production, the design of objects (4D) is based on knowledge of what is real and what is possible (without being sure of the acceptance of products by the public).

In a universe of disciplinary scientific origin stemming from linear causalities, thinking about design implies adopting probably a less classical vision and therefore closer to humility in human actions, in order to admit that the action of disruptive innovation, once seized in unstabilized interdisciplinary and organizational dynamics, can escape in part from the initial intention. In this changing environment, the desire for control, rationalization, standardization of behaviors and reduction of risk-taking, applied to uncertain, unstudied and increasingly complex systems, reinforces the risks of failure and potentially their difficult governance, committed to instability, by obscuring disciplinary hierarchies, tensions, questioning, understanding of oppositions between process and content, and the qualities of interdisciplinary interactions, which are nevertheless necessary for their functioning.

In a very schematic way, Figure 3.1 shows the temporal evolution of the performance linked to a rather radical innovation. From promising niches, the new technology spreads to the entire material or immaterial production system, until the next stage. But, as Jacomy (2015) reminds us:

Often, several inventions are concomitant, and quite naturally, from the moment when the environment becomes conducive to their appearance and when someone, perhaps more astute than the others or more open to innovation, to the creative spark [appears]. It is not only the 4D printing domain that is moving! But, in any case excited by their project, their first practical step is however retrospective: they must return to a set already constituted, formed of tools and materials; to make or remake some [...]. All these heterogeneous objects constitute their treasure, they question them to understand what each of them could "mean", thus contributing to define a whole to be realized, but which will finally differ from the instrumental whole only by the internal arrangement of the parts. (Lévi-Strauss 1990)

The new, by principle of continuity, is not completely new, it is generally based on existing techniques, except in great disruptions (the passage from the candle to the electric bulb, etc.). But the field of 4D printing probably does not constitute in itself a revolution of such a scale.

In this figure, when a technological field reaches its peak, it may be advantageous to return to ideas or devices that have been abandoned (for various reasons) in order to progress; in fact, one remains, at least in part, controlled by one's past. But, in any case, one must invest "a lot" to have an innovation (which must be accepted by future users). Going to market brings up incremental research and development needs whose results support performance improvement, but, after a while, progress is increasingly costly and one may be waiting for disruptive solutions (Autio et al. 2015; Leyden and Link 2015; Acs et al. 2017). When an

industry chain approaches performance saturation (S-curve plateau), it may be advantageous to revisit the stock of previously abandoned ideas in an attempt to make further progress. The present situation shows increasingly short durations between these transitions, which force companies to enter more voluntarily into the (infernal) circle of innovation.



**Figure 3.1.** Continuous and threshold changes in technological performance

Investing in research and development seems obvious in this context, where the appetite for the new is maintained, confirmed by the significance of the expenditure that companies devote to it (as well as national and European state support). Yet it is a risky investment, as the production of knowledge has very different characteristics from those of a good or service: knowledge is not a product to be sold; it is always unique and different from the previous; its usefulness cannot be determined in advance; its final cost is never perfectly known; its value is unpredictable (Kratzer et al. 2008; Bernstein 2015; Bronzini and Piselli 2016; Howell 2017; Yoon 2017).

Figure 3.2 shows the current situation of the 4D context, located between scientific knowledge, proof-of-concept, design and industrial applications. This presentation is, however, reductive with application domains of different amplitudes and complexities. Nevertheless, the areas written in gray represent, for a large part, spaces to be conquered.



Figure 3.2. Between 4D scientific knowledge and industrial achievements

# 3.3. From matter to 4D form; from 4D form to function

To try to define the limits of the "material to the immaterial", it is to try to decipher and to give substance to the distinction so hackneyed of the sensitive and the intelligible, of the permanent and the intangible, of the concrete and the volatile. (Dulau 2005)

Because it belongs to the meaning of the mathematical project to establish a uniformity of all bodies according to space, time and relations of movements, this project allows and at the same time requires, as an essential mode of determination of things, the common and equal measure, that is to say, the principle of mensuration of the unthinkable diversity. (Heidegger 1988)

But to know or to be aware is to have time to avoid and prevent the moment of inhumanity. (Levinas 2006)

Basically, the question arises as to whether we have or will one day have a practical capacity to become engineers of the future of matter. By exploiting self-organizing potentials, we have shown that there can be spaces (admittedly limited in volume) where these can be exploited in order to achieve desired spatial and functional transformations (Demoly and André 2021b). But, our ignorance on this subject is still very great... and, behind this incompleteness of knowledge, a very important ethical question arises, that of the possibility of making our desires

and of making them express themselves on the matter, or even further... It is an interesting debate, but that we did not want to start in this work, because it could not, in the field of 4D printing, be supported by other opinions, more enlightened than ours. Before being a qualifier, algorithmic, a noun, refers to the decomposition of a precise calculation and its formalization in a generic calculation operation: "Good model + Good data = Good orders". It is about transforming a situation into sequences of logical operations, written in the form of diagrams, schemes or networks (Reigeluth 2016). The problem of algorithms is associated with the successful movement of "data processing" of society, a movement that has taken on unprecedented dimensions with the Big-Data.

However, this principle of control (social formatting) has already conquered our society. Harari (2017) introduced the concept of "dataism" which originates from the life sciences with their biochemical algorithms and digital algorithms (from which additive manufacturing and 4D printing are derived) and highlights close mathematical formalisms between the two, yet highly disjointed, fields. A central question is to know if everything human can be reduced to a set of operating rules whose application allows for solving a problem stated by means of a finite number of operations. A projection into a conceivable future (it is not certain that it will come true) would be to see the barriers between the two domains fall, with other problems to be dealt with in depth, such as that of the risk of entrusting our future even more to digital machines (Huxley 2013) and, as is often the case in this work, the (less oppressive) question of the treatment of the "inverse problem". But, there is the question of the finesse of the rules and the management of data, probably badly conditioned, and then of the "algorithmization of the human being" and with the "humanization of the algorithms". Moreover, for Houdé (2019), intelligence could work in three circuits: the short circuit of approximate heuristics, the long circuit of exact algorithms and the inhibitory circuit that blocks the heuristics, depending on the goal and the context. With feedback, the linear and determined approach would enter into learning processes. It is therefore not entirely certain that human functioning can be easily controlled by knowledge of the right data.

The shift from a homocentric to a data-centric worldview will not simply be a philosophical revolution. All truly important revolutions are practical [...]. Ideas only change the world when they change our behavior. (Harari 2017)

Classical mechanical algorithmics, as we know it in additive manufacturing or 4D printing, is subject to the deductive logic of mathematical demonstrations – "that is, to the movement from an abstract universal knowledge *a priori* to the knowledge of a particular case *a posteriori*, and thus to a temporality whose linearity tolerated only the expected deviations or variations" (Reigeluth 2016). The epistemological shift involved here goes much further. The reduced conception of intelligence and

learning as a nervous activity can undoubtedly be the object of modeling based on logical schemes, with the contribution of neural networks, etc. (Marsland 2015). One can then imagine a homology between the transmission of bits in an electronic circuit and the transmission of information adapted for the functioning of the brain, or, for 4D printing, the transformation of matter whose modification of form is imposed by algorithmic ways. Concretely, the dimensionality of the algorithm is then questioned (Witten and Frank 2005; Marsland 2015). If digital technology can handle a very large amount of data, a complementary question must be addressed aiming at linking digitized information to a certain mass of matter. This "small" problem is, to the authors' knowledge, not addressed... And yet, it is paramount!

The last great difficulty is the field of complexity and bifurcation! In spite of these obvious facts, scientists are quietly talking about a new, fascinating and somewhat disturbing world; that of stem cell engineering, biomaterials, micro-patterning and bio-printing, bioreactors and modeling of the living. They claim that we are already partly able to rebuild our organs, liver (Samsara 2016), kidneys, skin, heart and why not brain from our stem cells and in 3D printing and explain to us how scientists, all over the world, are making giant strides in this field of bioengineering, bio-printing (a form of 4D printing adapted to the living) and reconstruction of the living. But which form of living are we talking about? Is it based on the classical criteria of self-reproduction, function, communication and evolution? And with which degrees? Should we or should we not be wary of the first results and the seductive analogies? (Danzo and André 2020).

On these bases, the initial vision of the engineer was to make simple, if possible within a causal thought, with the tools at their disposal. The living matter can be ordered, meaning that its form can be, at least in first approximation, described geometrically. We can think that structures, more or less self-organized, must be associated with certain types of physical or physico-chemical conditions. However, from a broader evolutionary perspective, it is impossible to predict the evolution of forms by deducing them from the application of simple laws that we could have identified. More exactly, in experience, if these laws are necessary, they are not sufficient. They form the inescapable background of an evolution which develops in a non, apparently, deterministic way by interaction of the natural systems between them and with their environment. The sciences of "morphogenesis" would therefore only be valid within the limits of ordinary or macroscopic physics. However, the latter is only an approximation imposing to chase linear determinism. Turing is therefore not dead!

We realize that in order to give substance to a sequence of events, we must break out of a memory organized in silos, flee from the schools of ready-made thinking to embrace the dynamics. (Cohen and Nowakowski 2020)

Figure 3.3, taken from Danzo and André (2020), shows that from the same initial conditions, the fate in terms of cell growth of two interacting species can be very different from one simulation to another. Alan Turing brought many disciplines (including biology and bio-printing) into the era of complexity and bifurcation. Even with very simple equations, it is easy to observe losses of determinism in temporal evolutions. It is reasonable to think that by developing much more complex algorithms, with a number of recursions, the relations between "inputs or order" and "output results" would be very far from deterministic reductionism. But, this question remains to be explored.



Figure 3.3. 2D random shot (Monte-Carlo) modeling of a two-composition cell growth

In the preamble to this general reflection, we presented a figure (see Figure 3.4) that brought together what is happening and what was happening in the "world before" with already enormous changes induced by the development of digital technology. This "map" remains interesting as long as we know how to formalize the links between control and evolutionary structures, but with data that probably do not easily accept errors and grammatical mistakes (a question of robustness). The active materials (or their stimulation systems) will have to free themselves (question of control) from the problems of complexity and will have to be connectable.

It is possibly as an agent of a power of evocation that the transformation of the matter induced by a given energy stimulation (resolved in time and space) plays an active role on the design of a 4D object. The designer then has several roles, because they must play with the spatial distribution of the matter, the spatio-temporal distribution of the stimulation(s). We can think that the problem they deal with is not of the same nature if we start from a 4D idea as it has already been discussed or from the integration of 4D ideas in projects considered as probable in the near future (see previous section). It can be associated with the various correspondences that are established between a place, a body, an object and the memory (the knowledge of

the previous art). But, starting from this knowledge, it will be necessary to give a form to an object which must reach specific objectives of uses. It is in the programming modes allowing the fabrication of a 3D (or 4D) object that the qualifier "precise" deserves (without it being discussed) particular attention. The design, from a cultural point of view, does not accommodate badly the to-be-accurate and could thanks to data processing not concede anything anymore to the orthogonal reference.



**Figure 3.4.** *Quantum-continuum transition in the development of active material uses and 4D printing* 

To go further, technical design, especially in emerging fields such as 4D printing, must involve a collective. This team, with different skills, responsibilities and objectives, can consider the 4D object to be built from several points of view; for example, it can consider the design from the material(s) or, on the contrary, from the object's properties. It will be necessary to take design out of its conceptual aspects by discussing how creativity can be associated with participation and experience, etc. The members of a design group must consider the object of design through its constituents. The object of design is to integrate them for an operative purpose. The next chapter will address the following question: how to harmonize the claims and proposals of the different design actors, from the initialization of the project to its confrontation with the initial specifications. How does a member of the design group transform his projections on the object according to the objectives, on what criteria does he evolve his ideas, etc.? (see Bucciarelli (2002) and Binder et al.

(2012)). For the moment, we will try to examine how knowledge of the material and its evolution can allow for a transition to the 3D object.

To my mind, the only purpose of science is to lighten the toil of human existence. If scientists, browbeaten by selfish rulers, confine themselves to the accumulation of knowledge for the sake of knowledge, science will be crippled and your new machines will only mean new hardships. Given time, you may well discover everything there is to discover, but your progress will be a progression away from humanity. The gulf between you and humanity may one day be so wide that the response to your exultation about some new achievement will be a universal outcry of horror. (Brecht 2007)

#### 3.3.1. General considerations

Marenko (2014) considers that we are "becoming animalistic" in a world of smart objects. The creation of 4D-printed objects falls under a domain (not considered at the time) of interaction design. Animism is exploited in this framework and corresponds for this author to a mythical narrative and an "embodied fiction" that stimulates innovation in the "fluid, productive and meaningful relationship between humans and interactive systems" (Van Allen and McVeigh-Schultz 2013). Laurel (2008), on the other hand, describes the impact of digital technology on this field and the conceived animism "forms the basis of a poetics for a new world", in which computing induces animistic responses with behaviors induced by human interactions with smart objects (or using smart materials or passive materials).

This author suggests that an animistic response emerges when the technological framework linking objects becomes simultaneously smarter, more pervasive and more invisible. This would be a psychologically appropriate and imaginatively pragmatic response to the particular qualities of the information jungle. He writes:

The smartphone is no longer a mere "digital Swiss Army knife"; it is an object to which we give our full attention and in which we become emotionally invested – not to mention the new repertoire of physical gestures we have learned to use. All of our (often slightly compulsive) gestures of touching, tapping, pinching, stretching, stroking, sliding, scrolling, and holding are a particular set of new phenomena that I believe are the embodied expression of more animistic inclinations. Brown (2001) observes that objects are subject to particular subject-object relations in specific temporal and spatial contexts. What, then, might be the nature of the subject-object relationship in a world populated by increasingly responsive things?

On the one hand, the development of the IoT (Internet of Things) concept, on the other hand, that of 4D-printed objects... this association that will be created in the future will be examined in terms of exalted support for the production of new types of active objects in all human activities. For the moment, there are no industrial 4D-printed objects, therefore no effects in synergies, but the theme is to be followed (even anticipated). It is all the more interesting because it allows us to link technical innovation to its links with the social body (Chapman 2016). For example, according to Kimbell and Blomberg (2018), the design of such services could be anchored on exchange relationships between actors in a service system (grouping of resources and skills in particular arrangements called service ecosystems or value constellations). This exercise could, in agreement with Ginja (2018), serve sociological work on the creation and use of objects in the present era. In particular, the emergence of 4D-printed objects in a largely saturated world could be related to minimalist philosophies, which insist on the reduction of production and use of artifacts. On the other hand, as in some 3D productions, it could be considered to examine how the principles of user participation in the final configuration of objects (DIY for Do-It-Yourself) with aspects of customization can constitute an original aspect of 4D production.

## 3.3.2. Algorithms by/for 4D printing

Algorithm: "Method for solving a problem in a systematic way". (Champin 2019)

Modern science was born when attention shifted from the search for the "what" to the investigation of the "how". (Arendt 1972)

An algorithm is an ordered set of operations or a chain of specific instructions that must be followed in order. The algorithm takes the form of a flow chart. Its objective is to solve a problem, that is it has a limited objective with a result at the end of the operation (see Figure 3.5). What is important to note is the systematic character (without initiative) of its execution. Any computer program has the structure of an algorithm. But, what happens is that in reality the phenomena to be treated can be more complicated, even complex. Thus, the form of the flow diagram that forms the algorithm can become a "tree" of instructions that, depending on its complexity, can even offer unexpected results. In design, based on a certain amount of knowledge, a digital programming language is used to develop algorithms and create a set of instructions that the computer will use to design and realize a 3D/4Dprinted object (see Massaron and Mueller (2017)).



Figure 3.5. Elementary algorithm

The object of our reflection is a little broader. With the digital technologies represented by additive manufacturing and 4D printing methods, we are used to this logic of action. The question we ask is to examine whether we can develop algorithms by other means: for example, to put it simply, by using an active, temperature-sensitive material, there can be a simple physical instruction created for a given shape of the object. The design of the 4D object thus allows a relationship between an input and an output and a non-electrical or electronic mode of activation which is heat. Then, to go a little further, the generic question of the appropriation of 4D technologies as physical supports of algorithms arises. It is then a question of exploiting localized transformations in time and space based on chemical, physico-chemical and/or physical transformations.

Almost 15 years ago, Giavitto et al. (2008) considered that the chemical paradigm could be an unconventional programming paradigm well suited for the high-level specification of parallel systems. However, this model lacked an explicit management of spatial relations, which the authors modified by uniformly managing the data structures needed for algorithmics as well as the distributed data structures needed for programming autonomous or amorphous systems (see also Scheffler (2008)).

The physical-chemical information principle would have been used to evolve a multitude of different computational devices implemented in physical materials (but not using 4D printing). One of the biggest problems in materials exploitation is finding a good computational abstraction to carry the computation over the underlying physical process (Nichele et al. 2017). For their part, Przyczyna et al. (2020, 2021) have shown that in two dimensions it is feasible to work with cellular automata (see, e.g., Nichele et al. (2017)). For Chiolerio (2020), working on robotic extensions that come close to 4D printing, it is possible to enclose the logic (fluid) part in a shell. The interior provides the necessary functions, including data storage, processing and relaying, external stimuli detection, mobility, and energy storage and

distribution, like biological cells. While the devices are not operational today, this author proposes interesting macroscopic avenues for changing functionality that may be worth exploring. On the other hand, Jaworski et al. (2019) consider the use of nanoparticles.

However, Cortes (2014) reminds us that moving systems with kinematic loop closure constraints remain a challenge for motion planning (4D printing and soft robotics). Indeed, such constraints imply changes in the topological properties of the configuration space, which require a reformulation of motion planning for open-chain mechanisms. Closed-chain mechanisms are, however, common in robotics (see also Perrin (2012) and Gothard et al. (2012)). Parallel robots are a clear example. Closed kinematic chains are also created when multiple manipulators grasp an object simultaneously. Algorithms for computing the motions of closed kinematic chains are also important for applications in structural bioinformatics.

One of the limitations of current bio-inspired research concerns the materials used to build objects and robots that are models of the living. They do not resemble biological materials in any way. (Guillot and Meyer 2014)

Recently, Trégouët (2021), reporting on the costs of storing digital data, reminded us of the existence of other paths currently being studied, such as those using polymers, DNA or biological or biochemical media. He does not take into consideration the multi-stimulation component nor the shape of the artifacts. He writes:

It is clear that the smart combination of quantum machines, biological information storage and the Internet of Things will allow the emergence of a complete virtual world, integrating all spatial, temporal and cognitive dimensions, and which will be capable of constantly self-enriching. Far from being a simple representation of reality, this autonomous and living virtual world will become consubstantial with our reality and our lives, to the point where it will simply become impossible for us to distinguish it from the real world, since it will have become a new dimension.

In short, this is a field to be explored that should bring 4D printing out of its still timid, interesting niche role of 3D printing by bringing just a plus, that of the "simple" movement and/or functionality. The author believes that this reflection should be taken into consideration because it opens up new areas of investigation combining materials and shapes, probably obtained using fairly traditional algorithms, but to produce active artifacts with functionalities that are unimagined today, but which could significantly modify our daily lives tomorrow.

#### 3.3.3. Preforming the material

A structure is an operational set of indefinite meaning [...] grouping elements, in any number, whose content is not specified, and relations in finite number, whose nature is not specified, but whose function is defined. (Serres 1969)

Since object technology came to prominence a decade ago, experts have repeatedly predicted its demise, with the inevitable periodic announcements of the so-called "winter of objects," an allusion to the "winter of artificial intelligence" that froze the spread of artificial intelligence in the late 1970s. But winter has not arrived. And all signs indicate that the spring will continue. (Meyer 1998)

But, to reach these challenging goals, it may be interesting to design a 4D object starting from the material. Several general questions can then be asked before choosing an additive manufacturing process:

- the intended effect;

possible stimuli;

- possible performances (mechanical qualities, response time to one or more stimuli, etc.);

- the need for a single or multiple materials;

- the mass of material(s) required;

- the associated costs;

- the form that the active (and/or passive) elements have: material to be transformed (e.g. resins that are photo-polymerized in additive manufacturing), generally in liquid form, powders, composites, composite threads, sheets, massive structure, etc.;

– cohesive qualities of multi-material systems (including printability);

- the issue of functional fatigue of smart materials;

- the selection of a (reliable) fabrication and actuation strategy of the object (with the question of the spatio-temporal positioning of the voxels and the energy stimulations with possible transfers and shadow effects);

- experimental validation and, if necessary, improvement of the additive manufacturing process combined with stimulation by re-examining unsatisfactory areas.

However, it is necessary to have an interdisciplinary knowledge base ranging from materials (active) to processes, considering the fact that today, there are many ways of falling through the gaps, partially masked by an almost exclusive coverage, in the recent literature, of the creation of 4D objects presented in the form of proofs-of-concept. In such a situation, it is difficult to put forward robust methodologies for producing usable 4D objects. To reach the public domain, it will probably be necessary for some time to get closer to the traditional design methods that have proven themselves in additive manufacturing. But, you never know...

Perhaps our most compelling duty is to never let go of the thread of wonder. (Singer 2007)

After the guardians have first made their domestic cattle dumb and have made sure that these placid creatures will not dare take a single step without the harness of the cart to which they are tethered, the guardians then show them the danger which threatens if they try to go alone. Actually, however, this danger is not so great, for by falling a few times they would finally learn to walk alone. (Kant 1784)

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# **Design for 4D Printing**

Rational knowledge and rational activities certainly constitute the major part of scientific research, but are not all there is to it. The rational part of research would, in fact, be useless if it were not complemented by the intuition that gives scientists new insights and makes them creative. These insights tend to come suddenly and, characteristically, not when sitting at a desk working out the equations, but when relaxing, in the bath, during a walk in the woods, on the beach, etc. During these periods of relaxation after concentrated intellectual activity, the intuitive mind seems to take over and can produce the sudden clarifying insights which give so much joy and delight to scientific research. (Capra 1975)

# 4.1. Introduction

The previous chapters have shown that understanding 4D printing in its entirety is a very complex process. This technology requires the consideration of multiple, generally disjointed forms of expertise (businesses and disciplinary fields). At the same time, it introduces new opportunities and freedoms to develop and manufacture objects and structures capable of evolving in their usage environments. The resulting potential technological applications give way to needs intimately linked to new modes of object-environment, object-object and object-human interaction. In this context, most of the research work – which is part of the supply and demonstration logic – suggests a promising future. But to transform the trial, it is important to refocus this technology – governed by the triptych process-materialenergy stimulus – in the design phase where decisions have a major influence on the success of a 4D project.

4D Printing 2: Between Science and Technology,

For a color version of all the figures in this chapter, see: www.iste.co.uk/demoly/4Dprinting2.zip.

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Over the last few decades, the integration of lifecycle constraints has been the subject of much attention from researchers and industrialists, leading to the concepts of "concurrent engineering" and "integrated design" (Sohlenius 1992; Tichkiewitch 1994; Prasad 1996). The design phase is certainly a "place" where the right decisions have to be made regarding the architecture of the product, its geometric definition, the selection of materials and manufacturing and assembly technologies (Otto and Wood 2001). It is then necessary to initiate actions involving the stakeholders concerned in order to make the designers aware of potential constraints or complications linked to the decisions made. The design phase is therefore a strategic phase for initiating/validating concerted creative steps guided by constraints and business rules. In such a context, it is important to remember that 4D printing technology highlights both the constraints of the additive manufacturing and the functional role of the active materials introduced, which bring behaviors that are still poorly understood in this phase. The interdisciplinary nature of the technology must be controlled on different levels of design ranging from system engineering, product design to material design.

"The notion of integrated design includes the integration of the product's life cycle during the design phase by integrating all of the actors who will be involved in the history of this product in the short or medium term" (Tichkiewitch 1994).

#### Box 4.1. Integrated design

In the same way that additive manufacturing was imagined by the author of Tintin and Spirou (see Introduction), 4D printing and in particular the inspirations around programmable matter were borrowed from science fiction and animation films, as has already been mentioned. These movies – which are intended to transport us into another universe – open up an interesting path, among others, of disruption, of imagining far-off places, in short, of creation for researchers wishing to go beyond current paradigms. They offer us in a way a possible finality, one that, generally, is very spectacular and allowed by 4D printing. We must now find a way to reach it; this is the challenge of this chapter.

## 4.2. How can 4D printing in design be approached?

Matter undergoes such changes that it ceases to remain what it was, a substance. (Dagognet 1989)

Through design, 4D printing generates technological inseparability when it comes to producing parts, through 3D printers, made active by stimuli, rather than through activities organized in different successive steps. It is therefore a major
disruptive integration that can revolutionize the field of design if it can meet industrial specifications in terms of mechanical strength, responsivity and fatigue (otherwise, one would have to be content with seeking niche applications, corresponding to a more modest attractiveness and a less exciting research momentum), or generate new products with new modes of interaction. Changes in production strategies, materials development and manufacturing techniques have always influenced how people think about and perceive physical products and objects, and especially activities related to design and engineering (Otto and Wood 2001; Ulmann 2002; Pahl and Beitz 2013; Ulrich et al. 2015). This is the case with additive manufacturing, as this technology challenges traditional design practices focused on subtractive and formative manufacturing by paving the way for multi-functional and multi-material parts with complex geometries (Beaman et al. 1997; Jepson et al. 1997; Gibson et al. 2010; André 2018). Building on this disruptive manufacturing technology, primarily focused on complex shapes and high-resolution problems, researchers over the past decade have paved the way for the integration of smart or active materials via user instruction or autonomous sensing in their use environment (Addington and Schodek 2005; Ge et al. 2013; Tibbits 2014). Printing active materials or smart voxels from a digital perspective is therefore driving related research fields and scientific communities toward new interdisciplinary and converging research paradigms, namely, bioprinting and 4D printing (André 2017a, 2017b).

Because of its transformative capacities through additive manufacturing, 4D printing has received sustained attention from researchers in fields such as chemistry, applied science, physics, materials science and engineering, and mechanics. By combining the deposition modes offered by this process, energy stimuli and material functionalities, proof-of-concept-oriented contributions are promising and multidimensional. 4D printing then allows for multiple transformation scenarios between two object states, whether between manufacturing and use phases (e.g. the case of origami-inspired deployable solutions using folding functions) for logistical and transportation reasons, or even between two use stages (e.g. the case of transformable/adaptable solutions using disparate functions) (Ferguson et al. 2007; Singh et al. 2009; Weaver et al. 2010; Sosa et al. 2014). These developments are extremely appealing from a conceptual point of view but only provide a partial solution that is difficult to implement in real-world applications. This is due to the fact that most scientific efforts follow an incremental approach leading to specific solutions and are mainly addressed by scientific mono-disciplines supporting technological innovations.

To reach a threshold of industrial maturity, the fields of chemistry, process and materials science must be: (1) appropriately aligned with end-user expectations/needs; and (2) arranged to operate at the system/product/object level, which involves applying a problem-solving approach (i.e. teleology or inverse problem questions, as

well as user- and system-centered design) (Brown 2008; Camburn et al. 2017; Perez et al. 2019; Tushar et al. 2020). Indeed, to date, little effort has been made to integrate 4D printing into the design and development phases of smart devices (Lantada 2017; Sossou et al. 2019a, 2019b).

Therefore, it has become urgent to bring these advances to the attention of designers and product engineers in order to address "design for 4D printing" issues, as has already been successfully done to integrate assembly (Design for Assembly), conventional manufacturing (Design for Manufacturing) and additive manufacturing (Design for Additive Manufacturing) constraints into product design (Boothroyd 1994; Demoly et al. 2011a, 2011b; Sossou et al. 2018). By considering 4D printing at such levels of expertise and operation, the next generation of smart products and devices should be able to respond to usage over time and, more importantly, satisfy industrial needs/demands and the market, which point us toward the futuristic applications in the previous section. Furthermore, bringing interdisciplinary knowledge on the scales of mechanisms of change of properties, functionalities, states and shapes into design and engineering would open a space of solutions with active materials integrating functionalities like sensors, actuators, transducers, etc. One possible way is, as shown in Figure 4.1, to get closer to what nature can do, via biomimicry.



Figure 4.1. 4D printing as a manufacturing technology based on biomimicry (Demoly et al. 2021)

Indeed, this search for transformation, adaptation and even reconfiguration in the systems/products/objects to be developed meets the principles of biomimicry (CNRS 2019). By endowing traditional 3D printed objects with behavioral capacities, 4D printing is one of the most appropriate manufacturing technologies to address this fashionable paradigm. Over the years, nature has received much attention from researchers and engineers to design and build novel forms and structures, whether in architecture, art and, more recently, mechanical systems (Benyus 2007; Liu and Jiang 2011; Goldfield et al. 2012; Fu et al. 2014; Zhang et al. 2016). This last point seems to be legitimized by the advances in additive manufacturing techniques and their adoption over time by industry. However, in order to manufacture biomimetic objects that ensure the translation of innovative mechanisms to solve engineering problems, additive manufacturing processes must be combined with another ubiquitous driving force – behavior (Kempaiah and Nie 2014; Gladman et al. 2016; Oliver et al. 2016). As such, Figure 4.1 illustrates 4D printing as a means of realizing the basics of biomimicry (structure, form and behavior), thus introducing issues of spatio-temporality and complexity (André 2020).

# 4.3. Opportunities and challenges in design: a strategic roadmap for research

Any path is only a path, and it is not offensive to yourself or others to leave it if you feel like it. Look at each path separately and deliberately. Try them as many times as you think necessary. Then ask yourself, and yourself alone: does this path have a heart? If it does, the path is good; if it doesn't, it is useless. (Castaneda 1977)

# 4.3.1. Evolution of technological solutions and associated challenges

The broad spectrum of research initiatives and current limitations of 4D printing demonstrate that (see Figure 4.2):

- this is a rapidly growing area of research due to the abundance of proofs-ofconcept and interest in fabricating active structures with additive manufacturing technologies;

- the underlying interdisciplinary facets can no longer be addressed in silos.

Interdisciplinary issues in product development have been addressed in recent decades regarding the types of technologies used in products and objects. This is because the manufacturing industry has been progressively forced to increase the capabilities of products and systems and reduce their lifecycles by ensuring a certain level of flexibility and efficiency as competitive advantages, especially in the product development phase where design activities and decisions have a significant impact on downstream processes (i.e. manufacturing, assembly, maintenance and recycling) (Demoly et al. 2013). This has been successfully achieved by considering:

- the capture and integration of lifecycle constraints and knowledge into product design (e.g. design for X approaches, advanced computer-aided design (CAD)) in mechanical product design to deliver products aligned with their lifecycle phases (Demoly 2019);

- the integration of other disciplines such as electronics in the design of mechatronic systems;

- the integration of computing capability in the Internet of Things (IoT) era for connected objects (Kiritsis 2013).



**Figure 4.2.** Evolution of technology products incorporating scientific issues, key results and their capability maturity level (Demoly et al. 2021)

Figure 4.2 shows that these types of systems have generated interesting and successful results in terms of theories, models, approaches and tools with a high level of capability (mostly levels 4–5 on the Capability Maturity Model scale). A promising and emerging trend in interdisciplinary engineering and design – as highlighted by the European Factories of the Future Research Association (EFFRA 2020), Manufuture (Manufuture High-Level Group 2018), the National Aeronautics and Space Administration (NASA) (Liu et al. 2018) and the European Space Agency (ESA) (Mitchell et al. 2018), to name a few – is to increase the responsiveness and intelligence of systems/products once they are manufactured based on user needs or customer expectations, and then increase their lifespan by providing them with an appropriate use configuration in a specific context and use environment (see the blue box in Figure 4.13) (Burman et al. 2000; Brown 2008; Christoforou et al. 2015; Camburn et al. 2017; Perez et al. 2019; Tushar et al. 2020). It is also about increasing the functionality of systems while connecting them with their digital "parent" twin, paving the way for digital, physical and even social

connectivity. We will then speak of smart systems governed by pre-programming matter (within material or micro/nano robots). On this last technological evolution, we can note scientific barriers to the integration of active materials, electronics and additive manufacturing in design, the majority of the work being oriented on the mechanical modeling of active material behaviors. These research actions are part of the broader context of Industry 4.0 and its components such as additive manufacturing, soft robotics and 4D printing in particular (see Figure 4.3).



Figure 4.3. Industry 4.0 paradigm

Over the last decade, the emergence of smart products/systems (i.e. capable of adapting to their environment, self-assembly, self-repair, sensing their environment and reacting accordingly in a programmable manner) and innovative additive manufacturing technologies has required a profound revision of well-established design and engineering models and approaches to provide dynamic product definitions (i.e. product geometry integrating one or more materials) at different levels of description (mechanical assembly, part, material, etc.). To move to an operational and industrial threshold – where inverse problems are mainly adopted by architects and mechanical designers in their routine activities – 4D printing must be considered from a design and development perspective. Implementing this manufacturing paradigm shift – with an emphasis on multi-material printing strategy – in the product development process poses great challenges and opportunities for the industry. The following sections aim to identify key research directions to successfully implement/apply a vision of 4D printing dedicated to smart system development.

# 4.3.2. Design for 4D printing

For 4D printing, everything leads us to establish a roadmap – called Multi-X (X here represents the different components of 4D printing) – which aims to map and take into account all aspects of 4D printing to develop innovative products/systems. The ultimate goal is to design and produce smart products/systems that make sense

for society, that meet the needs of users and associated interactions and, in a way, that "live" (evolve in terms of form, property and/or functionality over time). To approach such a long-term vision, it is relevant to consider the multiple aspects concerned by the technology, that is multi-material, multi-domain, multi-process, multi-function, multi-physics, multi-world, multi-representation, multi-sector/niche, multi-scale and the associated temporality(ies), for which knowledge, models, methods/processes and tools/machines have to be elaborated in the process of designing smart systems/products/objects. This can also be done using reasoning procedures such as parametric/topology optimization, bio-inspired reasoning and connectionist/symbolic artificial intelligence (AI). Figure 4.4 provides an overview of the above research directions. It also shows the underlying complexity of the facets of 4D printing and presents many scientific opportunities and challenges if combined with each other.



Figure 4.4. Design roadmap for 4D printing (Demoly et al. 2021)

In this context, design for 4D printing must allow engineers and designers to develop solutions that can be realized by additive manufacturing and assembly (if needed) and that must be compatible with usage modes involving the environment (which can play the role of a stimulator) and the end user. The integration of this technology in design therefore requires several means to be harmoniously addressed, namely:

– Design with active materials (Liu et al. 2018), whether they have the roles of actuators, sensors, converters or multiple roles, spatially arranged in coherence with the stimulation strategy.

- Design with the stimulator, whether natural or artificial, global or local, internal or external, single or multiple, consistent with the active materials.

- Design for additive manufacturing (Rosen 2007; Sossou et al. 2018), thus enabling consideration of manufacturing constraints in the context of homogeneous, heterogeneous or hybrid structures.

- Design for assembly (Demoly et al. 2011a), at the material scale (material compatibility and voxel assemblies) in hybrid 4D printing, or even at the object scale (assembly during and/or after manufacturing) in heterogeneous 4D printing.

- Design for/with the end user. Although every solution is designed to satisfy needs, 4D printing and its evolving solutions require being able to take into account potential new modes of interaction. The end user must be able to play a role in the design process. This is all the more true with the democratization of additive manufacturing where the "maker" designer is also the end user.

# 4.3.3. Methodological framework for the design of energy-sensitive structures

With the massive development of proofs-of-concept to illustrate stimuliresponsive behaviors in objects and structures, it becomes important to think about a general design framework for 4D printing that promotes an inverse problem-oriented approach to build necessarily innovative solutions that are expected to be satisfactorily robust. With respect to design methods for additive manufacturing, conventional manufacturing and successfully adopted assembly (Rosen 2007; Gibson et al. 2010; Demoly et al. 2011a, 2011b; Meisel and Williams 2015; Thompson et al. 2016; Richter et al. 2017; Sossou et al. 2018), the level of complexity induced by the multiple human actors, on the one hand, and influencing parameters, on the other hand, involved (i.e. the architect, the designer, the materials expert, the process planner and the mechanical engineer), and the spatio-temporal behaviors inherent to active and passive materials require a progressive or integral integration of the constraints of 4D printing, whether at the functional, logical/behavioral, structural or geometric level, as well as the introduction of user-centered design innovation mechanisms.

A proof-of-concept is (simply) a demonstration of feasibility. It is, however, a realization whose mission is to show the feasibility of a process or an innovation. The notion of robustness does not usually enter into the development of proofs-of-concept.

Box 4.2. Proof-of-concept

Thus, Figure 4.5 represents a design for the 4D printing process, covering the conceptual design, embodiment design and detail design phases. It highlights the top-down definition of stimuli-sensitive systems/products/objects and the concurrent consideration of stimuli, active materials and the constraints and knowledge of additive manufacturing. Coupling with innovation approaches provides flexibility to focus along the way on the desirability of all stakeholders involved and the opportunity to inject creative, inspired and user-centered contributions into the design stages (Perez 2018). This coupling will allow engineers and designers to exploit the potentials of 4D printing while projecting toward possible innovative objects/systems as imagined in section 4.2.



Figure 4.5. General framework of innovative design for 4D printing

In considering such a general framework with design theory and methodology for additive manufacturing (Yang and Zhao 2015), new critical steps have been introduced and those already adopted now require new considerations (Perez et al. 2019). Each step, shown in Figure 4.5, represents a set of opportunities and challenges in and of itself, and requires decision-making supports, heuristics, best practices and rules. In their current form, the main steps can be handled proactively to involve active materials and critical additive manufacturing decisions leading to development from different perspectives (i.e. with respect to structure, geometry and properties) in the early stages of design, and offer enough flexibility to incorporate any reasoning procedure (Roucoules and Demoly 2020).

## 4.4. Capture and reuse of 4D printing knowledge

Reality is movement. Now, life is an evolution. We concentrate a period of this evolution into a stable view which we call a form, and when the change has become considerable enough to overcome the happy inertia of our perception, we say that the body has changed form. But, in reality, the body changes form at any moment. Or rather there is no form, since form is motionlessness and reality is movement. What is real is the continuous change of form: form is only a snapshot taken on a transition. (Bergson 1907)

Following an inverse problem approach leads to the application of a top-down design strategy. To do so, this broad area of research requires the establishment of a solid knowledge base regarding energy stimuli, adaptive or smart materials, product models and manufacturing/stimulation/actuation processes. Among current efforts to formalize knowledge on conventional materials (Biswas et al. 2008; Ashino 2010; Zhang et al. 2015a, 2015b; Ghedini et al. 2017; Zhao and Qian 2017) additive manufacturing (Dinar and Rosen 2017; Jee and Witherell 2017; Liang 2018; Kim et al. 2019; Sanfilippo et al. 2019) and product models (Gero 1990; Tichkiewitch 1995; Fenves et al. 2001, 2004, 2008; Sudarsan et al. 2005; Demoly et al. 2010), we can see that little attention has been paid to formalizing and capturing 4D printing knowledge. Apart from Nam and Pei's (2019) initiative on a possible taxonomy of shape-shifting behaviors, it is indeed the interdisciplinary knowledge inherent to this technology that needs to be formalized and related. The importance of knowledge capture/reuse in design approaches for X (X being a lifecycle or a technology) is now recognized by the academic and industrial communities.

It is in this context that a domain ontology – developed in the framework of the French national research project PIA ISITE-BFC HERMES – has been created to formalize the knowledge of 4D printing at the product and object scales (Dimassi et al. 2021). This ontology is based on the high-level Basic Formal Ontology (BFO) (Guarino 1995; Arp et al. 2015) which enables, through the philosophical theories of endurantism and perdurantism, and even mereotopology, structuring domains and associated knowledge (see Figure 4.6). This type of construction through the BFO guarantees its accessibility for any other domain ontology such as those promoted in the international initiative Industry Ontology Foundry (IOF)<sup>1</sup>.

<sup>1</sup> https://www.industrialontologies.org.

#### DEFINITIONS.-

- Ontology: "An ontology is a formal and explicit specification of a shared conceptualization" (Studer et al. 1998).

- *Endurantism*: philosophical theory according to which the material objects are persistent three-dimensional individuals entirely present at each moment of their existence (Sider 2001).

- *Perdurantism*: philosophical theory according to which the material objects have temporal parts throughout their existence (Sider 2001).

-*Mereotopology*: a first-order theory of formal ontology (branch of metaphysics) that expresses mereological and topological concepts, relations between parts and the whole, and their boundaries (Smith 1996).

The capitalization and formalization of knowledge must be thought through beforehand, not only in terms of their reuse and reasoning scenarios, but also in terms of research work and published standards (ISO/ASTM 52900 2015). Given the strong growth of 4D printing in terms of the number of scientific publications, the implementation of knowledge change management mechanism is to be expected (Pittet et al. 2014). Efforts should therefore address this issue and extend the knowledge of 4D printing, whether at the system and material level or even to cover soft robotics (Marechal et al. 2020) and structural electronics, while ensuring their semantic alignment (Hagedorn et al. 2019). If this research direction is appropriately addressed, then symbolic AI reasoning procedures can be developed as in various selection problems and as design decision-making supports recommendations. They can also play a key role in making sense of connectionist AI-based design approaches (Demoly et al. 2019; Landgrebe and Smith 2019).

What Figure 4.6 shows us is that the HERMES ontology is constructed by considering both a top-down approach (guided by philosophical theories and the study of material objects) and BFO classes of high level of abstraction as well as a bottom-up approach (guided by both theoretical and experimental published research work). The objective of formalizing and representing design knowledge for 4D printing thus requires us not only to map concepts (called "classes") to properties of the studied domain, but also to provide formal specifications in order to be interpretable by computer for different uses and different business actors (product architect, designer, mechanical engineer, materials expert, process planner, etc.). Figure 4.7 shows the first-level concepts of the HERMES ontology built from the higher level BFO on the concepts of "continuant" and "occurrent", allowing the spatial and temporal/spatio-temporal regions, respectively, to be structured.



**Figure 4.6.** HERMES ontology construction strategy to formalize 4D printing knowledge and reuse it in different scenarios (Dimassi et al. 2021)



Figure 4.7. High-level concepts of the HERMES ontology (Dimassi et al. 2021)

On the other hand, Figure 4.8 provides a "design" oriented view of the ontology, with the other views detailed in the work of Dimassi et al. (2021). As this technology is mainly characterized by form and function as well as by property transformations and multi-material printing capabilities, it is indeed important to revisit established design patterns to support knowledge and rules from different perspectives in space and time. This view can be seen as an integration lever to connect all aspects of 4D printing. Designing a product/object for 4D printing highlights the need to provide an appropriate knowledge representation. According to the BFO, a material entity can be either an object or an aggregate of objects, but not in an approximate way; therefore, it provides an interesting knowledge modeling strategy to represent objects from different perspectives. In this case, the "Object" class can be about a 4D printed part as a whole with multiple states (e.g. manufacturing state considered as initial state and use state designated as final state) in space and time, but it can also be related to a component to be integrated (e.g. an electronic part, a liquid crystal elastomer film and an electrode). The different states and related geometric definitions of 4D printed objects - which can be 1D, 2D or even 3D - are made possible by the transformation function.



**Figure 4.8.** Example of the "design" view of the ontology highlighting object concepts and properties related to the decomposition of objects into voxels that can carry materials and properties (Dimassi et al. 2021)

Using a spatio-temporal modeling approach, an instance of the "Object" class carries the identity of an object that evolves in space and time under its different representations. The "Object\_aggregate" class, on the other hand, plays the role of a collection of objects. A spatio-temporal entity is a representation of real-world entities, which consists mainly of an identity, descriptive properties and spatial properties. While the identity describes a fixed specification of the entity, the alphanumeric and spatial properties can vary over time and thus represent its dynamic part. When the identity transforms into a new entity (Gruhier et al. 2015). Identity can be defined as the uniqueness of an object, regardless of its attributes or values. Identity is essential in conceptualizing and modeling a phenomenon (Muller 2002; Del Mondo 2010).

Following the same logic of the above object classes, the "Voxel" class is described as a volume element arranged and spaced in a "Grid" (considered a type of spatial region in the BFO), which carries multiple data properties (e.g. color, position and scale in terms of size in a grid), multiple mechanical properties and multiple materials. It can be refined to a specific spatial resolution and appropriately sub-divided with quadtree and/or octree data structures for geometric representation, numerical simulation or reasoning based on machine learning or deep neural networks. The voxel can be considered as a tool to decompose any 3D object into multiple sub-objects in order to allocate materials, simulate their behavior or even increase additive manufacturing capabilities.

In order to be used in an adequate way, the ontology requires reasoning layers, either by a system of logical description rules for verification, classification for example, or by other mechanisms such as natural language processing (NLP) to interpret the tacit knowledge contained in patents and scientific publications (Siddharth et al. 2021). In order to increase its operational level, it is also appropriate to work on application programming interfaces (APIs) dedicated to semantic alignment and specific communication with design, simulation, manufacturing planning and even digital twin tools, as specified in the possible use scenarios (see Figure 4.6).

The mathematician's patterns, like the painter's or the poet's, must be *beautiful*; the ideas like the colors or the words, must fit together in a harmonious way. (Hardy 1956)

#### 4.5. Functional design

## 4.5.1. Functional modeling and solution principles

Considering evolutionary requirements to develop deployable, reconfigurable or transformable systems introduces additional transformation functions or dynamic functions to cover different states (whether from manufacturing to use phases or simply during the use phase) (Stone and Wood 2000; Hwang et al. 2021; Ferguson et al. 2007; Singh et al. 2009; Weaver et al. 2010; Sosa et al. 2014). If detailed enough, these functions can be directly linked to solution principles based on active material/stimulus associations. These can be incorporated at this stage of the process because active materials have a functional facet. The functional modeling of systems responding to a stimulus requires the mapping of technical functions, materials, signals and information. As such, the use of the "Black Box Model" as defined in works by Stone and Wood (2000), Otto and Wood (2001), Hirtz et al. (2002), Ulmann (2002) and Pahl and Beitz (2013) may be an appropriate strategy, to which the challenges are:

- composing transformation functions with multiple basic functions (e.g. a fold can be decomposed into multiple expansion/contraction functions);

- consolidating technical functions with multi-material and multi-function objects;

- integrating technical and multi-material functions, focused on the user and their interactions;

 – advancing design for additive manufacturing and innovation by integrating processing principles, multi-material printing capabilities and functional augmentation of products (Rosen 2007; Gibson et al. 2010; Perez et al. 2019);

– developing creative and technical bio-inspired additive manufacturing design principles that combine form, behavior and structure with active material and stimulus distribution patterns (Daly et al. 2012; Rosen 2014; Fu et al. 2016; Yilmaz et al. 2016; Sinha et al. 2017; Abdelall et al. 2018; Richter et al. 2018; Blösch-Paidosh and Shea 2019; Hwang et al. 2021);

- establishing mechanisms for recommending design principles.

Following the initiative carried out by Sarica et al. (2021) to generate pre-concepts from semantic technologies, it would be appropriate to reconcile 4D printing knowledge with natural elements by analogy. This direction could greatly contribute to the emergence of innovative solution principles.

## 4.5.2. Smart material/stimulus selection and processing planning

"What matters" said Nietzsche, "is not eternal life, but eternal vivacity". All the drama is, in fact, in this choice. (Camus 1955)

The selection of the best concept to develop requires a comparative analysis of the generated solution principles against the end-user requirements and the functional specifications. The concept can emerge either from a single design principle or from a concatenation of several principles. However, in order to enable engineers and designers to make good decisions, it is important to provide them with assistance in selecting compatible and available active materials and stimuli. This can be addressed by establishing a transformation sequence that describes the desired changes over time. To do this, a strategy for applying stimulation, whether global versus local, internal versus external, or a combination, must be defined depending on the mode of stimulation (autonomous with the external environment or controlled with the integration of a stimulator within the structure).

Once the transformation strategy has been defined and adopted, the selection of active materials/stimuli can be performed. However, it should be noted that this critical step suffers from a lack of selection tools (Ramalhete et al. 2010). This is mainly due to the complexity of the ad hoc experiments proposed in the literature that highlight a wide spectrum of parameters (stimulus amplitude, stimulus flux, composition, size, shape and distribution of materials, 3D printing parameters, etc.). Currently, there is no well-suited database for comparing active materials and stimuli. Although the HERMES ontology (Dimassi et al. 2021) provides mechanisms for structuring material/stimuli data, developing a standard protocol for testing active materials would greatly increase their adoption in design engineering. A first issue is to establish key performance indicators (KPIs) for active materials similar to those that exist to measure actuator and sensor performance, as presented in Chapter 1, section 1.2.5.2 (Volume 2). Another possibility is to use AI techniques to capture data and information from the published literature and populate a 4D printing ontology. The ultimate goal is to appropriately select active/stimulus materials capable of meeting the desired change performance requirements. This research direction should be approached in a manner that is consistent with current American Society for Testing of Materials (ASTM) standards.

# 4.6. From architectural design to detailed design

The universe is seen as a dynamic web of interrelated events. None of the properties of any part of this web is fundamental; they all follow from the properties of the other parts, and the overall consistency of their mutual interrelations determines the structure of the entire web. (Capra 1975)

#### 4.6.1. Definition of design spaces and CAD representations

Throughout the design process for 4D printing, the CAD representation of the solution to be defined must be approached from different levels of abstraction, viewpoints and scales. In Figure 4.9, top-down design – advantageous for integrating knowledge and constraints – has highlighted CAD modeling strategies, whether they are behavioral specifications with geometric skeletons (which may include lines, points, planes, sketches, etc.) (Demoly et al. 2011; Demoly and Roth 2017) or 3D design spaces (incorporating interface skeletons, functional surfaces, volume-envelopes, etc.) with geometric modeling to specify the overall product/part geometry and active regions to be developed or even for properties, materials and stimuli with voxel-based multi-scale modeling.





These strategies are necessary to approach the inverse problem necessary for addressing design for 4D printing and offering different levels of abstraction to introduce advanced reasoning procedures. As far as the design space is concerned, it is indeed a matter of constructing geometric skeletons to control the shape change behavior of the structure and also to allocate regions to be activated by stimulation where a particular attention in terms of simulation is needed. Based on this geometric input, the multi-material distribution requires a specific CAD representation in order to determine the appropriate properties at the right place, thus responding both to a desired transformation at the digital level and to a multi-material 4D printing. Thus, voxel-based multi-scale modeling appears to be extremely strategic in enabling both material design and design with material (Liu et al. 2018).

The multiple representations distributed over the architectural and detailed design phases require the ability to be aligned and reconciled (Roucoules and Demoly 2020). The progressive evolution of abstraction levels offers CAD representations suitable for knowledge reuse and the generation of solid and/or multi-material lattice solutions. However, the complexity of the generated 3D models requires the adoption of an appropriate data exchange format, whether to capture critical information at different scales or to exchange information to the manufacturing stage (Qin et al. 2019). Some of the most widely used data representations for additive manufacturing include Standard Tesselation Language (STL) (Roscoe 1988), Geometry Definition File Format (OBJ) (Wavefront Technologies 1995), Additive Manufacturing Format (AMF) (F42 Committee 2016) and 3D Manufacturing Format (3MF) (3MF Consortium 2018), which are receiving a lot of attention from the industry. In terms of structure, the AMF format seems to be the most appropriate representation format to accommodate multi-scale and multi-material data and is of common interest to academics and industry alike.

## 4.6.2. Voxel-based modeling and simulation of active material behavior

How to live without a stranger in front of you? (Char 1987)

Designing a shape or property change through heterogeneous 4D printing leads us to place the right material in the right place consistent with the stimulation strategy and transformation sequence imagined in Figure 4.5 (Khou and Tan 2007; Biswas et al. 2008; Li et al. 2020). The arrangement of materials in a 3D object leads to the construction of heterogeneous objects (except in homogeneous 4D printing) and goes against classical modeling approaches oriented mainly on geometry and topology (Stroud and Nagy 2011). The integration of materials, or at least information carried by materials, into defined spaces requires a change in the geometric modeling paradigm or at least the introduction of a new representation in CAD, voxel-based modeling. This type of discrete modeling has been the subject of much research in the past (Kaufman et al. 1993) and is at the core of thinking in the additive manufacturing community for controlling the deposition of multiple materials on the object under construction (Chandru et al. 1995; Hiller and Lipson 2010; Doubrovski et al. 2015; Aremu et al. 2017). The work of Westbrook and Qi (2008) and Hiller and Lipson (2010) demonstrated that the spatial distribution of active materials within structures plays a prominent role in order to achieve the expected behavior once subjected to the stimuli. Since then, this pathway has become essential to print physical voxels (Hiller and Lipson 2010; Lipson and Kurman 2013; Gershenfeld et al. 2015; Huang et al. 2015; Cramer et al. 2019; Jenett et al. 2019), provided that the few additive manufacturing processes capable of realizing them are taken into account or new ones invented.

In this context, the freedoms offered by heterogeneous 4D printing, and to a lesser extent hybrid 4D printing, and the material distributions made possible (see Volume 1, Introduction, Figure I.5(a)) lead us to address a voxel-based representation in order to: (1) simulate the behavior of active materials; (2) compute complex material distributions as a function of a targeted change in shape or property (case of the inverse problem); and (3) identify counter-intuitive materials distributions via AI.

The proposed approach is based on the primary assumption that the material properties within a voxel are homogeneous, that is a voxel can only accommodate one and only one isotropic linear material (or a homogeneous mixture of materials). On the other hand, two important aspects can be considered together. The first one concerns the use of a lattice, in order to quickly simulate and obtain a relevant qualitative response of the behavior associated with a distribution under a specific stimulus; the second one integrates two well-established techniques in the field of 3D computer graphics: skinning and rigging, techniques currently dedicated to the animation of virtual characters. The rigging technique controls the deformations of an object by using a set of deformation primitives related to the movement of the skeleton (which is made of bones and joints) (Jacobson et al. 2014). To replace a mass-spring system - usually used to study the behaviors of mechanical systems between adjacent voxels, "beam" type elements were used, as shown in Figure 4.9. These beams are resistant to traction, biaxial bending and torsion. They are governed by the Euler-Bernoulli theory, neglecting the influence of shear. The voxels located at both ends of a beam - can then move in space. We can also specify that each end of a beam has six degrees of freedom; the beams extend between the centers of adjacent voxels, half of their length being inside one or the other of the voxels, and will thus depend on the properties of the affected materials.

This modeling strategy allows us to quickly explore possible distributions in the embodiment design phase, as well as to expose large deformations (e.g. hydrogels can contract up to 400% and shape memory polymers can have an elastic deformation capacity exceeding 200% in many cases). Indeed, numerical

simulations using the finite element method require, as a general rule, a significant amount of computation time and memory to provide answers that are of a different order than those expected in the present case (answers of a qualitative order before the detailed design phase).

The fundamental prejudice is that order, clarity, everything systematic is necessarily inherent in the true essence of things; and that, conversely, what is disordered, chaotic, unpredictable only appears within a world of falsity or recognized as incomplete. (Nietzsche 2021)

To do so, the voxel-based modeling approach must be articulated with the skeleton-based and solid modeling techniques in order to be harmoniously integrated in the embodiment and detail design phases. Figure 4.10 illustrates the steps of the approach as follows:

- Definition of a design space (volume-envelope) – This step consists of modeling a design space or an active region to be designed with a volume in which the geometry must fit and also with potential skeleton elements and functional surfaces to interface with inert spatial regions and/or other product parts.

- Voxelization (discretization) – This step aims to cut the previously defined design space into equal-sized cubes (voxels), in order to be able to control the granularity/precision of the material distribution. As explained above, the voxels here are connected at their center by 3D beams. These beams then form a 3D lattice structure, acting as a control structure for the deformation of the geometry.

- *Material modeling* - Assuming that each voxel is homogeneous and made of a single isotropic linear material (or a perfectly homogeneous mixture), this step concerns the introduction of properties such as the Young's modulus (E) and the shear modulus (G) in a first step, other properties will be added according to the stimulus sensitive behavior of the selected material.

- Skeleton extraction – Once voxels have been assigned a material, their properties are directly linked with the beam-level properties. This inheritance is useful for calculating compound values from the connection of voxels carrying different materials. It is important to specify that the active materials involved are those whose response to a stimulus is either elongation or contraction (but not bending or folding at this time).

- Calculation of the skeleton (structure) deformation – This step is necessary to calculate the deformation of the lattice structure. First, a calculation will be performed to find out the degrees of freedom of the structure and then its deformation using the direct stiffness method (Okereke and Keates 2018).

- Deformation of the voxelized object - Once the deformation of the beam structure has been calculated, the voxels will be controlled using the "skinning" technique so that the shape can follow the structure; the deformation of the structure, in turn, follows the laws of physics.

In this way, this innovative approach, although composed of bricks already accepted by the mechanical/digital community, offers the advantage of federating several scientific communities belonging to the fields of digital mechanics, manufacturing and 3D computer graphics.



Figure 4.10. Voxel-based modeling and simulation of active materials (Sossou et al. 2019a, 2019b)

# 4.6.3. Distribution of active materials

Perhaps we still don't know everything about how ideas can act on ideas. (Rosset 1967)

Based on this material modeling approach, it becomes equally important to be able to explore relevant material distributions. From the literature, we learn, to this effect, that smart material distributions in structures are generally ad hoc, except for the quite original work of Maute et al. (2015) on a topological optimization approach (level-set method) for predicting the behavior and spatially arranging shape memory polymers within a passive (inert) material. In order to achieve a desired shape change (i.e. from an initial shape to a final shape), having a few materials and a stimulus, it is necessary to be able to determine a spatial distribution of materials that enables the desired change (see Figure 4.11).



Figure 4.11. Methodology for computing material distribution (Sossou et al. 2019b)

To do this, a three-step methodology - illustrated in Figure 4.11 - can be proposed as follows:

1) *Voxelization of the initial shape* – This step allows for voxelizing a shape corresponding to an initial state, that is a state resulting from a print. This step also includes the generation of a control structure made of points and planes. The points correspond to the centers of the voxels (or their centers of gravity), and the planes to

a median plane of the voxels. These elements are used to capture the degrees of freedom required for the desired deformation: points for displacements and planes for rotations. This enriched structure allows us to manually transform the initial shape into the desired shape to be reached by stimulation.

2) *Comparison* – This step consists of comparing the two voxelized models to obtain the degrees of freedom associated with the nodes of the control structure.

3) Material distribution computation – The distribution here is the result of an inverse problem. Given a desired deformation vector and a given stimulus, this step will determine the nature of the material of each voxel, in the manner of an integer linear optimization problem. The goal is to find a material distribution such that the resulting deformation produces a field of degrees of freedom close to that required for the desired shape change. In other words, a distribution that is such that when the initial shape is subjected to the stimulus, it deforms into a shape very close to (or even identical to) the desired final shape. To do this, the materials to be affected will be selected from a set of materials including a conventional (passive) material and a finite number of active materials. Thus, a computed distribution will be represented by a series of integers to inform the material allocated to each voxel. Figure 4.12 illustrates how voxels are materialized via the combination of two materials (#0 and #1). In order to determine a suitable distribution, an "objective" function must be optimized. In the proposed approach, the sum of the difference in squares between the calculated degrees of freedom (for a distribution) and the desired degrees of freedom was chosen. A distribution that minimizes this function will be suitable. As this function is not a linear function of a distribution, an evolutionary approach exploiting a genetic algorithm (where the genome is the material distribution and the genes are the inputs of the matrix representation) - may be relevant to solve this optimization problem. This type of approach is effective for solving nonlinear integer programs (Yokota et al. 1996).



**Figure 4.12.** Example of distributions of an active material (#1 in red) within an inert material (#0 in blue) (Sossou et al. 2019b)

True progress [...] always appearing, as it does, in the form of the will and way to *greater power* and always emerging victorious at the cost of countless smaller forces. The amount of "progress" can actually be *measured* according to how much has had to be sacrified to it. (Nietzsche 1994)

## 4.6.4. Distribution of active materials by integrating empty elements

The determination of material distributions is a crucial step in designing heterogeneous 4D printing behaviors. Whether they are predefined or numerical, the distributions bring freedom in design and must be defined according to the "capabilities" offered by the multi-material 3D printing processes. While the solutions generated may appear satisfactory in terms of the desired change in shape, attention must be paid to the still limited performance in terms of energy consumption and the use of active materials in the structures. It is then possible to couple the reasoning efforts with material shrinkage approaches as treated in the vast field of topological optimization. While the majority of the current work in additive manufacturing process, it is appropriate here to exploit this strategy to minimize the use of active materials and/or to facilitate the transformation (in the case of a change in shape in particular). This strategy – consisting of removing material that does not influence the mechanical stresses for example – can be exploited in two ways by:

- Developing new "topologically optimized" distributions for reuse in design.

- Determining, in the case of an inverse problem, a material distribution including empty elements.

The development of topologically optimized generic distributions can be operated by different simulation/optimization cycles. Figure 4.13 highlights optimized voxel-based distributions that can complement the known distributions adopted by the 4D printing community (see Volume 1, Introduction, Figure I.5). The proposed solutions have the advantage of minimizing the use of both materials, reducing the mass of the system while maintaining the desired curvature at constant stimulation. The example introduces a temperature sensitive hydrogel (E = 1 MPa, G = 0.3 MPa, LCST = 33.5°C, K = 2,  $v_{min} = 0.098$ ,  $v_{max} = 0.4$ ) and an inert polymer (E = 10 MPa, G = 3.7 MPa) distributed on a specimen of dimensions 27 mm × 7 mm × 2 mm. This initiative has real interest for the development of multi-material metamaterial structures and can be studied for gradient functionality distributions.



**Figure 4.13.** Development of bilayer and patterned distributions to provide a "bending" type transformation between a temperature sensitive hydrogel (red) and a passive polymer (blue)

In an inverse problem logic, the direct determination of numerical distributions is both an original and complex path. This research direction can be approached by topological optimization as some authors have done in the case of multi-material structures made by 3D printing (Hiller and Lipson 2009; Vaezi et al. 2013; Zuo and Saitou 2016; Sundaram et al. 2019; Wu et al. 2021). The goal here is to go beyond the search for mechanical performance at the static level but to improve the actuation of the heterogeneous 4D structure while minimizing the use of active materials. Recent efforts have been undertaken by the authors to integrate material removal directly into the determination of material distribution. This involves introducing either an extremely passive material or an "empty" material into the genetic algorithm from the work of Sossou et al. (2019a, 2019b), which consists of distributing three materials in the 3D structure intended to change shape under thermal stimulation. Figure 4.14 shows initial results exploiting the VoxSmart tool within the Rhinoceros3D/Grasshopper environment. Figure 4.14(a) shows a numerically distributed structure obtained by the genetic algorithm with an initial population of 150 and 450 generations (Figure 4.14(d)) and a structure incorporating patterns; the representation of the simulated structures shows extremely close results (Figure 4.13(b)), in perfect coherence with a "hollowed out" version (Figure 4.14(c)). The initial assumptions concerning the introduction of an extremely passive material to simulate empty elements are encouraging to distribute active, inert and empty materials within the same reasoning.



**Figure 4.14.** Determination of numerical distributions of materials incorporating void elements with the VoxSmart tool and comparison with a structure with a patterned distribution: a) voxelized structures with three materials (active in red, passive in blue, extremely passive in green) before stimulation; b) stimulated structures; and c) multimaterial structures integrating void elements. The convergence toward such results requires considering a population of 150 and a generation of 450

Confused notions have an advantage: they allow agreement on formulas to be reconciled with disagreement on their interpretation. (Perelman 1990)

#### 4.6.5. Additional scientific challenges

The transformation function to be designed and implemented and its required performance lead to problems of distributing multiple materials and related properties to the right place in the final geometric definition of the product/part. The implementation of a primitive behavior in a primitive shape can be done by using well-known distribution models shown in Figure I.5 of Volume 1's Introduction. However, such a numerical definition of the multi-material structure can only be established by first considering the constraints of the additive manufacturing process and transformation sequence. The selection and planning of additive manufacturing processes correspond to a well-known and well-addressed research question (Ghazy 2012; Wang et al. 2017), but multi-material 3D printing processes and techniques introduce compatibility issues regarding materials and distribution patterns that are intrinsically related to deposition capabilities (layer by layer or even volume by volume). This selection step must be performed according to the stimulus strategies. For example, if a stimulator is to be integrated into the structure to produce internal and local stimulation, the manufacturing "range" will have to take into account the integration of this element during printing or afterward by an assembly operation. The selection of the additive manufacturing process thus guides the design choices in terms of materials and their distribution in the structure. In such a context, many challenges may arise to address the multi-material/stimulus distribution, especially if a multi-scale modeling strategy is applied, and can be developed as follows:

– Multi-scale modeling (bottom-up, top-down and homogenization approaches) and multiphysics simulation of spatio-temporal phenomena in multi-material structures. The scales of application of 4D printing present different dynamics and temporalities. Therefore, extension of scale-specific models through homogenization approaches will play a role in coupling particle-based and continuum-based models. Achieving this long-term goal will contribute to the real-time simulation design of active/stimuli materials with quantitative prediction capabilities of their structure and properties (van der Giessen et al. 2020).

- Computation of the materials distribution in 3D design spaces (where active regions have been defined) at different scales (macro, micro and nano) on a voxel/octree basis so as to promote an inverse approach.

- Integrating multi-scale (continuous or discrete) topological optimization in multi-material structures coupled with parametric optimization (Wu et al. 2021; Zhu et al. 2021).

- Discovering material distribution models, metamaterials and multi-scale mechanisms based on connectionist AI (e.g. machine learning and deep neural networks).

- Considering bio-inspired generative design strategies for voxel-based structure definition. Possible scientific directions to voxel automata, Lindenmayer's growth system (L-systems) (Prusinkiewicz and Lindenmayer 1990), numerical morphogenesis, etc., would provide ways to ensure the growth of multi-scale geometric definition.

What its simplest expression summarizes to the hermeneutic unity of the veiling-unveiling: what shows us a thing is hidden while it shows itself, independently of any human subject. (Kacem 2004)



# 4.7. Digital chain for 4D design and prototyping

**Figure 4.15.** Proposed digital chain dedicated to the design and prototyping of 4D systems (Demoly et al. 2021)

The application of a design method also requires a dedicated digital chain (Kim et al. 2017; Singh and Wilcox 2018; Pang et al. 2021). Figure 4.15 illustrates a digital chain for building smart systems with 4D printing technology, which consists of systems engineering, product design, multiphysics modeling and simulation, manufacturing planning and process engineering. This chain makes sense if it is appropriately coupled with a FabLab 4D value chain, which starts from raw materials to combined manufacturing, assembly and 3D printing steps, followed by material training, stimulation and actuation steps. The digital chain can be implemented by connecting information systems and related tools covering both digital and physical models. To do so, a scientific challenge is to introduce a solid and active knowledge base associated with a digital twin so as to (1) improve

theoretical and simulation models from physical data; (2) ensure cyber-control of physical systems; and also (3) enable a seamless reconciliation between the two worlds through feedback and backward information flows.

When something becomes dismantled, one can support it [...]. But to undertake to recast such a great mass and to change the foundations of such a great building, is the business of those who, in order to clean [a painting], erase it, who want to amend particular defects by a universal confusion and to cure diseases by death. (Comte-Sponville 2020)

#### 4.8. Claims and practical constraints

In all that has just been shown, we note that a general approach to modeling a 4D structure can be envisaged. However, if the bricks are beginning to stabilize, the complexity of 4D systems with the transformations induced by energy stimuli still limits this field of full development (and which, in any case, will be essential in the future if we hope to see 4D printing reach industrial environments). Several considerations aiming at a satisfactory robustness must be taken into account:

- The satisfactory realization of a 3D object from active and passive elements. For example, Yang et al. (2021) recall what quality requirements are claimed for traditional additive manufacturing.

- Particular specificities linked to the evolutionary nature of objects such as cohesions between voxels, functional fatigue aspects of materials stressed by stimuli and aging. The spatio-temporal effects of the stimuli must also be taken into account by the modelers, etc. These different elements have been described throughout the body of this text and, along with others, add complexity to what has just been proposed.

However, as it has already been pointed out, 4D technology is recent and has yet to find its place; its whole place. The idea of attacking the development of 4D printing with different methods is undoubtedly a good way to achieve this goal.

We must consider the present state of the universe as the effect of its previous state and as the cause of what will follow. An intelligence which [...] would recognize all the forces of which nature is animated and the respective situations of the beings which compose it, would embrace in the same formula the movements of the largest bodies in the universe and those of the lightest atom: nothing would be uncertain for it, and the future, as well as the past, would be present to its eyes. (Laplace 2020)

### 4.9. Conclusion

Over the past decade, 4D printing has attracted greatly increased academic interest, initially from researchers in the fields of materials science, process science and mechanics. Given the large number of proofs-of-concept proposed in the literature, it is clear that this technology – highly interdisciplinary – offers greater freedom (and maybe too much freedom) to build structures capable of evolving over time under energy stimulation.

This tangible benefit – which is characterized by the functional increase over time via consolidated parts – brings about complexities and paradigm shifts in design. This chapter was conducted to establish a long-term scientific roadmap dedicated to "Design for 4D Printing". Overall, the opportunities and challenges have been highlighted and developed in a way that initiates the transition of this emerging technology to an industrial threshold. In its current form, 4D printing has useful capabilities to play a crucial role in the Industry 4.0 era, and also introduces new usage scenarios that deserve to be explored in terms of human factors and experience for the adoption of stimuli-responsive structures in society.

Time, is a three-fold present: the present as we experience it, past as a present memory, and a future as a present expectation (Kant). By that criterion, the world of the year 2025 has already arrived, for in the decisions we make now, in the way we design our environment and thus sketch the lines of constraints, the future is committed. (Dutton 1996)

A scientific discovery, or the most systematic, or the most seemingly universal of new theories, will apply to some immediate element of novelty or antinomianism in an enormous mass of tacitly accepted knowledge, experience, beliefs and presuppositions. Our progress is thin, it leaves intact an immense universe and assumes it to be accepted. (Oppenheimer 1955)

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# Conclusion

# A Conclusion Without a Real "Roadmap"

A modest start (artisanal) to finally arrive at an entry into a fully constructed universe, freed from its constraints. It is, according to us, the very trajectory of creation. [...] Man thus invents his world. The machine participates directly in creation: not only does it accelerate the speed of manufacture, not only does it ensure the passage of the negligible (water, fire) to useful energy, but its mechanical power cannot be compared to the neuromuscular forces of the workers. Therefore, thanks to it, we note prodigious increases. (Dagognet 1995)

# FOUR STORIES TO START:

- 1) Nokia rejected Android.
- 2) Yahoo rejected Google.
- 3) Kodak refused digital cameras.
- 4) Blockbuster rejected Netflix.

# LESSONS:

- 1) Take risks.
- 2) Accept change.
- 3) If you refuse to change over time, you will become obsolete.

# TWO OTHER STORIES:

- 1) Facebook takes over WhatsApp and Instagram.
- 2) Grab takes over Uber in Southeast Asia.

4D Printing 2: Between Science and Technology,

For a color version of all the figures in this chapter, see: www.iste.co.uk/demoly/4Dprinting2.zip.

First Edition. Frédéric Demoly and Jean-Claude André.

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# LESSONS:

1) Become so powerful that your competitors become your allies.

2) Reach the leading position and work intelligently with the competition.

3) Continue to innovate.

# TWO OTHER STORIES:

1) Colonel Sanders founded KFC at the age of 65 years.

2) Jack Ma, who could not find a job at KFC, founded Alibaba and retired at 55 years old.

#### LESSONS:

1) Age is just a number.

2) Only those who keep trying will succeed.

# LASTLY BUT BY NO MEANS THE LEAST:

- Lamborghini was founded as a result of the revenge of a tractor owner who was insulted by Enzo Ferrari, the founder of Ferrari.

# LESSONS:

- Never underestimate anyone, ever!
- Keep working hard. Invest your time wisely.
- Do not be afraid to fail.
- Let us go!

A few decades ago, there was a very simple method to make the prospective experts of "France Telecom" of the time laugh: it was to speak about the cell phone. For them, it was science fiction. Neither data compression nor the miniaturization of computing was conceivable at the time. (Klein 2016)

We adhere to a kind of "aesthetics of the moment", we want to seize the mythical moment when one passes from the misunderstood to the understood, to detect the lightning germination of the revolutionary idea. (Klein 1994)

# Warning

4D printing is a technology whose mission should be to provide society with fruitful applications that it will appropriate if they are profitable and/or desirable. In this sense, science must allow this blossoming (and not only satisfy the production of egocentric publications). This being the case, before structuring a community around a structured "roadmap", both coming from Descartes Country, it seemed to us, in this work, opportune to know where we can and want to go, why and how, with, as far as possible, long-term visions (foresight or forecasting including designers, dreamers and engineers). It seems, however, that 4D printing has a real ascensional power (in the sense of Bachelard). This is what we have tried to produce in this book by illustrating possible fields of research, by proposing to limit others, etc. A basic robust roadmap for the short term should take into account the real and the possible by associating all the essential scientific disciplines that can participate in this necessary success (through the convergence of scientific knowledge and interdisciplinary projects).

However, it is still necessary to bring together research results that are still scattered to integrate them into a set that is as coherent as possible, to imagine applications that carry meaning (and economic market) to constitute a new source of imagination, with the stabilization of the myth created in 2013 by Skylar Tibbits. It is therefore already through some successes that new promises can emerge, and, by passing through fiction, that new narratives can be proposed. It is true that the cultural baggage that should be used to move forward on this buoyant, spectacular and original theme, associated with our vision of the future are priority elements for any emerging techno-scientific activity. But, in the present situation, it is not a question of getting on a moving train, but of knowing where it is going and why. Thus, these restrictive comments do not allow a stable scientific progression to be defined, this must be co-constructed by associating the time of exchange and debate for interdisciplinary works to be carried out.

# C.1. Introduction

Small seas do not require the same sailors as large oceans. (Serres 1985)

What degree of reality can we [...] attribute to things that are born and die, that all do and undo? (D'Espagnat 2015)

This book gives arguments that strategic intelligence devices do not contribute in human society and means on the one hand, and do not make the success of radical innovations reliable on the other hand. But it opens up avenues for reflection and possible actions. The challenge of "moving" the lines is crucial both for those who are committed to renewal, as has been shown, in a difficult but promising way, and for those who are not ready because of the important changes that will be initiated by others. This type of phenomenon is particularly problematic for all authorities, private or public, who find it difficult to question their knowledge and experience, especially when these form the basis of their activities and success (Blanco 2008). But the 4D adventure could/should be associated with a capacity to anticipate change (prepare for it and identify decisions to act upon).

However, "emerging" technologies such as 4D printing are booming, growing and transforming at a rapid pace with very high growth rates. These "neotechnologies" are, as we have seen, part of a whole with shifting outlines, but with high potential fields: digital sciences and technologies, engineering sciences, material sciences, biology (for bio-printing), etc. With an image of a "clean" process associated with 3D printing (André 2020), we can also associate a dynamic high-tech aspect with a sustainable image. But we still have a long way to go before we win! What this joint work shows is that it is not possible to locate precisely the origin of the "disruptive" character of this concept (apart from the "breath" introduced by Tibbits (2013)). What is also shown is that the different technologies involved in 4D development feed off each other, and in their interdependencies, their joint effects should exceed that which they would have had separately (hence the concept of convergence). Within this same framework of all-out dissemination, they envisage the penetration of numerous application markets likely to transform them. In this sense, we could say that they are both convergent and divergent, extending their branches in all directions (Andler 2020).

While additive manufacturing is often presented as likely to radically transform the way objects are designed as well as produced, the organization of the productive system, its large-scale dissemination still remains largely dependent on a significant number of technical advances whose speed is difficult to anticipate. At present, machines and processes do not meet all the constraints of industrial production and progress should be made in particular to accelerate the rate of production. (COE 2017)

At the same time, the arrival of a technology that eliminates many design steps by providing a tool that is positioned (in principle) between the computer and the object puts it back in the hands of designers and "makers" who should be able to make innovative and otherwise inaccessible objects "in their garages" (as long as the chemical materials are readily available). The expansion of this new relationship between thought and tool may lead to cultural evolutions in the act of production with the desired aspects of reintegration of certain productions on the national territory and possible uberization of activities (with low-cost machines). What is partly true for 3D printing with the concept of the "FabLab" (Gerschenfeld 2005; André 2017a, 2017b, 2017c) may, given the youth and operative complexity of 4D, be called into question, at least for the moment. The components of 4D printing should still progress/evolve from both the scientific and technological (and perhaps social) perspectives.

Engineering sciences and the disciplines that contribute to their development are not static data, from which the question of what scientific knowledge represents could be asked, but a convergence of integrable knowledge in view of operative ends whose manifestations are visible. To reach this objective, there is an alliance between uniform research models and experiments, the only current ways to master these needs. For all that, this stabilized form of access to the intelligibility of new knowledge must not eliminate the power of creative invention to imagine and then explore less usual phenomena, even that considered today by many members of the body of academic research as exotic, or even for some, without interest, because they are located outside their mono-disciplinary spectrum of competence. This creativity, which is essential to the mobilization of researchers motivated by divergent thinking and collective approaches, must however respect the arrow of time, the breaks in symmetry between the before and after, with a constant reminder of the second principle of thermodynamics, and therefore of entropy and irreversible processes (Clausius), synonymous with forms of impotence in the fight against degradation processes. It is this Sisyphean struggle that is the spice of shared disruptive operations.

Scientific, technoscience and engineering disciplines increasingly shape the emergence of new technologies, as was the case with 3D printing in 1984 (André et al. 1984) and to an extent with 4D printing (Tibbits 2013). But, as we have seen, disciplines holding a robust "paradigmatic dogma" have a conservative character acting as a flywheel, with institutions (such as university departments and so-called "learned" scientific societies) and pedagogy (textbooks and curricula) designed to propagate, and also perpetuate, existing approaches. In the case of 3D printing, the first publication on the subject was rejected because the concept was not stabilized or accepted. This situation has changed a little since then.

For CSET (2021), a similar disciplinary situation emerged for missile defense in the 1970s and 1980s, when physicists and software engineers viewed its prospects from different angles (Slayton 2013). To change the metaphor, scientific and engineering disciplines can provide a kind of friction, an additional resistive force that emerging technologies must overcome. Paying close attention to this tension could help ensure that established disciplinary interests do not impede the development of new technologies.

However, in a country that operates on the basis of five-year plans, such as France, or on a slightly different time frame, such as the European Union, initiatives do not come from decision-makers who commit their countries to a strategic vision, nor from users who could be the first to use the new technology. Rather, the initial impetus comes from motivated middle-ranking individuals. Only after accepting a possible fantasy associated with actual success, can senior leaders sustain a program. If it takes time to pass through these different filtering levels and reach political decision-makers, the descent in the form of action programs (ANR in France or Horizon Europe for the EU) follows a similar filtering path. Indeed, it is necessary to convince the program managers who mediate between science and technology on the one hand and strategy and policy on the other hand. They are often slow to translate technological opportunities and challenges into economic and social strategy and policy (and vice versa). However, if the message is well conveyed, it should not take longer than the exploration of the theme to reap the benefits in terms of competitiveness. This is what can be asked as a basic question for these emerging technologies.

While historians have long recognized that many technologies have international roots (Mokyr 1990), while it is not always possible to access literature in English (or in French), the siloed nature of science and technology may be responsible for innovations not being taken into account. Such was the case of the tracking of submarines through the infra-sound waves they emitted and which were observed by oil companies looking for oil slicks to exploit... or of the theory of wavelets developed by geologists and not analyzed by information processing specialists, constituting another community, that of signal processing. Another difficulty is thus revealed. "Scenarios allow us to broaden our mental boundaries and to develop a greater openness to new knowledge. They are multidimensional (...). They are an interdisciplinary and multicultural exercise" (Barbieri-Masini 2000). One of the possible difficulties of the exercise is to "arbitrarily" multiply the possibilities of disruption, in defiance of the knowledge of the forces at work, the other being to reason about a world of predictable and therefore reassuring continuity, by concealing the disruptions, if only with the help of oratory precautions.

In order to think of positive futures from a technological point of view, having both an attractive force and a credibility that does not relegate them to utopia, nor to "reassuring" conservative continuity or to ordinary, vaguely unhealthy lobbying, an approach that combines the potential of science fiction, the rigor of foresight and the collective and shared dimension of participatory production would seem desirable today (Luck and Aubert 2021). Foresight is based on rigorous methods and is "serious", science fiction, synonymous with the improbable, the impossible, is more fanciful. But we did not find any media that mention 4D printing, except for "*Terminator 2*", which is far too far from a future that is possible today. The survey carried out on the possible future of 4D printing did not allow us, for various

reasons, to go very far even if we had thought to leave a large place to creativity, in a participative approach which could arouse the adhesion and multiply the possible visions of futures opened by 4D printing. The foresight exercise was not easy, the exploration of new methods such as this survey could broaden and enrich the public's ability to help academic research free itself from the reductionist reflexes to which practice forces it on a daily basis (Saives et al. 2019).

# C.2. The myth of 4D printing

Great things are done by a series of small things brought together. (Van Gogh 2020)

[Art]: species of representations which have their end in themselves and yet, without any other purpose, promote the cultivation of the faculties of the mind in their relation to social life. (Kant 2015)

Engineers tended to separate a system into sub-assemblies with Cartesian logic. Those were the happy days when you could repair your Citroën 2CV with bits of string and office paper clips. If, in the mind of designers, this division of tasks or functions remains present at all stages of the design of an industrial artifact, the physical realization increasingly integrates diversified knowledge included in increasingly black boxes (Duhart 2021). The development of digital sciences has been one of the culprits of this de facto confinement. To make up for this, we can write that we can imitate nature and that thanks to the virtues of bio-mimicry, designers now know how to integrate decision-making systems, sensors and actuators, intelligence, etc. (up to commanding a machine by thought? according to MCE (2021)). Is this not a bit like what we could try to do with 4D printing?

Independently of this desire for integration, for just under 10 years sciences and technologies have tended by "classical" nucleation to gather together in a stimulating association of people committed to the future of 4D; this is structured and becomes the bearer of methods and methodological habits likely to give strength, but also to make the field lose part of its richness and its initiatives. Does this form of unity, of cohesion, lead to a community sharing an ethos, an exemplary know-how? Or, should we, having 3D and/or 4D know-how, think about the positioning of these scientific technologies for the realization of tools, gadgets or applicable devices?

The field of exploration of smart or active materials involves using, in a clever and imaginative way, materials or stimulable materials in which all the elements can (or will be able to) be directly integrated. There is also the question of the elegance sought in the energy stimulation of active materials... (but Chapter 1, which should have been devoted to this aspect, was very short, due to the absence of credible elements to present to the readers). Many targets can be envisaged: varied and evolving mechanical properties, digital and electronic circuits revisited, new types of energy for motorization and actuation, adaptive chemical reactors, drug delivery, direct brain-matter communication, adaptive deformable materials, etc. The list is even longer as all dimensions of space can be covered and there is a strong demand for soft robotics, medical applications, home automation, etc.

Everything is still to be done and it sticks to us like a band-aid on an index finger or on the thumb of Captain Haddock in *Tintin and Snowy* drawn by Hergé... We do not get rid of it just like that, one small problem solved chases the other without being able to master the immense complexity associated with this emerging field with its fantastic openings, but always far from robust and industrial results. Inspired by the Palo-Alto School, cannot we go beyond the indispensable creativity (defined as a capacity to change one's perception of the world) to a true innovation capable of changing the reality of this world?

## C.3. What can we do?

The main principles of startups, which are becoming the laws of the new economy, are *a priori* contrary to the image we have of established champions: to have a revolutionary idea, to do simple, to do fast, to operate in small teams, to have eccentric, out-of-the-box leaders. (Beffa 2017)

From the point of view of the authors, it is a question of respecting the opinion of Jürgen Habermas (1968) on the integration of Science-Society; he wrote: "The real problem is to know if, once a certain level of knowledge is reached, one is satisfied to put it at the disposal of Men occupied with technical manipulations or if one wants them to be citizens communicating with one another so that they again take possession of their own language". It is somewhat on these bases that we discussed a principle of co-construction representing a fruitful alliance between applicant and designer in 4D printing. For us, this mode of action avoids "autism", withdrawal into oneself, or even self-citation where the researcher risks becoming their own tautology (Supiot 2006). This remark therefore provides a way of developing the culture of researchers so that they take into consideration the fact that they can be influential members (but not more so) of society, that their direct or indirect technological contributions can change and shape it (Hacking 2001). But for all that, the prospective thinking or the potential of a science fiction story through its scenaristic nature and its reference to social myths are generally missing elements to try to convince decision-makers. Science fiction stories can indeed serve as an amplifier of a society's fears and hopes for its future, but this is outside the normal

activity of the researcher. A proactive relay is therefore necessary. The current situation of scientific writings reveals a form of unthinking, of randomness (but in 4D printing, with a potential strongly oriented toward polymers) with defects of anticipation and mobilization on the construction of the technology-oriented future, which supposes to better discern the serious and emerging trends, the discontinuities and the disruptions. For Chognot (2021), the foresight approach makes it possible to overcome inertia or determinism and to mobilize a community for a chosen future. She writes: "By opening up the frameworks of analysis, conceiving the alternative, and choosing an aim, it reveals itself as fundamentally political".

Normally, the researcher is not an isolated castaway on a desert island. They are members of a large research organization or a university. They are trained and have means at their disposal. However, these organizations are increasingly concerned with economic development, in parallel with their traditional missions of teaching and research. Commercialization can take many forms: technology licensing to established companies, startups, etc. (NBER 2021). But, the technologies developed in university laboratories are generally more embryonic than those of their industrial counterparts (Jensen and Thursby 2001). In the absence of the creation of new companies, there is a risk that these discoveries will not be commercialized and will not be perceived, unless a responsible and motivated choice is made on the part of the supervisory bodies that support them, as representing a technological advance that could contribute to the competitiveness of national or European companies.

From the leader's point of view, the startup looks like a troublemaker, an agitator to be chased away quickly, a small annoying voice to be stifled quickly. (Beffa 2017)

But, to make the problem of the robust emergence of the 4D domain even more complex, it is a question of investing in the aspects of interdisciplinary and inter-technological convergences, the difficulty of which is recognized by all. Indeed, if we place ourselves in a broader evolutionary perspective, as has been proposed, it does not seem possible to be able to predict today the evolution of the production of 4D printing works by deducing them from the application of simple laws that we could have identified, and this being at different scales (not even from the resolution of generic problems). Note that companies also use the term "coalescence", defined as the ability to grow faster by being together. But words and semantics may be of modest use to the development of the field.

For a long time now, science has been divided into disciplines, sometimes operating in watertight silos. Vigoureux wrote in 2020 that "until the final year of high school, [he] thought of science as a French garden where all the paths were marked out". We have thus added to the silos, a knowledge raised to the rank of intangible, sclerotic, predictable knowledge! The walls are therefore thick... And

then, the creative person is a generally worrying character who would have advantages that the others do not have. Intuitive, no doubt, but certainly presumptuous! Moreover, they are difficult to manage because they are too independent and everyone recognizes that, while they have ideas, they can also change them a lot... The litany is long and quickly dries up any debate. How, in this sometimes deleterious context where trust is modestly present, to pay attention to the beginnings of ideas, to the signs revealing something unexpected. How can we then help chance?

Rationality imposes only rather loose constraints on half-understood ideas: their internal coherence and mutual evolution cannot be assessed. (Sperber 1996)

However, on the knowledge front, we realize that when dealing with boundary objects, with inverse problems that lead to innovation or invention, it is necessary to pool knowledge from several disciplines and several origins while accepting that they should be questioned. This knowledge has its own paradigms, its own dynamics, its own sensitivities, its own ways of working and its own specific evaluation mechanisms. It is possible to show that in order to escape quasiprofessional incremental research, a certain amount of courage (or innocence) is required because it corresponds to operations that are ultimately very difficult to put in place, without being sure that success will be at the conclusion of the project.

This is what Roach (2021) recently produced in his thesis, in which he considers that 3D printing of adaptive materials should allow, as a first priority, innovative applications that are difficult to achieve with conventional manufacturing processes. However, the potential applications of 4D printing go far beyond a simple change of shape with active structures using metamaterials capable of changing shape (self-supporting tents, flexible micro-robots, drug delivery, etc.). He reminds us that other properties sensitive to energy stimuli, such as self-repair and self-sensing, are beginning to be integrated into 3D/4D printed structures. Other advances such as color-changing textiles that can help individuals hide in certain environments or computing to monitor health. What is interesting is that there are individual initiatives in the 4D field (even if in this bottom-up logic, they are free and spontaneous), initiatives that it could be useful to federate to try to go a little further in terms of disruptive research proposals.

Contrary to the great change that followed the Industrial Revolution during the 20th century (on which research, to a greater or lesser extent, continues to be successfully carried out), it is no longer the states that are at the heart of the process (even if they finance research), but increasingly the consumers. They are the ones who decide to adopt or reject new products or new service models. In this sense, they are "consum'actors". They are in fact the main actors of the Industrial

Revolution 4.0 (André 2019) that we are experiencing: the revolution of uses. This new era feeds on previous revolutions: that of mechanization, energy and more recently that of digital technology. It is amplified by the connected mobility of objects and people, but relies on tangible elements hidden from them (materials, processes, energy).

Among the slogans associated with the future of our consumerist society, we can find expressions such as: "Data is the new oil"; "Artificial intelligence is the new electricity" and the "Internet of Things is the new nervous system"; "Technology is exponential"... (Leonhard 2018). Everything is therefore possible: "We just need to agree on what we want"... There are, to recall some obvious facts, only social and environmental reservations to this future that prepares us (if we believe in it) for Trans-humanism, for the Augmented Human and for digital immortality. This is easily allowed especially if we have some ideas, if possible, disruptive. Fortunately, some people have a position closer to reality and rely on the knowledge of the real and the possible to make realistic predictions (which remain deeply technological). But, between the inertia of traditional processes and the explosion of digital technologies, there is undoubtedly a need to define areas of innovation that could make a link between "tradition and modernity" within the framework of mastery, under constraint, of the exploitation of digital technologies in order to enter into the realm of concreteness based on the spatial mastery of the transformation of matter.

The fact remains that such anticipations cannot simply consist of calculations, precisely because reason is a motive, i.e. an object of desire, and not merely a consequence of the present. (Benyls and Harlan 1994)

On the other hand, in this same context and in a pre-paradigmatic creative phase, the social and economic realities of additive manufacturing were quickly decisive, leading to a cultural body today in the process of a certain stabilization, a body that has a small recognition within the disciplinary scientific system insofar as the field has defined its "doctrine body", reducing by (necessary) disjunction of many disciplines that could contribute to its development. While it is not seeking, in a determined way, to confront "recalcitrant" problems, the field remains alive, as long as it keeps an "acceptable" contact with the anticipation of the needs of society (in different ways), which poses the delicate question of the voluntary exit from the habits perpetuated by the "system", its social organization, its ends. But this is not yet the case for 4D manufacturing, an emerging field that is still fragile. The question of a post-mortem survival not anticipated toward the opening of new fields is however posed in a world that supports a posteriori risk-taking, creativity and disruptions too much, but that does not hesitate to pick them up again with ease.

The economic success of 3D printing is a beacon that attracts (somewhat a posteriori) funding for the improvement of methods, processes, materials, etc. In

this dynamic, it is partly up to the academic world to provide the socio-economic and technological development sought at a lower cost. So, in this situation, science is not asked to produce new disruptive ideas (as in the case of 4D printing in particular, and even less to produce eccentric ideas or ideas considered as such on new uses of informed or activatable matter), but to follow a demand coming mainly from those in power who are listening to users. At the same time, the system is segmented into macro-regions within which horizontal relations are dense or, on the contrary, distant due to a lack of adequate organizational fluidity. In fact, the necessary relations are not horizontal, they remain subject to domination and subordination (Olechnicka et al. 2019)... But it is still to be undertaken!

#### C.4. A look back at a non-draft roadmap for 4D printing

Utopian or dystopian, we find ourselves safe in a (supposedly!) virtual environment. We can therefore disregard the limits imposed by reality! (ATD 2020)

Life can only be understood backwards; but it must be lived forwards. (Kierkegaard 1988)

In Genesis, the devil says to Adam and Eve, "For God knows that in the day you eat of it, your eyes will be opened and you will be like God, knowing good and evil". This sentence quoted by Korn et al. (2008) clearly illustrates our unfulfilled but perennial desire to surpass ourselves, but never without knowing where we place the cursor between good and evil! Has the "tempter" fooled us? In a socially stable world, it would have been possible to begin this chapter and, no doubt, to end it with this sentence by Desroches et al. (2003): "Acceptable risk is the value of a risk resulting from an explicit decision established objectively by comparison with known and accepted natural or technological risks in certain branches of activity". In a framework of obvious uncertainty, many avenues can be exploited to pursue the promotion of technological progress: from social debate to the shaping of citizens, from democracy to the "best of all worlds"... We can therefore imagine a fundamental difficulty linked to a difficult interaction between science and technology on the one hand, and positive and negative effects for the public on the other hand, an interaction that reinforces the responsible dimension of research. Who, in this context, is responsible for defining the "Good"?

Starting from an important applicative potentiality, Verchère-Morice (2006) writes that it is not easily conceivable today to impose *de facto* applications of nanotechnologies on citizens, without resistance and without negotiations. She writes that "it is thus unlikely that 'small objects' will be massively incorporated into individuals, without there being 'pockets of acceptance' that allow a

dissemination and a trivialization of these forms of use within society". However, more than 1,000 "nano" products are already being manufactured. This illustrates rather well a traditional hiatus between research and the everyday reality, between what is offered and what is accepted, perhaps without knowing it... But, we have not yet controlled, nor "increased" Humankind by this means.

These few sentences, quite interesting for the exploration of the theme of acceptability, or even desirability, lead the authors to ask the question of what the acceptable represents, of its temporal evolution linked to a certain form of "mental plasticity of the social body", or even the opposite, but for the same goal to be reached, of a "shaping" of people, of a displacement by populism of "social tectonics" (Minc 2005) or of a status quo of the utilitarian morality, of the generation of crises by the contribution of true/false new knowledge (management of controversies), etc. Prigogine and Stengers (1986) contrast this notion of "plasticity", which suggests a form of flow under constraint, with complexity. However, our socio-technical systems, with their numerous changes of scale, are constantly becoming more complex. Under these conditions, according to these authors, the system associated with apparently deterministic technical progress offers numerous possibilities of bifurcation and destabilization, leading to possible crises. In addition to the shaping of the acceptable, there is the reaction of certain militant social groups under conditions that are not totally predictable. According to Benasayag and Sztulwark (2002), the cultural crisis that society is undergoing is based on the difficulty of differentiating between the "thinkable" and the "possible". They write that "not everything that is possible is necessarily thinkable". However, neoliberalism, that is to say the society of the individual, claims that in the name of economic profit, everything that is possible is thinkable. It is in this context that a classification criterion must be found, essential in a society committed to continuous technological progress, that of confidence, associating with a symbolic knowledge and belief system (Simmel 1996).

Independently of just the scientific aspects, it is indeed a question of leaving the perpetuated habits of scientific onanism to make original boundary objects emerge which could constitute elements of convergence of interest, of demonstration of the possibility of a true interdisciplinarity inside the 4D domain, of a new reference of innovation of disruption on real artifacts, of support to the divergent thought using 4D as a basis of demonstration. It is indeed necessary to go further. It is naturally not envisaged to address in this project these conservative visions, but to show that taking into account the original questions resulting from various forms of scientific and technological research or from expressions of social demand are likely to bring about, by desired proactivity, the local research from its now too traditional role of competent follower. This desire leads us to modify our relational modes in order to focus our research work on innovation that will bring about the future and

resourcefulness by engaging in risky operations, a condition linked to the nature of the disruption.

To be successful in discovery, you have to have improbable approaches. If you think like everyone else, if you bring people who have the same way of thinking together to discover something, the chance of discovering something is very low [...]. Discovery is as beautiful as the chance meeting on a dissection table of a sewing machine and an umbrella, that is, something improbable. (Ducasse 2011)

In this book, we have reported (several times) on several ways of making 4D objects based on homogeneous, hybrid and/or heterogeneous principles recalled in Figure C.1.



Figure C.1. Fourth reminder on 4D printing processes (not taking into account the stimulation aspects)

We have shown that the sole focus on the two homogeneous and hybrid aspects was not able to cover all the application needs (but that, on the other hand, it allowed the constitution of a prolific scientific community with rates of increase in the number of self-positioned publications of more than 40% per year). However, the promotional work by Tibbits (2013) was remarkable in creating a founding myth that made one forget the less imaginative attempts in the 1990s to realize industrial actuators (which met more easily industrializable criteria) using heterogeneous technologies (e.g. Ballandras et al. (1996)). His perfectly successful account was able to provoke a feeling of familiar strangeness that gave the impression that the 4D future was possible, attractive because it was spectacular, accessible and close, if not almost familiar. It was not a question of science fiction with real breakthroughs, but a dreamy presentation of a technology that was accessible and widely expected to succeed.

"Either the future is presented attractively, and then it is not credible, or it is presented credibly, and then it is not attractive," Klein (2021) recently wrote. Tibbits has succeeded in creating an imaginary that is both desirable and potentially credible! In his presentation it was a quirky but not overly close proximity, which allowed him to criticize the case just to enrich it and ultimately promote it. The result is obvious. New functions could be integrated, more than 30 years ago, to 3D objects by adding components or features. But, it is true that the associated more laborious aspect, less spectacular, less sensitive for a decision-maker in a hurry than the magic of the 4D movement inaugurated by Tibbits could make us forget for a few decades this field of possibilities allowing 4D printing to find its applicable potential market... or at least widen it!

"The more a market society mediatizes itself, the more it must devote a significant part of its activity to the production of demand, investing ever greater resources in attention-grabbing devices" (Citton 2014). But this demand does not have to be produced by the economic community; it can result from an approach that is closer to the end users, which would bring science, technology and society closer together and lead the research community to a more responsible approach (André 2013). The European Union in its H2020 program recommended the application of the following criteria regarding innovation (Mazzucato 2018):

- Inspiration and boldness, which in turn bring relevance and a major contribution to society.

- Ambition, with realistic and innovative research actions.
- Fostering interdisciplinary and cross-sectoral innovation.
- The establishment of targeted, measurable and time-bound guidelines.
- Implementation of multiple solutions with a bottom-up approach.

Our own brains harbor two competing systems. One is an exhibitionist incentive to do things; to create; to express. The other is an inhibitor, the critical alter-ego that questions, weighs and criticizes our ideas. To embark on a new enterprise, we must strike a careful balance between them. (du Sautoy 2020)

The designer who has explored (with inspiration and audacity) and deepened their initial idea can put forward a certain number of hypotheses on the elements of their perception of reality with a projection toward its use as long as they have an adapted vision (with a minimum of sharing). However, we have shown that the number of potential applications of 4D printing was already particularly large, which strongly limits the purpose. Success after success should develop other applications to be explored. If one is able to consider the proposal of a draft of use (let us recall the already old example of the Minitel with its unenvisaged detour at the beginning toward the pink Minitel...), one does not have all the assets... But as long as a pilot study is not around, the project remains at the state of a chimera. Thus, at least two actors must be taken into account: designer and user.

We must constantly go back and forth between the designer and the user, between the designer's user-project and the real user, between the world inscribed in the object and the world described by its movement [...]. It is the user's reactions that give content to the designer's project, just as the user's real environment is partly specified by the introduction of a new device. (Akrich 1987)

Figure C.2, extracted from a television program (Arte 2021), illustrates the reality of sensitive subjects (global warming), the existence of well-separated populations (but which can be satisfied with numerous cognitive biases), each defending a more or less ideological point of view or, to put it another way, more or less rational. The approximation does not seem to be an easy phenomenon (the majority of opinions have no reason to be right; see the Galileo syndrome) and yet, what we show is, as far as 4D printing is concerned, a need for approximation between scientific communities (but not only) that are currently too disjointed to favor innovation, too concentrated on quasi-individual activities. The change of scale therefore seems to be a necessity, but for many reasons corresponds to a very complicated path.



Figure C.2. Disjunctions between populations using social networks on the aspect of global warming

Unexpected events cause reality not to follow the envisioned sequences, outcomes not to be as expected, and individuals to take too long to recognize that their predictions are being dashed and the increasingly critical problem is emerging. (van der Veen and Wakkee 2004)

On another, more disruptive register, there is the idea (which should really be explored) related to the possibility of moving toward learning 4D printing. So, rather than imposing a clear, programmatic, long-term strategy, can we not simply support operations where applicants and designers would be enthusiastic about sharing, mutualization and a little less about mono-disciplinary mutilation to find disruptive ways out of the usual? It is better to dream together, especially if the results illustrate the interest of sharing!

In sum, as Richard Feynman predicted in 1959, the era of "Smaller, Faster, Cheaper" whose initial idea was aimed at nanotechnology and nanoscience (manipulating matter at the atomic and molecular scale in order to design and realize sub-micrometer components and systems) could be applied to 4D printing. If immense promises can be associated with this integrative concept, the population of researchers who can exploit their creativity and their time is however not extensible. Although it is not possible to know the size of this emerging body of research (around a thousand?), the number of applications (non-exhaustive) demonstrated by the existence of recent publications is too high (more than 200) for specialists in the field to operate solely on the basis of ("bottom-up") supply. There are reasons behind the campaign in favor of a "top-down" approach, that of the interdisciplinary sharing mentioned above, but above all that of reaching, through a convergence of interests, exemplary successes that can be industrialized, which require an alliance between the applicant(s) and the designer(s) (and other forms of reasoning). It is from successful and communicable examples that we will be able to train young people involved in the promotion of this very open field and that we will be able to remember Richard Feynman's prediction (without forgetting his humility and his humor). But, this will probably be for later...

Intelligence generally designates the cognitive resources that allow any living being to adapt to its environment or, on the contrary, to modify its environment through invention in order to adapt it fruitfully to the needs (or demands) of society. Everyone can see, even in their own disciplinary silo, that it is deployed in many ways in intellectual and professional fields, etc., in order to meet specialized objectives. Without wishing to intervene in the scientific background avoiding an irrepressible need to categorize the phenomena, it would undoubtedly be better to speak of a spectrum of talents and abilities than of multiple intelligences; but then, if to identify them more closely, instead of expressing ourselves in terms of multiple intelligences or not (Gardner 1983, 1998), why not take into consideration all the qualities of training, mutualization, exchanges, trust coming from the other elements of the social body in order to move forward?

Leblanc (2017) wrote: "Why is the robot scary for jobs, while the 3D printer is a dream? [...] The 3D printer makes everyone dream, because it appears at a time when consumers are tired of throwing away and buying back". This is certainly a simplistic image, but it is useful because it presents a recent technology in a positive light (this is probably not as true if we look deeper – see André (2017a, 2017b, 2017c, 2020)). Let us hope that together we can create a collective imagination that is just as useful and desirable by using 3D printing to promote one of its "daughter" or "son" domains, 4D printing!

# C.5. Resuming the campaign to go further together?

Socrates knows well that to be right alone against everyone is not to be right, but to be wrong, to be mad. (Lyotard 2012)

Many grant submission and review practices discourage researchers from submitting high-risk, high-return proposals due to an inherent conservatism in the review process, a tendency that becomes more problematic as competition increases. Many practices have been used by granting agencies in specialized high-risk research programs, such as reducing the length of proposals with anonymization of applicants, to attract less typical proposals. (CCA 2021)

The concept of industrial clusters comes from agglomeration economics:

A coherent whole in which the territorial production system, culture, technology, firms and institutions are closely related. In this environment, trust and reciprocity are two fundamental concepts. The system is then based on a set of implicit rules and cultural norms and on the institutions that support innovation and ensure its flexibility. (Camagni 2005)

This strong proactive associative principle reminds us that if there is a "real" will to revive the industry, with enlightened strategic choices on high-tech themes, the research and development of complex disruptive projects with technological aims must necessarily involve the sharing and pooling of conceptual and practical knowledge between upstream and downstream sectors (Lorenzi 2021). In short, rather than a campaign, should we instead consider using a sledgehammer to destroy the comfortable silos that protect established situations?! There are, and this is the wish of the authors, less radical ways to involve colleagues in this adventure!

Without great scholars, there are no excellent students who will then sow the seeds of new technologies in the laboratories of companies. Without brilliant universities, it is illusory to want to water companies through the research tax credit. It is like watering the sand or, in the case of France, let's be fair, the scrubland [...] (Trannoy 2021)

In fact, especially in France, researchers have been forced to focus on reductive aspects of bibliometrics that are probably from another age. These scientists may think that they are controlled by a new "Big Brother" called the Web of Science and/or Scopus with its high priests of "orthodox" evaluation who may only imperfectly master the processes of qualitative evaluation... But this situation, which has not yet led to a "religious war", is not unique to the Gallic village! Marginson and Xu (2021) remind us that, in the last few decades, the exploration of the so-called fundamental sciences entrusted (because they are not profitable?) to the university or academic world has been transformed quite profoundly. It has become less elitist, more a "mass science", more global and much more networked, and more distributed and diversified (which is reflected in the use of quantitative indicators).

Under these conditions, discipline and a certain monoculture remain the order of the day (with good reason, if only in the use of specialized jargon, in the exploitation of shared paradigms and in the optimized choice of associated scientific equipment). However, it would be necessary to understand the dynamics of the research system leading to innovations that radically underestimate what is being done, what is being thought, what is being tried outside one's own square meadow or that of one's colleagues, members of the same "club"... For Marginson and Xu (2021), there is a debate; they write:

The centre-periphery model is unduly determinist, in the outcome reinforcing the Eurocentrism it opposes. [The paper argues] for a critique of hegemony, not centre-periphery, focusing on cultural factors as well as political economy, and for an "ecology of knowledges" approach as the way forward.

This is undoubtedly to be done, but it requires a real sharing, which may not be spontaneous...

The novelty of subaltern cosmopolitanism lies above all in its profound sense of incompleteness without aiming at completeness... The diversity of the world is inexhaustible and this diversity still lacks an adequate epistemology. In other words, the epistemological diversity of the world has yet to take shape. (Santos 2007)

"My belief is that science is a human activity and that the best way to understand it is to understand the individuals who practice it"... so wrote Freeman Dyson (cited in Lemire (2015)). If this inclusion of science as a social actor is true and enlightening for a Science of knowledge, which is inscribed in the general policy of a State, it is even more credible for a technoscience which has an interest in its primary motivation because it must allow for imagining, socially accepted, applications entering in the economic competitiveness of our country. Indeed, we are no longer in the distant past when a monk, isolated in his monastery in the Austro-Hungarian Empire (the current Czech Republic) conducted, admittedly by cheating a little bit on the results (it is true that since Ptolemy, some people had been used to doing this for reasons of esthetics or power or fear of the latter), studies on peas, probably without thinking of the immense disturbances that these scientific works would bring about. If the emergence of research works with a radical impact on science sheds light on the deep nature of society (at the time, Mendel had forgotten to publish his results in English and in a journal with a high impact factor and a reading committee!), this is not the case for 4D printing, which is well integrated "in the century" because it is a technological extension.

Since the epic breath of Tibbits (2013), the environment has been favorable to a positive destiny, to a "tipping point" of 4D printing in our daily lives, with all indicators in the green. But, between a premonitory vision and the practical facts, if it is obvious that convergent approaches must largely define the daily life of researchers, it seemed interesting to us to examine where the intellectual beacons that illuminate this field are located today. The idea is not to reflect on the "how", but to have a small idea of "where" it is developing? A MAPI CEO Survey (2020) suggests that an eco-systemic approach can help companies accelerate digital transformation and boost productivity and business performance. However, we need to deal with the confusion of a certain number of executives who do not have the right reading keys. They need to be convinced that integrating into ecosystems would enable progress and efficiency factors much faster than for organizations in organizational silos. It is this general attitude that is sought here. The return of these change-averse business leaders has already been approached, in another context, by Montaigne (2020) when he wrote: "Our appetite despises and oversteps what is in its grasp, in order to run after what it does not have: it disdains what is within its reach and pursues what escapes it" or, to put it simply, it is better to hold on than to run, according to the old adage.

Jacquin (2021) reminds us that directive management can hinder collaborative management involving cooperation because each operator (alone or as a group) considers that only the achievement of assigned objectives matters. Working in silos requires a hierarchical system and a pyramidal management with clear objectives. Decisions are top-down and levels of responsibility are upward. The experience of external partners helps improve relations and promote the success of a project: better coherence, better reactivity and increased competence.

Box C.1. A matter of management

Everyone knows that we live in societies that are more or less developed from a technological point of view, but that are increasingly socially fractured internally, with social groups that have antagonistic lifestyles and competing imaginations. On the one hand, there are always those who unwaveringly support technological innovation and, on the other hand, those who see it as a threat, with the environmental problems and other major risks associated with it. So it may be interesting to examine where the quality of an environment conducive to the scientific and technological development of 4D printing lies. The "H-truism" of scientific evaluation (see Hirsch's H-factor (2005)) is misleading when the action has to face events that inaugurate, with a complexity to be discovered.

The choices necessary to engage in a future yet to be invented are strategic, rarely optimal and rather unpredictable and, in the scientific and organizational difficulties of an emerging subject, it may be interesting to avoid additional difficulties by not supporting risk-taking. Indeed,

starting from purely local and short-term linear models, or even from supposedly "sophisticated" models, but always ignoring the "black swans" that these elites push for "optimization", whether in private or public action. Like viruses that kill even their host, their models impose the reduction of costs and the maximization of the profitability of financial assets, while the ontological status of money (trust, consideration, liquidity, holding, etc.) as a source of creativity (the basis of capitalism) and an incentive to innovation is radically ignored. (Le Méhauté and Héliodore 2021)

The innovator [...] does not respect the rules laid down from on high [...], hijacks them or secretly invents new ones. And, finally, if society and the administration are not completely blocked, it is thanks to all those who, consciously or unconsciously, cheat and play on the sidelines. (Crozier and Tillette 2000)

To do this, we have taken up the publications presented in Chapter 1 dealing in particular with the potential applications of 4D printing in order to have a certain categorization of the major geographical areas from which they originate (note that it is not a quantitative result that is expected, but a certain indication of the states or groups of states that can serve as a reference to go a little further on the subject). However, in the perceived "artisanal" or individual approach, this indication, presented in Figure C.3, is probably not binding, as the emergence of other islands or other lighthouses and/or the strengthening of other credible strategic positions have not been stabilized.



Figure C.3. Approximate distribution of states or groups of states involved in research work related to 4D printing (1: China; 2: EU; 3: Asia (minus China); 4: English-speaking world)

As a remark, Joshi et al. (2020) report in their study on 4D materials a slightly different hierarchy as presented in Figure C.4. By analyzing the publications year by year, China seems, by covering practically all the fields discussed in this work, to have an increasingly amplified dynamic relative to other developed countries of the OECD. Its scientific leadership is becoming visible. It will be interesting to examine in a few years the evolution of the data presented in this figure.

Clearly, China, with its current population and dynamics, is at the forefront, while the European Union and the English-speaking world are positioned as "acceptable" players. What does not seem to be expected from current images is the modest role of other Asian countries (South Korea, Japan and Singapore) in this dynamic. But, even if it is not very original, everyone knows that the size of Singapore is not quite the same as that of China! Excellent publications come from this city-state, and the contacts we have with the academic community do not seem to show any loss of momentum. Finally, this knowledge could lead one to think that everyone can have the same assets in the competition on this 4D printing theme. However, it may be interesting to remember ancient history: at the time of the great discoveries (printing, explosives, reaching America, etc.), the very multi-cultural countries of the great Bosphorus region, in relative peace with a number of religions and turbulent neighbors, found themselves very far from these technical advances. The latter came from geographical areas where the citizens were very much controlled by political and/or religious powers that were in clear competition with their neighbors.



**Figure C.4.** Publications by states concerning 4D printing (Sg: Singapore; SK: South Korea; Fr: France; Au: Australia; In: India; Ca: Canada; Jap: Japan)

COMMENT ON FIGURE C.4.— This figure is only an approximate indicator, since only a few references on a leading topic are cited in the bibliography. It would have been easy, for example, to double or triple the number of publications from Singapore on the 4D printing theme, but this is not what we are looking for.

When the truth of a thing is not known, it is good that there is a common error that fixes the minds of Men. (Pascal 1976)

The future is already here. It's just not evenly distributed. (Gibson, cited in Gauthier and Hanifa (2020))

Goulding (2021) reminds us that this large country, China, maintains a strong general dynamic in the field of manufacturing in the broadest sense (see Figure C.5). The representation in Figure C.3 illustrates a buoyant positioning on this prospective 4D printing axis which is real and which, it seems, is tending to grow from year to year. This interesting and credible observation should force other developed countries to position themselves on this scientific adventure, which may have particularly strong economic and strategic consequences. Furthermore, the *New York Times* (2021) considers that the Chinese leaders are in the process of tracing a path to go it alone in the industrial field. They are promising to spend heavily to close the innovation gap and avoid dependence on the United States and other developed countries:

Neither the United States nor China can imminently achieve true self-sufficiency in the myriad of advanced technologies needed to run a modern economy and military. A proxy struggle is being established as both countries strive to secure "missing pieces" from other countries. (VEI 2021)

Continuing on this theme, according to *China Magazine* (Chine Magazine 2021), Chinese President Xi Jinping said in 2013, "Innovation is the soul of a nation's progress and an inexhaustible source of prosperity for a country. It is also an essential part of China's national character."

These and other trends are transforming the global scientific landscape. Businesses like the EU are increasing investment in R&D, with a diminishing willingness to support risky research (Larivière and Sugimoto 2018). China and other emerging economies are becoming increasingly important in the field (CCA 2021). Between 2000 and 2018, the global share of published papers in basic and engineering sciences devoted to China increased from 5% to 21%, and China has long surpassed the United States in the number of PhD degrees (NSB 2020).



**Figure C.5.** Evolution of manufacturing aspects in the economy of a number of developed countries

In a more global framework, Figure C.6 (Couet 2021) highlights countries that have made research a priority, such as South Korea, with other developed countries,

such as France, that are much less involved in this process. However, France, within the "Research Tax Credit" process of stimulating innovation in companies, supports R&D proposals to the tune of  $\epsilon$ 6 billion without it being clear whether this funding is beneficial to French companies. Without wanting to show a classic pessimism in our country, it is possible to remember this remark: "We live and think in the element of this moderately optimistic presenteeism, colored by worries, without strong beliefs, with a limited, impoverished imagination. The most derisory claims have replaced projects, social jealousy and resentment have driven out enthusiasm" (Taguieff 2021). In short, the risk of a follower mentality that has won over the French with rights and few duties should be the subject of deep reflection!

There are about 60 BPI France (*Banque Publique d'Investissement*) support mechanisms that mainly target small, recent companies, particularly in the technology sectors. Their number makes it difficult for some companies to mobilize them. The communication of public operators on their programs is not sufficient to respond to the difficulties of access to information by private actors, particularly in SMEs (BPI 2021). It would seem that this innovation support policy is helping to improve France's position in international rankings, with undeniable progress in the ecosystem of startups that has reinforced its strengths in terms of research quality (with a positive effect of aid on spending or employment in research and development). Despite this, France's position continues to erode relative to its competitors, particularly China.



# Box C.2. France

Figure C.6. Average R&D effort of developed countries since 2005 And then, to finish with an (almost) pirouette, we understood that the technological obsolescence that is desired and/or natural obliges us for the moment to accept the innovations of the "disposable", and this being in temporal domains increasingly short. If 4D printing partly fills this trend, it will be all the more accepted if through its use(s) we can exploit aspects of repair, recycling, etc., the place where Leblanc (2017) has positioned it. This could be realistic as long as the 4D objects have a use still adapted to the technological evolution of the moment (question of the obsolescence of artifacts). And it is indeed due to the competence of the applicants (in the broad sense) that the researchers, in the paradigmatic disruption desired by the authors, will be able to leave their pure science, where the in-depth study reigns, science theoretically disconnected from reality to help with the realization of potentially public utility artifacts.

And as for constraint, since constraint there is, happy constraint which frees us from uselessness, from childishness and from slavery! (Claudel 1997)

Kingdon (2003a, 2003b) introduced the concept of a "window of opportunity" to explain the development of a public policy. This window only opens if the following elements are taken into consideration:

- the existence of a problem to be addressed;
- the possibility of finding solutions for its treatment;
- inclusion in the overall policy.

While the first two aspects are addressed or can be addressed (at least in part in this book, even if developments are still needed), the inclusion assumes the existence of informed decision-makers to support possible futures. But, for example, with the crisis associated with the current pandemic, it is understandable that choices must be made. For all that, it seems necessary that researchers convinced of the interest of a privileged support for the development of 4D printing come out of their ivory tower to try to convince their hierarchies, or even those higher up. On this point, Lévy-Leblond (1994) writes:

A founding myth is needed that allows a society to devote itself to science and to believe in it, to believe in it absolutely, exaggeratedly I would say, because if it only believed in it relatively, reasonably, which would seem normal and healthy to me, it would not believe in it enough to give itself the means!

To go a step further then, the OECD (2021a) refers to "Mission-Oriented Innovation Policies" or MOIPs as a type of systemic intervention to address growing societal challenges. They aim to mitigate some of the most common weaknesses in many national innovation systems, including a lack of holistic strategic direction and policy coordination. MOIPs combine initiatives to "push" the market by acting on the supply side and "pull" it by acting on the demand side. The expected characteristics of MOIPs are defined along three dimensions:

- Strategic direction: legitimacy, directionality, intentionality and flexibility.

- Policy coordination: horizontality, verticality, intensity and novelty.

- Policy implementation: consistency of policy mix, funding capacity, evaluability and reflexivity.

France does not seem to be involved in this process, but has proposed specific support for several years within the Investments for the Future program, PIA (*Projets d'Investissement d'Avenir* in French). In either case, it is easy to imagine that a place allocated to 4D printing could be a particularly effective lever for the deployment of this promising technology. The traditional administrative system is in opposition to a desire to engage in something new, unstabilized, with heuristic evolutions associated with numerous possibilities of bifurcations and destabilizations, even possible failure. This is what, for the authors, contributes to the satisfaction of thinking and undertaking in a very open scientific and technological environment. Today, if we think that we must continue to engage in this path of invention, it is because it can be profitable for our country, to face reality, to learn from others, etc. We willingly take on the risks associated with creation in order to be able to try, but human and material resources are necessary, so PIA or MOIPs, anything can be done if we can, for a while, support the risk of disruption in research.

Because all the great changes do not mobilize only mental processes. It takes desire, emotion, often passion to transform relationships to [...] (Viveret 2012)

However, Crozier and Tillette had written in 2000, before we spoke of 4D printing, these sentences, unfortunately realistic in other situations, adapted to the national reality:

Innovation is not sufficiently visible and dynamic to stimulate the whole field, to create waves of passion that could shake the establishment. Moreover, when we get excited about a discovery, the disappointment is usually strong because the implementation is too slow. Finally, we are victims of the lack of follow-up due to the fact that strategic decisions on resource allocation are made by a bureaucratic and egalitarian system of committees whose choices are constantly questioned [...]

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After testing the EU operation, the change is not very visible, especially since we are trying to work at high TRLs for which the risk-taking is rather modest. Perhaps by changing space, we will find scientific allies who will be able to convince (their) decision-makers of the value of risk-taking for action?

A certain amount of madness (not furious), of creative incoherence, is necessary for the proper breathing of the research system if it is not to lock up researchers who must continue to take initiatives to continue to invent, and to avoid detours and games at the margins. However, by dint of waiting for disruptive ideas to come from other countries and to impose themselves on scientific communities, is there not a risk of an important logical error, a form of cognitive bias, between the asymmetry of action and that of all our non-decisions?

To have passed from cannon fodder to consensus fodder and to the mixture to inform is certainly progress. But flesh spoils quickly: the consensual matter is essentially putrescible and is transformed into a populist unanimity of silent majorities, which is never innocent [...] (Chatelet 1988)

The field of 4D printing is experiencing – at least temporarily – an acceleration in terms of the number of publications, processes, materials development, etc. This is made possible by advances in scientific instrumentation, increasingly sophisticated 3D printing processes, computer science and, to a lesser extent, methods for predictive calculation of active structures and material properties relative to demand. The continuation of this dynamic is essential to enhance the development of the field to facilitate mass customization.

Nevertheless, there are a number of challenges facing modern materials science. These include the need for a big data infrastructure: global management of materials data repositories and data curation strategies; lack of coordination, redundancy, and/or dispersion of instrumentation and technical expertise; need for interdisciplinary research, development, and training; and lack of ecosystems that can help create new supply chains. (OECD 2021b)

This idea of pooling a certain number of shared ideas and the means to investigate them may be of interest as long as we go in directions that are at least partially known. And, if a certain number of barriers have been identified, we are not there yet... On our part, there is no commitment to a programmatic approach that favors a "transversal coordination of activities, the sharing of objectives and stakes for the benefit of a global strategic vision" (Schuster 2021). So, our simple wish is to get out of Simpson's paradox (Delahaye 2013) and thus to participate in the search for solutions (for a problem that can be solved through 4D printing) from a spectrum

of alternative solutions, depending on the scientific and/or technological possibilities, the environment and its estimated evolution, all the while trying to bring about the tensions between these components, the compromises that need to be negotiated, while imagining new uses, etc. In short, real life! With the associated risks...

Simpson's paradox is statistical in origin: a phenomenon observed by several groups may appear to be reversed when the groups are combined. There must be a variable that affects the final (group) result. This confounding factor affects the analysis of the result because the sample under study is not homogeneously distributed. When these two conditions are met, Simpson's paradox can occur.

#### Box C.3. Simpson's paradox (Delahaye 2013)

The precedent is only valid where everything is repeated. Analogy is justified only in a stable universe where the deep causes are involved in easily recognizable external forms [...]. But when everything changes rapidly, the sets disintegrate [...] As for extrapolation, it is content to prolong the current trend which is the result of the deep causes. (Berger 1967)

The appendix to this conclusion gives an idea of the authors' proposal for integrating 4D technologies into our country's economic adventure. But, in general, this adventure is timely, even if some hierarchs consider it premature because they have not yet resolved the old paradigm of the chicken and the egg, which prevents them from taking any risk. The paths to industrial success involve technical, organizational and broader political aspects. The latter are based on safeguards and/or competitive know-how of a technological nature, but technical safeguards can only function within a political framework, such as that of the desire to reintegrate certain manufacturing processes on national territory by making choices on the privileged support of techno-scientific domains that are thought to remain relevant in 10–20 years' time. To achieve this broad objective, several questions can be asked:

- Do 4D printing technologies, if supported upstream today, have the potential to attract the public? A future submission by seduction? Do they induce risks for the users?

- Is it possible to promote greater academic cooperation within the 4D printing field? How can it interact with the business world? How can it achieve sufficient momentum and critical mass in international competition?

- It may be recommended to harmonize the parameters of the tests in order to allow reliable and robust comparisons between laboratories working on 4D printing.

The objective is to try to unify the way scientists design their projects, to produce standards of excellence for the validation of results and to evaluate the best associated evaluation criteria. Would this be enough?

- Basically, can we propose a winning strategy<sup>1</sup>?

- Do they have the power to supplant or compete with traditional manufacturing and usage technologies?

- Can they reinforce the national industrial dynamics? Can they be the cause of an effective redeployment?

- Are they associated with specific risks for users?

- Are they environmentally friendly, recyclable, cost-effective and provide social benefits? Greener, cheaper, safer?

- How can we promote the development of 4D printing technologies, apart from the widely known spectacular aspects?

In this item, promising applications are essential, but most of them are yet to be discovered, or at best to emerge from proof-of-concept. Great progress has been made in the application of materials science and engineering, from the development of materials with significantly improved properties, new manufacturing techniques (including 4D printing) with many application targets such as bioengineering, food, home automation, comfort, transportation and energy production. Computational capabilities have developed considerably and enable understanding, designing, manufacturing and controlling systems in a way that was not conceivable before, capabilities that are yet to be exploited in the 4D domain.

The question of the appropriation of increasingly digital technologies, such as 3D printing, has yet to be answered. For example, in the depressed industrial areas of a French region such as the Ardennes, which used to have a large number of jobs in the metal industry, the expression of union power is not very engaging: "To make 3D parts, it takes a phenomenal amount of time. For me it's a gadget", according to one of the local employee representatives, "It will never replace the jobs that have been destroyed. For there to be jobs, there has to be speed and volume", according to

<sup>1</sup> Several designer perspectives should be considered: mechanics could consider an outcome from the perspective of evaluating the operation of the 4D object as a whole combining practical aspects and the effect of new active materials and their stimulation on the design, operation and maintenance of the 4D system as a whole. On the other hand, materials scientists could create new systems such as meta-materials and new stimulation modes to improve the performance of 4D systems. In this framework, risk tolerance is necessary for their industrial development. Testing, design and manufacturing could be carried out in parallel in the shortest possible time, which probably requires decisive actions with still highly limited information (idea of Plan Bs to be considered?).

another (Notre temps 2021). The same concern will soon arise for 4D printing (as it does for automation and robotics). So it is not only a question of science and technology, but also of aiming for the best possible integration of this small revolution in industrial production.

Given that major advances in one technology often stem from the application of advances in other areas, the question may then arise: Are there subsidiary applications of 4D phenomena for the benefit of society that are now conceivable, but should be explored for exploitation? Can the focus be on identifying new innovative applications that might now become practical through advances in 4D printing technologies? In short, we have probably not yet finished trying to position the 4D adventure in its future applications and its inclusion in national and/or European, or even more widely worldwide, economic schemes! Dear reader, whatever your field of interest, let us take into account, for 4D printing, this reflection of P. Massé, written in 1965: "A fact carrying the future is tiny according to its present dimensions, but immense according to its virtual consequences"... We (you) will have to point out the blind spots on what could become the seed of a strong potential of change and demonstrate it. But, considering the great number of obstacles to be overcome, this may take time and a lot of effort. The scientific truth requires courage because it always implies going beyond false promises through work that is sometimes laborious but essential to make a scientific and/or technical activity credible.

How do we divide the primitively perceived continuity of the material expanse into so many bodies, each of which would have its substance and its individuality? Undoubtedly this continuity changes aspect, from one moment to the other: but why don't we notice purely and simply that the whole has changed, as if we had turned a kaleidoscope? Why do we finally look, in the mobility of the whole, for tracks followed by moving bodies? A moving continuity is given to us, where everything changes and remains at the same time: from where do we dissociate these two terms, permanence and change, to represent permanence by bodies and change by homogeneous movements in space? (Bergson 2012)

If this necessity of the higher culture is not satisfied, the further course of human development can almost certainly be foretold: the interest in what is true ceases as it guarantees less pleasure; illusion, error, and imagination reconquer step by step the ancient territory, because they are united to pleasure; the ruin of science: the relapse into barbarism is the next result (Nietzche 1910)

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# Appendix

PIA4 PEPR – Programme et Equipements Prioritaires de Recherche Exploratoire – French Priority Research Program and Equipment Proposals of the Authors (2021) on the Theme: "Research, Innovation and Industrial Developments Concerning 4D Printing"

We need "a process of knowledge production that is open to epistemic diversity. It is a process that does not necessarily abandon the notion of universal knowledge for humanity, but embraces it via a horizontal strategy of openness to dialogue between different epistemic traditions". (Mbembe 2016)

#### A.1. Elements of context

Born in 1984 from a French patent issued by a CNRS research lab, additive manufacturing has developed spectacular performances over the last three decades: the complexity of objects is now (almost) free in terms of manufacturing, and the material is used for the right purpose with lightweight objects exhibiting similar mechanical performances, thus minimizing the environmental impact. The current world market is worth around 30 billion euros with an increase of 20% per year (but with a modest French presence). This digital technology is one of the pillars of Industry 4.0.

In 2013, in the United States, a disruptive approach emerged, aimed at using active matter in the additive manufacturing of objects. Thus, 4D printing enables, in principle, the production of objects capable of changing shape, property, functionality under the effect of a simulation (magnetic and/or electric fields, temperature, pH, humidity, light, mechanical, biological, etc.). While, in principle, this technology has the same advantages as additive manufacturing, it opens up new fields within and outside of Industry 4.0. However, in the present situation, 4D printing is not able to contribute to the development of the Industry 4.0 principle.

From a factual point of view, a thorough bibliographic analysis of the scientific situation shows:

- a volume of articles from all continents, but with a very "artisanal" French presence, in modest numbers (a few thousand publications, compared, for example, to 160,000 on 3D printing);

- a very high growth rate (44% on 4D printing corresponding to the use of non-living materials; 30% for bio-printing, the 4D of the living);

– a representation of these publications very centered and "bottom-up" in terms of offer, without a direct link with possible applications (performances very far from the needs, simply in situations of proof-of-concept);

– a focus on the use of 3D printers with one or more active materials, without exploring heterogeneous systems located between additive manufacturing and robotics (enabling us to get closer to the performances claimed by the industrial world);

- for the biological component, an avalanche of publications aimed at reconstituting tissues, or even organs, while missing shorter term applications, via the 4D manufacturing of organoids (which mimic a sick organ or not), leading to optimized care (personalized medicine), etc.

An analysis of the demand has also been carried out, it shows about a hundred application subjects where 4D printing could bring a decisive advantage in terms of innovation, high-tech industrial developments, jobs, dynamic image, all within a positive environmental framework (just like 3D printing). This last aspect is all the more reinforced as it is envisaged to produce self-repairing 4D objects, avoiding aspects of programmed obsolescence or not. The Appendix gives some indications on the (numerous) application paths already identified (which do not all concern the same scientific disciplines and their interactions).

French forces in the additive manufacturing field are already widely dispersed, and most of the published research work (apart from a few examples concerning 3D volume manufacturing) concerns incremental innovations. However, while the academic world is spread out "as best it can" on the national territory, the industrial world is up to the challenge with a few leading companies, such as Addup, Prodways, 3D Ceram, Ciny, Poietis (bio-printing), Microlight 3D, Pollen AM and Inetyx. These companies can serve as national technical support points for additive manufacturing. However, this solid but modest sector (3%) is in strong competition with other countries: the United States, Japan, China, and also Germany (with EOS).

What is true for additive manufacturing is even truer for 4D printing, which is less developed. It is possible to let the world of research interested in the field continue to operate on the basis of supply, with the number of publications in scientific journals as the only objective. It is also possible, and this is the purpose of the proposal, to function by crossing informed demands, in the short, medium and long term, with generic know-how from additive manufacturing (and from all the sciences that can participate in this development). It is a question of covering the entire value chain, without being able, given the very large number of applications, to robustly commit to the development of a coherent 4D printing sector. The medium and long term will allow us to refine this need.

#### A.2. The project, oriented application(s)

The part and the whole must be understood both as an aspect of each other and as analytically autonomous – although the degree of relative independence will obviously be more or less complete depending on the historical moment. The theoretical consequences are clear: systems composed of complex parts can expect change to come not only from the evolution of the whole [...] but also from developments within the parts whose movements are endogenously determined. (Smith 1979)

"Customers" (manufacturers, end users) are increasingly demanding "personalized" products (because each customer is unique and wants a purchasing experience that matches their aspirations...), which in the long term requires flexible, agile and reconfigurable production. This observation is reinforced by the need to develop products that meet the needs of the user and their environment. In short, the inherent spatio-temporal facet of the 4D printing technology allows it to adapt to both humans and the constraints of the environment, naturally increasing the functional potential of materials/products, replacing or merging functions performed by components with classic functional materials and otherwise exploiting the energy available from the environment so that changes can be made autonomously (the principle of biomimicry applicable to the Internet of Things). In this disruptive context, 4D printing is becoming essential in the development of Industry 4.0. For all that, what the analysis of the literature shows, it is for the publications of the academic world, the use of more or less mono-disciplinary opportunities satisfying the new and the university criteria of scientific evaluation. While 4D printing must keep its high-level scientific anchorage, exploiting all the potentialities of the "soft" matter (in the sense of De Gennes), it will only be able to function validly by a fruitful crossing between the upstream and downstream sectors, the demand being an essential lever to impose a vision of opportunity, to dynamize, structure and drive the research works for the producing action, not only of knowledge, but also of strong social utility.

In this startup of a recent technology, each country can enter positively into the dynamics of innovations resulting from science. However, by playing with different partners, the associated top-down approach imposes a teleological approach (governed by the goal), which must be translated into interdisciplinary operations. While this situation is risky, it is positive by placing all partners from different backgrounds on the same level and linking scientific silos that are still too disjointed today.

Under these conditions, we propose:

- on the one hand, to rely on research units linked to large organizations (like the main national research centers in France: ONRs) that will engage in taking generic themes associated with 4D printing and bio-printing further (specifically oriented toward personalized medicine). Table A.1 gives indications on the generic actions that today must/can/could serve as pillars for the development of 4D printing (see also Tables 1.1 and 1.3 in Chapter 1);

- to map the knowledge developed (as well as other complementary knowledge) with the demands of the economic world in order to manage interdisciplinary projects for industrial and/or medical developments;

- in association, to maintain a scientific and technological watch of the prospective for all of the actors and consider all the possible openings allowing our country to appear in dynamics (such as the one that it should have had if it had not scrapped the first patent on 3D printing in 1984).

# A.3. Areas of scientific research to stimulate 4D printing technologies and their applications

In the proposed matrix approach, it is essential to progress generic themes for a leadership in the 4D printing field (estimated world market of about 100 billion  $\notin$ /year). 4D printing deals with the evolution of objects, created by adding material, over time. It exploits skills from different disciplines and irrigates others in terms of applications, ranging from science to society. Table A.1 only concerns the

elementary bricks solicited for this development and must however make it possible to cover material uses (from living to non-living), varied spaces (from nano to transport), sensors, actuators (from structural electronics to soft robotics), active materials (with their stimulations), etc. At the same time, design approaches adapted to this field (inverse problem solving) must allow for a better link between the disciplines involved in an application activity (complexity management, multi-scale approach). It is a question of engaging in work on interdisciplinary knowledge coupled with the development of models to reconcile the different physics, properties and spatio-temporal phenomena that we wish to bring into play and to serve as an active support for the development of new approaches to design, modeling and simulation for the digital domain.

Advances in digital technology must be able to make 4D printing processes evolve and vice versa if they present capabilities that increase/stimulate the imagination. The integration of manufacturing processes in engineering is important. Know-how in terms of elaboration of active or functional materials and manufacturing by adding material within the different scales deserve to be articulated, or even combined, in order to develop structures that meet industrial and societal expectations.

Generic themes	Comments
4D design and processes	Creativity; epistemology; transition from homogeneous to heterogeneous 4D printing technologies; process control; heuristic approaches
Digital models, methods and tools	4D knowledge base; ontologies; multi-scale models; bio-inspired methods, etc.
Living, organic and inorganic active materials, and their stimulation	Structure-reactivity relationships; self-organization; biocompatibility Metamaterials and auxetic structures; bistable structures; shape memories; external and/or internal (to the material) stimulations; spatio-temporal associations of stimulations
Digital modeling	Anticipation of effects, robustness; inverse problem; collective transport; self-organization; differentiation and cell growth Innovation
Mechanics	Solid mechanics; soft matter; µ-fluidics; collective transport (swimming robots)
Fatigue	Anticipation of effects (mechanical, chemical aging, etc.); self-repair
Chemistry and photochemistry	Photochromic systems; functionalizations
Soft robotics	Robotics; cobots; biobots
Adaptive optics and photonics	Use of light as a stimulation mode; use of 4D printing to optimize optical processes
Micro-nano 4D	Nano-optics, nano-sensors and nano-actuators; adaptive optics
Soft electronics	Structural electronics

Medical applications	Biocompatibility; drug delivery; smart orthotics for high-performance sports
Biology	Interactions of cells with each other and with the environment and their studies
Bio-printing/ precision medicine	4D of the living; organoids; μ-fluidics

 
 Table A.1. Targeted scientific disciplines or areas to support the development of 4D printing

Change in science can occur in any scale, though at any time one or another scale can be decisive. World-systems theory's debacle in prediction, the manner in which it became decoupled from the actual history of science (and of the world order) shows that deductive reasoning from generalisations at the level of the stratosphere is a precarious method. Context and contingency matter. Theories are tools to be mixed and matched at need, not iron-clad formulas. (Marginson and Xu 2021)

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If the capture and reuse of 4D printing knowledge is necessary for this objective, the conclusion leaves the existing myth around the 4D printing theme and proposes a "draft" roadmap that should be the subject of reflection and scientific debate on a concept that is still immature, but full of promise.

**Frédéric Demoly** is a Full Professor at UTBM, France, and a department director at the CNRS (French National Center for Scientific Research). His research focuses on design for 4D printing using computational intelligence, and on multi-material additive manufacturing processes.

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