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Programmable Matter or Smart Matter, Stimulated Organization and 4D Printing

"It is not reason which speaks within technical objects, it is desire, and it is concrete existence which will bear the brunt of these dreams". [CÉR 11]

"The most successful entrepreneurs don't begin with brilliant ideas; they discover them. The great advantage of working in this way is that when trying to do something new and uncertain, we rarely know what we don't know". [SIM 11]

"The answer reassures us. The question merely serves to worry us. In the literal sense of the term, it compels us to react, indeed to invent. Raising the question, out of necessity, forces us to compel us to change our emphasis, to reconsider the hypotheses, to review the rules and to unearth the obscure". [LAG 12]

"Something that it has distractedly enabled [Man] into the present which would open up the future to him/her". [ZAM 89]

"Speed both demands and creates insensitivity to anything that can delay, to all forms of friction and hesitation which make us feel that we are not alone in the world; by slowing down, we again become capable of understanding, of getting acquainted with, of acknowledging what we have and what is transmitted to us, to think and imagine and, in the same process, to create with others relationships are not themselves captured". [STE 13]

"The concept of engineering is not realizing the given use until we discover its existence". [FRE 94]

"There are therefore no 'rules of induction' of general application, thanks to which we may derive the solution mechanically or infer hypotheses or theories from empirical data. To go from the stage of data to theory, it is necessary to undertake a creative work of imagination". [HAM 14]

"Initially, we learn that thinking too quickly may be a very bad idea. Infobesity surrounds us and it is a serious threat to our creativity". [LEW 16]

"The undertaking will be difficult, as it goes against the dogma of the Science's so-called 'Immaculate Conception'. According to this, scientific truths being designed without sin, are all beneficial. So that the sanctity of Research will not be diminished by the uses that the World puts it to and that it is not for those who are ignorant to discuss the use of funds – indeed always insufficient – which are allocated to them". [CÉR 17]

"'Festina Lente' (Hurry up slowly) was the motto of Augustus". [LEW 16]

1.1. Introduction

In the introduction to this book, DNA origami was mentioned as an example of a self-organizing system. Indeed, this was already chemically stimulated organization as it is necessary for us to have attached links in the DNA fixing onto the macromolecule to define a given form anticipated by the designer. The notion of stimulated self-organization used by the scientific community is perhaps not adapted to reality, or nature doing what it can. It is this issue which we will investigate in this chapter and indeed those that follow.

This is the case for molecular motors whose movement is enabled by external energy sources. Richard Feynman had predicted in the 1950s the development of molecular nanomachines such as those procured by Jean-Pierre Sauvage in 1983 when he invented a chemical method that enables the intertwining of two molecules taking the form of rings, thus forming a chain known as catenane [CNR 16, COL 01]. Thanks to an electric or light stimulation, it was widely shown that it is possible to cause molecular rotation in a controlled manner. This is similar to the given operation of a motor. We may therefore consider these examples as already being representative of what might be programmable matter. This has been of no interest to researchers for several years without an ability to detect a trend (whether linear or exponential) in the growing number of publications associated with the key words "programmable matter" (see Figure 1.1). The total number supplied by university libraries demonstrates a figure of the order of 1000 publications which should be compared to the 50,000–100,000 concerning additive manufacturing.

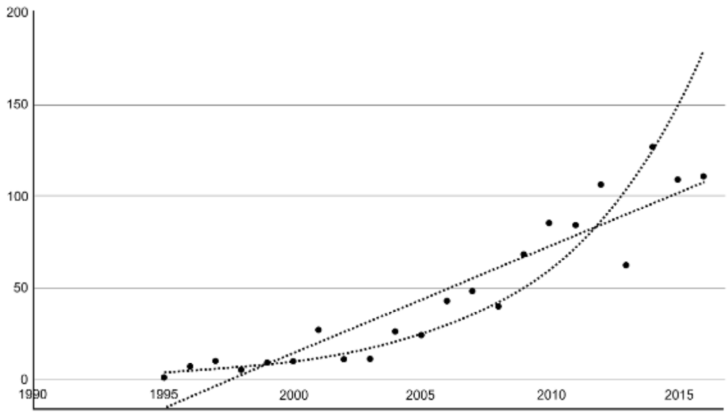

Figure 1.1. *Development of the number of publications on the subject "programmable matter" (University of Lorraine, France)*

As the focus of the subject is 3D printing, it appeared necessary to leave this sphere to approach the concept of additive manufacturing, which effectively involves the notion of additivity. We need at least two molecules to begin discussing additive manufacturing.

The aim of this chapter is to examine how we can exploit the overall principle of gathering separate components. We can thus make up a "self-organized" shape (to keep the term we used previously). Thus having reviewed this principle, it will be possible to envisage examples of self-organization which will partly serve as examples of 4D printing.

1.2. Natural (spontaneous) self-organization

Stating that matter can, in certain situations, be ordered means that the mold can, as a first approximation, perhaps be described geometrically in a simple way, by using for ease the concept of symmetry (for example, the case of a crystal). This is the case even if there are sub-structures which are more or less complicated, which can even take on a fractal aspect. However, the general form remains simple. We may think that structures, which are more or less self-organized, may be combined with some types of physical or physicochemical conditions [CHO 15, CHO 96].

The term "self-organization" designates a spontaneous or stimulated formation of a dynamic order leading to forms of organization stemming from a set of homogeneous or non-homogeneous units, which are interacting. Having placed the energy in the correct place and at the right time, in a given environment, the initial system, whether or not it is random, is organized to achieve a given form, if possible that sought by the given experimenter [MOR 04]. From a thermodynamic point of view, this "shaping" assumes that the transport of matter and energy within the system which is created should be subject to functional constraints in the absence of an external energy supply. The object is constructed, having the appearance of causal relationship (the laws of universal physics), which may be shown by a type of selection between several potential alternatives [CAM 74]: "Thus, its role is the construction of higher-level entities, formed from the (re)structuring of units which are intrinsically made up of material causality" [MOR 06].

1.2.1. *Nonlinearities*

The dissipative structures are processes in which fluctuation is stabilized to make up a dynamic macroscopic configuration. This is enabled by the flow of energy through the system under construction. These macroscopic structures necessitate, so as to increase in size, a continuous supply of energy in a range adapted to its cohesion. "The term 'self-organization' instead implies spontaneous creation of a given order or organization that refers to an agent-based identity, which is able to self-determine or form itself in a direction which involving a functional plasticity" [ALV 04]. Ultimately, we are concerned with respecting Bruter's concept [BRU 76]. He wrote, "Events shape the object with time. It is, above all, piercing the secrets of its history, of the line which it comes from, both the culmination and the projection".

A classic example is that of living systems which manifest behaviors and self-supporting capabilities. These include growth, differentiation, self-repair (at least partially) and other features. Such systems are capable of altering their behavior and their structure according to exterior parameters (nutrients, for example, temperature, gravity and other factors).

Quoted by Thuillier [THU 80], Stéphane Leduc wrote: "The elementary act of life is diffusion and osmosis". It was this professor of medical physics from Nantes who manufactured shapes and structures "resembling those of living beings" (in the early 20th Century). In 1910, he published a work entitled: "Physicochemical theory of life and spontaneous generations". Of course, it naturally conflicted with the stance of the French Academy of Sciences. Figure 1.2 represents one of the osmotic productions of Leduc [THU 80]. He wrote in 1912: "When we come to know the physical mechanism for the production of a given object or phenomenon... it becomes possible... to reproduce the object or the phenomenon. At that point, science becomes synthetic. Biology is a science like all others… it must sequentially be descriptive, analytical and synthetic" [LED 12].

Figure 1.2. *Osmotic productions by Leduc (copy from [LED 10]; see also [THU 80, EAS 09])*

This causalist theory and the methods flowing from it are to be applied to these constructs that aim to obtain finished and optimized products after "they have been initiated", in particular as regards matter and energy consumption, the notion of the voxel which may then reach the size of a molecule, indeed a cation or an atom then becomes relevant. The idea is that, as for all of the (or the majority of) engineering sciences, these methods are still often thought to be determinist: the various phenomena are controlled by laws that we must discover and apply as faithfully as possible. There might therefore not be (too much) space for the random.

Although this was the case, based upon an energy spatially positioned within a known manner, with cooperative components able to self-assemble, it would suffice to wait a "sufficient length of time" to reach an equilibrium leading to an object taking a given shape. Moreover, resuming the same experiment, all things being equal, should provide the same object (taking the same shape and size). Thus, by using this causalist hypothesis, it is not possible to predict the evolution of shapes by inferring them uniquely from the application of simple laws which may be able to be identified. This result illustrates an evolution that develops in a non-determinist way by the interaction of natural systems both with each other and their environment. However, by way of example, Prigogine [PRI 94] recalls this experiment. If we cool sodium chlorate at rest, the crystals produced are optically inactive. If, during the same time period, we shake the solution, we obtain either levorotatory crystals or dextrorotatory crystals. This is a bifurcation. The first provides either a skewed shape or a straight form and the initial germs which will participate in the crystallization process are either skewed or straight. There is therefore a break in the symmetry of the reactive system. This example serves to show that irreversible phenomena do not necessarily reduce down to an increase in chaos. However, there can exist constructive roles for this irreversibility of time as is the case with life. Hawking [HAW 89] recalls on this subject: “A strong thermodynamic arrow is needed for intelligent life forms to act”. These aspects will be reconsidered in Part 2 on bio-printing.

In these conditions, Turing [TUR 52] showed, using other examples, how reactions between chemical substances that he called morphogens, coupled with a process of diffusion of these substances through living tissues, could give rise to the appearance of specific structures or regularities (spots on a coat, the shape of a crystal, a shell or other examples). In his founding article, he introduced a mathematical model known as “reaction-diffusion equations” as well as the study of this model in certain relatively simple cases.

The dynamics of this basic model which he explains results from the coupling between chemical reactions and reagent diffusion, within a system which is continuously fed. The Turing structures correspond to periodic spatial variations of concentrations of chemical species [LES 13, ZEN 13, SHA 15, JOS 12] which compete with the mechanisms for molecular transport. To observe spatial variations in concentrations, it is therefore necessary for other mechanisms to come into play. It introduces a scenario involving two species A and B, known as “morphogens” and the combination of the following properties:

– species A is capable of inducing its own production;

– species A can produce the second species B;

– species B inhibits the production of A;

– B diffuses quicker than A.

It is thus sufficient that a small local fluctuation at a given point in space induces a slight excess of A so that the production of both A and B is higher than the average at this point. B, diffusing quicker than the excess of A, creates an inhibitory crown around the initial point, isolating the peak of A by an area which is richer in B. These random evolutions can happen in other places in the given area with other peaks of species A. “These activator peaks arise from selective ranges of fluctuations, which in very small quantities yet numerous, which spontaneously modifies the state of the homogeneous mixture. The distribution which is dynamically the most stable of these peaks forms a periodic pattern, such as spots that we may note on the cheetah” [CHO 15].

James Clerk Maxwell wrote in 1876 (cited by Thom, [THO 83]: “When the state of affairs is such that an infinitely small variation in the present state will only alter the future state by an infinitely small quantity, the state of the given system, whether at rest or moving is known as stable. However, when an infinitely small variation in the present state may cause a finite difference in a finite time, the condition of the system is known as unstable. It is obvious that the existence of unstable conditions makes the prediction of future events impossible, if our knowledge of the present state is only approximate and inaccurate”. The question of the control of the “correct” main parameters of influence on stability or instability is therefore paramount, but risks not being sufficient.

1.2.2. *Achieving the desired form?*

Once science starts to understand the way that nature has selected certain forms and not others, as much in the physical as the biological world, it can seek to apply/transpose corresponding mechanisms with a view to resolving engineering problems. This is advantageous for the manufacturing of given artifacts, tools or final articles. The large variety of given applications in a generic form such as the manufacture of a functional object is not inspired from nature. It is worthwhile investigating this if we seek to use knowledge of “morphogenesis” to achieve the desired form through self-organization [BAQ 04].

This is the inverse problem applicable to the direct manufacturing of objects (and not by additive manufacturing, on a voxel-after-voxel basis) and to create (in a way) three-dimensional structures. These are of varying sizes from a pseudo-homogeneous environment. This is a traditional problem “normally” not respecting entropy principles. It forces us to know from what energy and in what form it should be available. This is because the process is anti-dissipative. Indeed, there is nothing in the demonstration of the second principle, which enables us to assert that there exists a causal relationship between entropy variation and evolution towards a chaotic state. However, as soon as several entities interact, or the effects are not proportional to causes, there may be the possibility to make nonlinear characteristics available, with specific effects (for example, snowflakes within systems outside the equilibrium environment with the provision of continuous matter or "photochemical muscles", which introduce tensions of the manner of mammalian muscles). A multi-scale approach could be an asset to dissipate a given energy surplus. In these recursiveness conditions, the structure is as it were both a cause and consequence of this dissipation (self-similarity, see Figure 1.3). Self-organization thus gives the impression that the structure emerges from the combination of interactions between constitutive entities (including energy).

Thom [THO 83] analyzed problems connected with the control of morphogenesis by relying upon the notion of the model "black box" with its inputs and outputs (some of which revert to the recursiveness of inputs). When choosing Ui inputs, it is possible to measure Sj outputs. This is provided that their existence has been identified and, thereafter, to define a relationship between the Ui and Sj respective inputs and outputs, as Turing proposed in [TUR 52]. The central question, well known in process engineering, is from the experiments completed to reconstruct the inner workings, which happens within the black box. Within complex systems, there may exist "exceptional" situations for which the relationships {Ui, Sj} come out what we expect as "normal". The research into these "divergent", but real, situations may lead to new and necessary information becoming more apparent as to what happens in the "box". At such times, if we do not know where to look (and what to find) in these situations from an experimental point of view, it indeed falls to modeling to help search for peculiarities. This takes place at different scales.

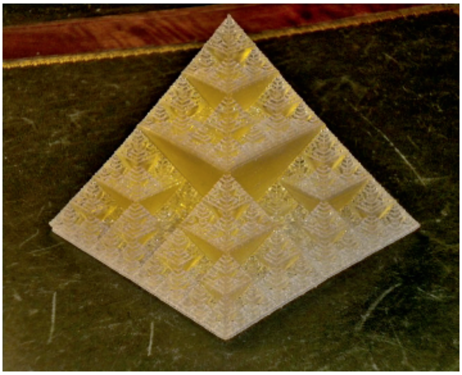

Figure 1.3. *Self-similarity – fractal pyramid produced in the 1990s [DIO 92/93]. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

Laurent Larger [LAR 15] states that nonlinear dynamic systems apply to all practical situations or movements appearing in space and/or time and for which there is a determinist description of development, taking the form of a differential equation (solely the temporal dynamic) or a partial derivative equation (spatio-temporal dynamics). Our world is of course 100% dynamic and nonlinear, but the current technological control relating to its operation is very often limited to situations with linear approximations. The example of morphogenesis illustrates not only the relevance of the nonlinear approach (a simple, deterministic, local dynamic model explains how nature constructs globally stable, reproducible, and robust forms, at the macroscopic level), but also the gap of scientific progress existing in the understanding, and then in the mastery and exploitation, of the associated mechanisms for concrete applications, with enormous technological potentials.

Finally, we must underline here the significance of the local and global notions within nonlinear dynamic phenomena. It is indeed necessary to understand the fact that a highly complex solution, at the global level, can effectively emerge in a determinist way from a uniquely local property (the dynamic determinist equation and local nonlinear coupling, which apply to all of the environments studied). It is relatively simple within its formulation.

From a point of view of modeling within the sphere, we may consider a system that evolves in time stemming from a given form to achieve another form (a dynamic approach) or impose upon stable form constraints of different forms (chemical, energy or other spheres) which will lead to a distortion (a thermodynamic approach). It may be useful to introduce, in the first instance, temporal elements which enable the definition of development at a given time. In the second instance, thermodynamic and chemical potential components are introduced. That being so, starting from a given Ui set, it is possible to define the Sj outputs in theory, provided that all determining factors are known. The converse problem aims to define the Sj outputs defining instructions and researching the Ui which meet the objective. This is a traditional problem that is always difficult in the case of linear systems, which are generally poorly conditioned (see deconvolution not least owing to noise and measurement errors; see [MUG 08]). This will be all the more the case since we will be positioned within nonlinear approaches with loud signals. That being said, the issue is the size as the control of the inverse problem can constitute an integral means to produce an object from a matrix which is easy to produce. The latter aspect and the distortion function in terms of time and energy supplied may enable the production of an object taking a complex form.

Chauvet [CHA 06] considered that when two units A and B are subject to certain conditions, they can do nothing other than join together (see the works of Turing mentioned in the introduction). For example, the dynamics of two biochemical pathways carrying a reagent from one to the other would be, per this principle, more stable than the dynamics of each entity when separated. This is the principle of stabilizing self-association per Chauvet, [CHA 06], Staetzler, Sonnenschein and Soto, [STA 11] and, Nykter *et al.* [NYK 08]. It can operate the same for enzymes (see [KEK 15]) with divisions between species which necessitate limited transport by molecular diffusion (solution of the Von Smoluchowski equation within a potential field; Kekenes-Huskey *et al.* [KEK 12]). Self-organized systems can be very stable relative to the range of a number of parameters, but they can diverge suddenly according to small variations of one or several influencing variables to reach a new stable state [NYK 08]. “Turing” evolutions with fractal approaches can also be put forward (see [BIZ 11]).

Thus, between the possible illusion of perceptible self-organization and the inverse problem, constituting a significant challenge, allowing the production of the researcher’s or engineer’s desired organized structure, there is a considerable gap to get over. Would it perhaps be necessary to consider nonlinear, out of balance and multi-scale phenomena? Perhaps it would be necessary to view the biological aspect (as was introduced in Volume 1 with DNA and DNA origamis or “smart” matter. Other biomimetic solutions are also envisaged using nanoparticles: Sarikaya *et al.* [SAR 03] Nikitin *et al.* [NIK 10]. Slocik and Naik, [SLO 10], Tamerler and Sarikaya, [TAM 07], Chiu *et al.* [CHI 11], Maye *et al.* [MAY 09], Laxminarayana, [LAX 12]). These involved original experimental pathways from the traditional physicochemical sphere to produce customized three-dimensional structures (although going beyond the nanometric sizes of origamis). Perhaps it would also be necessary to move away from traditional reductionist methods to involve holistic thinking in order to explore this small element of complexity. Nowadays, this point partially moves away from the topic explored, but this work is an enormous task which, at least from this author’s point of view, must be looked into more deeply. This will be shown with the example of bio-printing (see Part 2).

Put simply, what the engineer seeks is to place matter "in the right place" and "at the right time" so that the organization leads to the object sought (and not something to which to get closer or nearer). As when dealing with an insufficient control of systems, he/she is obliged to attempt to find situations where he/she resumes control of the overall complexity, through using "smart" matter. The results of these efforts are shown below.

1.3. "Smart" matter

The underlying concept is highly different. It starts from a given geometry so that it becomes, through the supply of energy, a mold corresponding to a given application (see Figure 1.4). By playing with spatial division with a range of form of this energy (from these energies), there may be also the possibility of having an adaptive system, such as an actuator, a structure evolving in time and perhaps in its functionalities [ZAR 17, LEH 02]. In passing, ideas of this sort were proposed in comics ([TUR 15]; Franquin published in Beaux-Arts, [BEU 15]). Only science-fiction films approach this type of subject thoroughly.

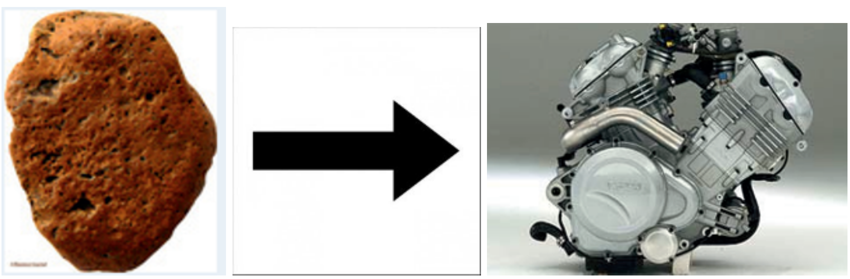

Figure 1.4. *The principle of using "smart" matter*

This context does not mean spontaneous complexity discussed above has been abandoned, but rather that the engineer and/or the designer attempts to take control of automated phenomena which are difficult to control, by accepting the manipulation of non-living matter and energy, so that initial shapes are tailored by their will. This section thus explores stimulated conversion of matter pathways. Lehn [LEH 02], said, backed up by Figure 1.5 in this paper, the "smart" matter referred to her falls within the exploration of adaptive supramolecular systems.

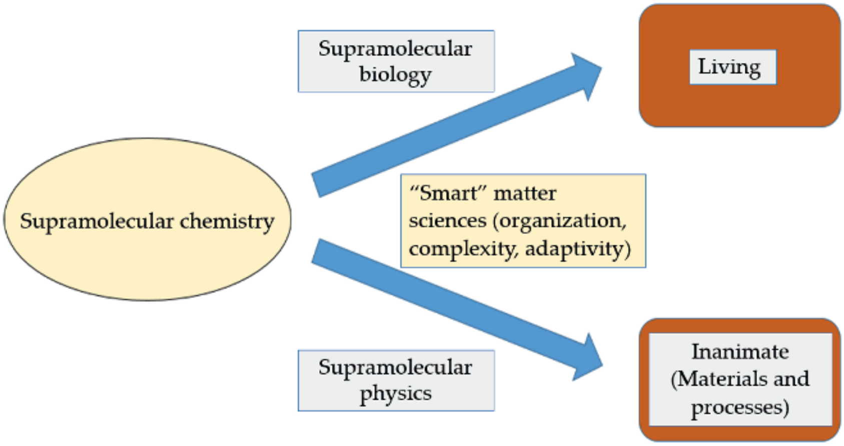

Figure 1.5. *Smart matter and supramolecular systems. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

1.3.1. *Active polymers: photochemical muscles*

One particular approach concerns the effect of light on polymer structure (the now famous expression "photochemical muscles"; Nakano, [NAK 10], Yoshino *et al*. [YOS 10]; Yu *et al*. [YU 03], Yu *et al*. [YU 15], Khoo *et al*. [KHO 15], André, [AND 17]) as Figure 1.6 indicates. This is extracted from Kuksenok and Balzs [KUK 13]. The concept involves irradiation inducing internal rotations in the material, which contributes to change the geometry of the irradiated area leading to distortions, which depend upon the flow received. The latter may be reversible if we have photochromic material available (which would both induce provisional conversion and be reversible).

1.3.1.1. *Photochemical changes*

All of these processes, apart from photodegradation, are shown in Figure 1.7 stemming from Khim *et al*. [KHI 14]. The curvature of polymeric fibers can be connected to a non-homogeneous division of photoproducts (see [GE 16a, AND 17]).

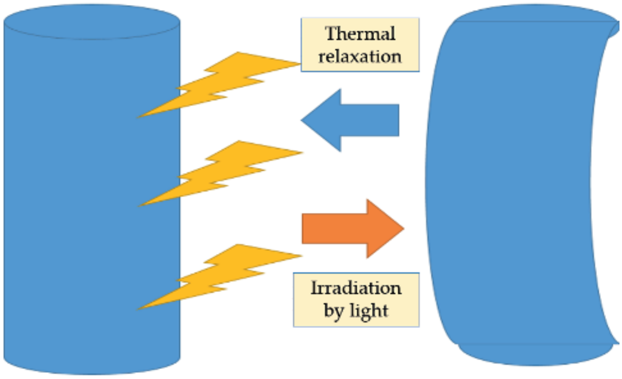

Figure 1.6. *Distortion induced by the light of a photo retractable polymer cylinder. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

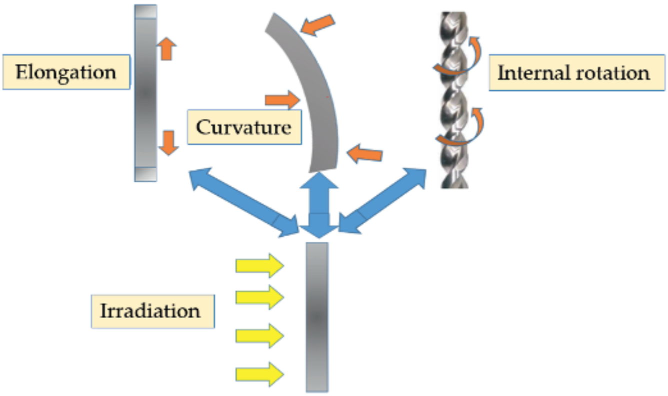

Figure 1.7. *Photomechanics of polymers. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

Another example, stemming from Mahimwalla's thesis [MAH 13], is the significant distortion of polymers containing azobenzene groups (see Figure 1.8 and [MAH 12]). These groups through irradiation go from configurations Trans to Cis depending upon the given wavelength used (see Figure 1.9). The effects are noteworthy as can be seen in Figure 1.12 (see also [DES 15, ROS 15]).

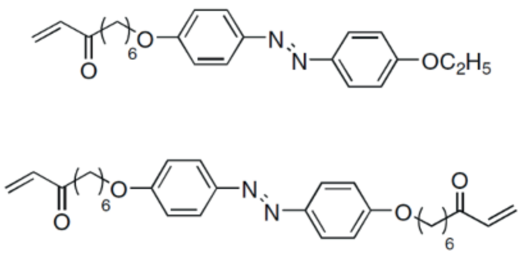

Figure 1.8. *Azobenzene polymers*

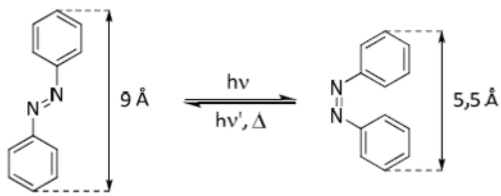

Figure 1.9. *Sizes of two isomers [DEL 07]. The conversion from Trans to Cis operates by ultraviolet means (300–400 nm), returning to waves which are visible (being between 400 and 500 nm)*

The azobenzene molecule is generally found in the Trans conformation, which may be considered as uniaxial. When this molecule is exposed to visible light, it can undergo photoisomerization to a Cis conformation (see Figures 1.8 and 1.9). The given molecule may then return to the Trans conformation consecutively with a thermically or optically induced conversion. Once this cycle is completed, the molecule may be located moving in a new direction. This process may be used to act upon the macroscopic direction of the molecules of a given film. When polarized light is linearly used to activate the cycle of photoreorientation Trans -> *C*is, only molecules for which the electronic absorption transition moment is located within the given direction (or within the vicinity) of the polarization absorb light and undergo a change of direction. In this way, it is possible to reduce the number of molecules oriented parallel to the direction of polarization and to increase the number of molecules oriented at right angles to this direction [ROC 15].

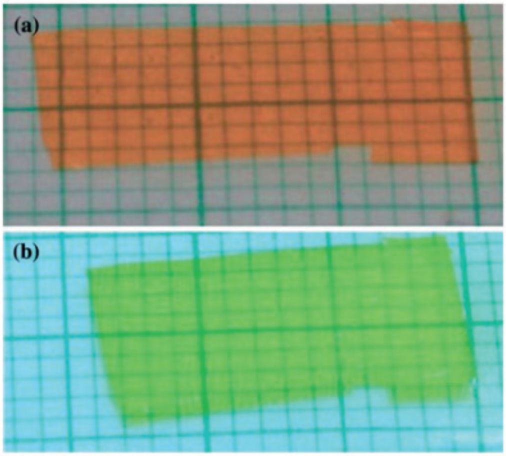

Figure 1.10. *Effect of irradiation on a polymer film containing azobenzene groups: a) before irradiation; b) immediately after irradiation according to Mahimwalla et al. [MAH 13]. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

These authors present the fairly spectacular results of Yamada *et al.* [YAM 09], corresponding to the distortion of an induced polymer film, both by UV irradiation (conversion Trans -> Cis) at 366 nm (240 mW.cm^{-2}) and in the visible domain (λ > 540 nm and 120 mW.cm) for the inverse conversion. Figure 1.11 kindly supplied by Kevin Ge [GE 16b] shows a reversible conversion of a polymer which is distorted with temperature. Figure 1.12 by Cheng *et al.* [CHE 10] completes the previous figure: having a double layer of polymers, one of which contains azotolane which can under a given isomerization Cis - > Trans thermic and Trans -> Photochemical Cis under visible light irradiation, it is possible to induce a temporary withdrawal of the irradiated film, followed after the irradiation ceases by a return to the initial position (see also Figure 1.13 by Zhang *et al.* [ZHA 15]). Other works are able to respond to this conversion principle [ERC 10, ILI 11, PET 15, GAO 16]. We may note that in view of the molecular structure of azobenzene and azotolane, it may be possible to take account of the direction of the transition moments of these molecules placed within a polymeric system to make conversions within polarized light determined with the given space, to create programmable diffraction networks (see above). Robotic applications are envisaged by Umedachi, Vikas and Trimmer [UME 13].

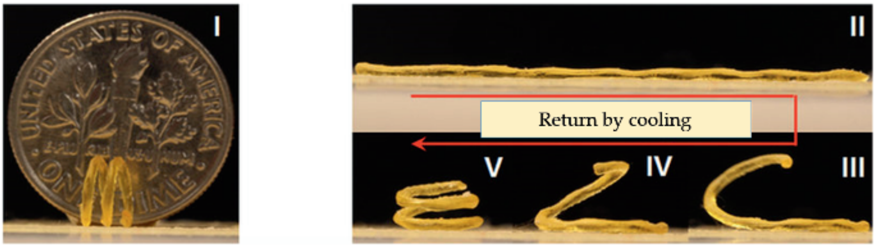

Figure 1.11. *Distortions of a light-induced polymer film. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

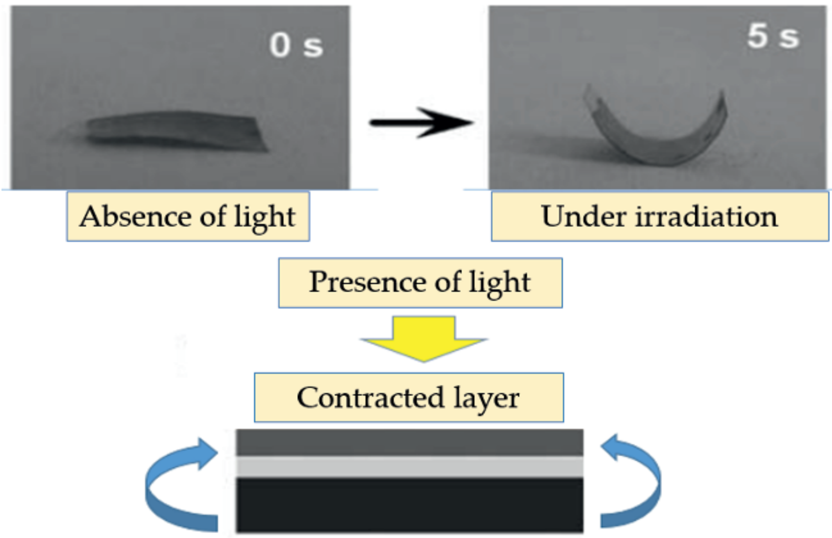

Figure 1.12. *Bimetallic element subject to light irradiation. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

Such systems can be produced by 3D printing with the assistance of various passive materials, but highly elastic, for example, with aliphatic acrylic epoxy-based monomer mixes (monofunctional) and urethane-based (bifunctional) substances. Details of these were recently published by Patel *et al*. in 2017 [PAT 17].

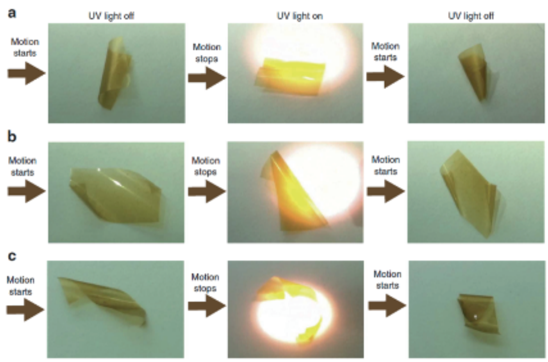

Figure 1.13. *Distortions of a light-induced polymer film according to Zhang et al. [ZHA 15]. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

1.3.1.2. *Thermic effects on polymers*

Zhao, Qi and Xie [ZHA 15] said that it is possible to have at one's disposal polymeric materials, the shape of which may either depend on the temperature or on photochemical conversions. These authors in their experiments were seeking to find reversible reactions affecting the space between molecules or macromolecules (see Figure 1.14).

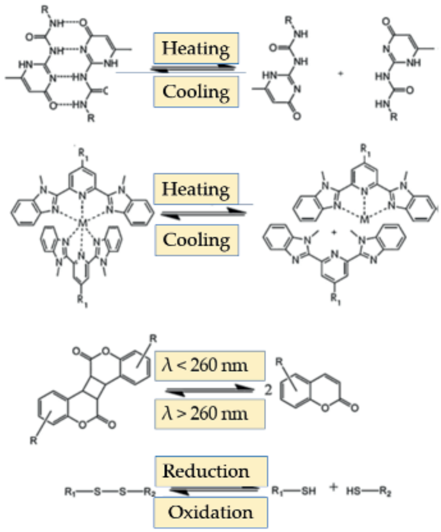

Figure 1.14. *Reversible systems having volume changes*

An explanatory example is given in Figure 1.15 illustrating the change of shape. Other examples are given in this article using various active materials.

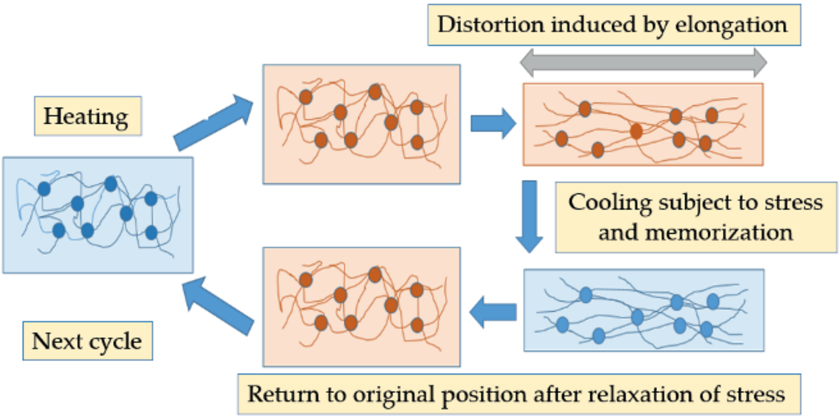

Figure 1.15. *Principle of thermically induced reversible distortions. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

Inks produced from hydrogels are capable of inducing distortion by humidity and by temperature [NAF 17]. Le Duigou *et al.* [LED 16] used wood particles as a charge within a given polymer. Oriented within shearing extrusion of this fusible polymer, they retain the memory for their given direction below the material's freezing point and can be seen to change their shape in the presence of humidity (or heating). Finally, from optimization view point, Zhang *et al.* [ZHA 11] added to a material sensitive to heat and light (poly-(N-isopropyl-acrylamide or pNIPAM), charges in carbon nanotubes. This facilitated the transfer of heat within the mass of material and the absorption of light by the given surface.

1.3.1.3. *Other systems*

In the same vein, a "paper" developed by researchers from the Donghua University in China may be qualified as "robot origami" with a composite material capable of reacting to temperature by distorting. A double-layer flat material may be, according to Rochefort [ROC 15], half made up of an oxide of graphite, the other layer of polymerized dopamine or polydopamine, a material that reacts upon contact with water. When exposed to light or heat, this layer shrinks. The other layer, having good mechanical strength, reacts to this action by bending or folding up. By using a high-precision infrared laser, researchers were able to develop tiny pieces of paper, capable of moving on demand. This research may serve to produce artificial muscles. Other examples are shown in Figure 1.16 of polymers sensitive to localized stimuli within space and which can be sensitively distorted. These were researched by Kevin Ge *et al*. [GE 16b] – see also Zark *et al*. [ZAR 15].

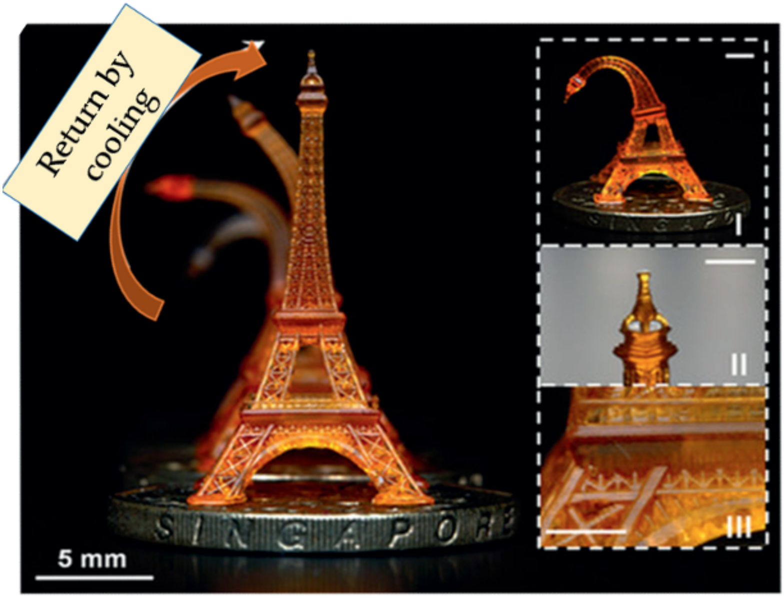

Figure 1.16. *Another example of polymers sensitive to given stimuli and validation by additive manufacturing. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

For their part, Grossweiler *et al*. [GRO 15] used polymers containing spiropyranes which are sensitive to mechanical stress. These may become distorted and change color with a relaxation time of several minutes (see Figure 1.17). This type of material enables them to produce micro pliers.

There is another example that taken from the works of Chen *et al*. [CHE 17]. Active polymers, ready to respond to a blue-light stimulation (from an LED), containing trithiocarbonates (TTC) groupings with an accordion-like structure may be activated by organic catalysts. In the presence of light, monomers are formed (the inverse process of polymerization), which indeed has the effect of drawing out the macromolecule. The monomers are distributed uniformly throughout the structure. They confer new properties on the material, which then enables us to modify the shape of the object. This complex technique necessitates the specific conditions for use.

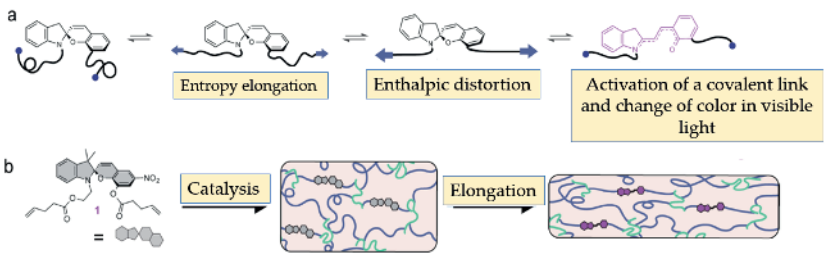

Figure 1.17. *Effect of a mechanical constraint on the chemical conversion of a spiropyrane-based polymer. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

Table 1.1, partly extracted from Gao *et al.* [GAO 16], gives indications as to convertible matter and upon the methods of conversion applied within this domain.

Type of materials	Mechanism and commentary	Bibliographical references
Polyethylene glycol; lipid bilayers	Contact with water	Jamal *et al.* [JAM 13], Villar, Graham and Bayley [VIL 13], Villar, Heron and Bayley [VIL 11]
Polymers at low-transition temperature (PNIPAAm; PCL); alumina ceramics	Transition temperature	Peppas *et al.* [PEP 00], Wang *et al.* [WAN 15], Bakarich *et al.* [BAK 15]
Magnetic materials	Precise positioning by the given location of the magnetic field	Kokkinis, Schaffner and Studart [KOK 15]
Polymers containing light-induced isomerizable components	Photochromic compounds	Zhang *et al.* [ZHA 15]

Table 1.1. *Various types of smart matter and associated mechanisms*

1.3.1.3.1. Biological materials

Natural plants demonstrate spontaneous curvature alterations due to the direction of the cellulose, which is subject to an anisotropic swelling when in the presence of water. Gladman *et al.* [GLA 16] imitated this process using a hydrogel containing such rigid cellulose fibrils. They spontaneously become aligned with shear points at the time of their flow through the nozzle of the extruder of the printer, which induces both swelling anisotropies and elastic module anisotropies. With a dual layer positioning, it is possible to achieve swelling differentials, hence a calculable curvature for the given object. By using anisotropy within the material, Qe *et al.* [GE 16b] show in Figure 1.18 how it is possible to develop the form of an object according to the stimulation it receives.

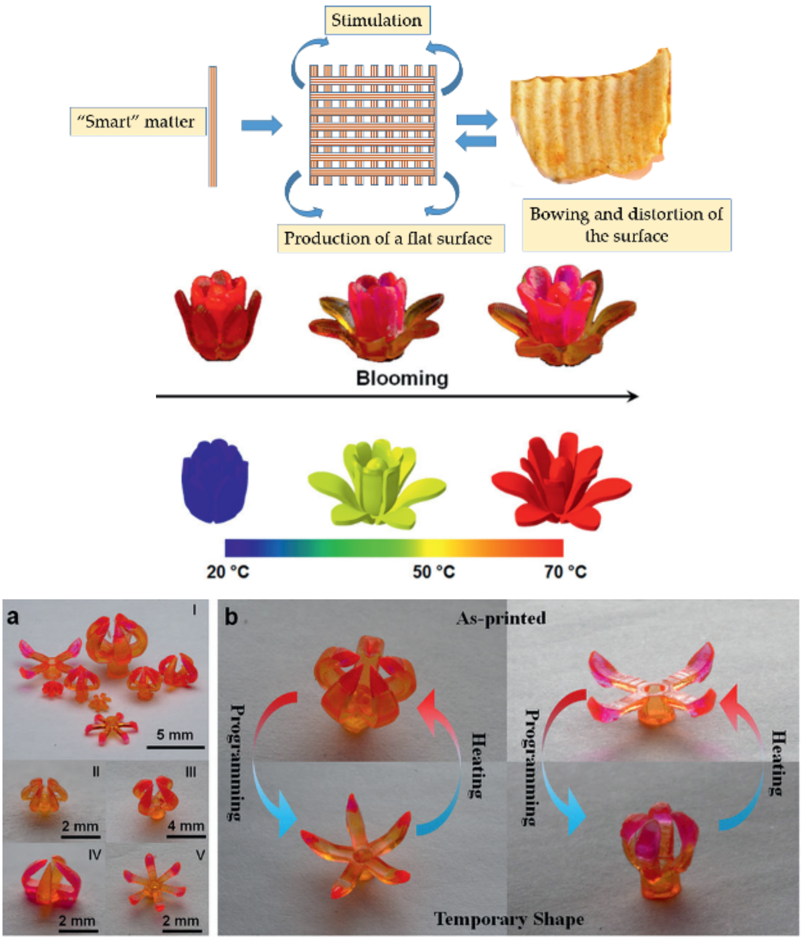

Figure 1.18. *The anisotropy of the material creates a curvature during the temperature change (we could have obtained the same phenomenon with the swelling of bilayer threads charged with cellulose). Complex morphology (here a flower) is generated with developments in the forms of objects. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

Lind *et al*. [LIN 16] used, as material able to be activated, the heart's muscle cells which contract under the influence of electrical stimulation. Figure 1.19 shows the phenomena of compression/expansion enabling a plastic material to bend (see also [JAC 16]).

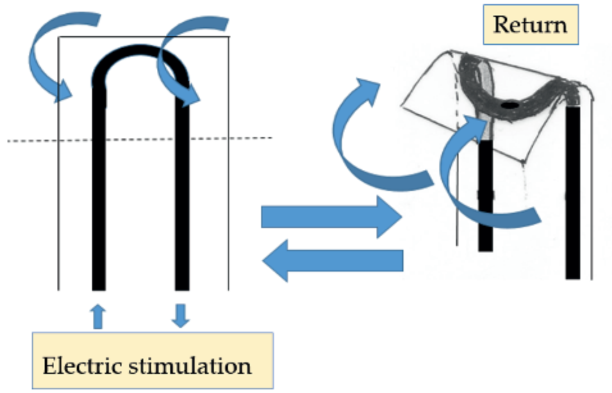

Figure 1.19. *Distortions induced by muscle cells. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

1.3.1.3.2. Active mineral materials

Dufaud, Marchal and Corbel [DUF 02] studied the effect of an electric current containing PZTs with the piezoelectric properties aiming to produce actuators.

1.3.1.3.3. Attempt at synthesis

Table 1.2, extracted from Formlab [FOR 15], enables a comparison between the materials used in additive manufacturing and those currently used in 4D printing.

Additive manufacturing	4D printing
Thermoplastics	Self-assembly materials
Metals	Multi-materials
Ceramics	Shape-memory systems, polymer couplings and systems for continuous change in shape (humidity, temperature, light irradiation and other factors)

Table 1.2. *Differences between the materials used in 3D and 4D printing*

1.3.2. *Physical alterations*

A surfactant is a molecule with a hydrophilic head and a hydrophobic tail, the hydrophilic head is directed preferentially to the aqueous phase, contrary to its hydrophobic tail. Thus, generally, the surfactants are positioned preferentially at interfaces water/air or water/oil. "Their presence stabilizes the interfaces, which implies a local reduction in surface tension. As the presence of surfactants on a given surface locally decreases the surface tension, it is possible to create a surface tension slope. From this results a movement of fluids towards the areas of higher surface tension" [MAR 14], as is shown in Figure 1.20. The addition of surfactant (shown in orange), leads to reduction of the surface tension from γ_0 to γ, which induces a distortion of the surface leading to a flow of fluids (blue arrows) towards the strongest areas of surface tension γ_0.

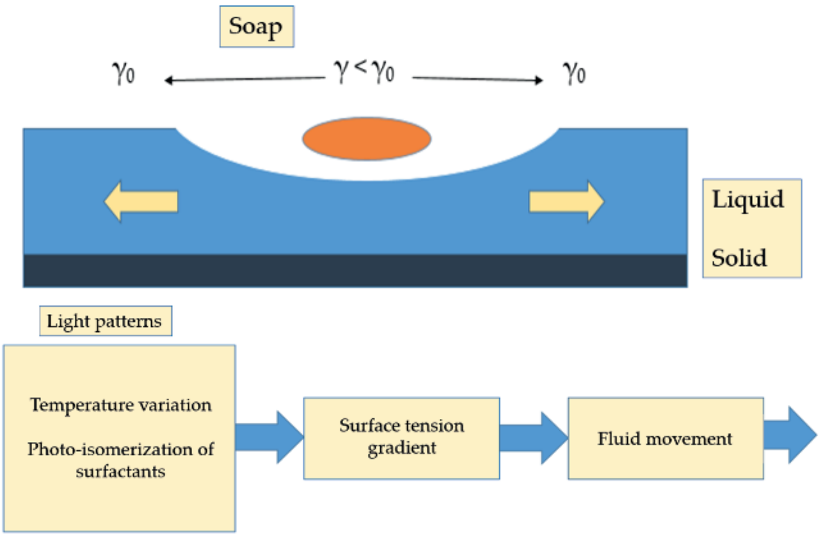

Figure 1.20. *Distortion of fluid induced by surfactants. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

If you use an active surfactant, for example, containing azobenzene, the isomerization Trans-Cis is likely to alter the surface tension, which may enable matter flows. In UV irradiation, the concentration in the Trans isomer diminishes, which increases surface tension. According to the light distribution, a surface tension slope may be obtained with the possibility of having a photo-induced Marangoni effect.

Thus, if a drop is placed on a surface, it will be able to move along this by taking advantage, as Figure 1.21 shows, of the light-induced alterations to surface tension [SHI 99].

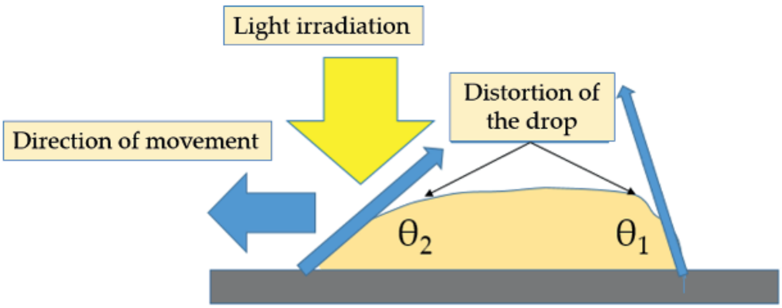

Figure 1.21. *Possibility of guiding the movement of a drop on the surface. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

COMMENT 1.– taking account of the forms of energies called into question, if this technology enables the movement of droplets on the horizontal flat surface, it is only possible through immiscible fluids of the same density (see swimming robots in section 1.4) for an application to additive manufacturing.

COMMENT 2.– with the Trans-BTHA [SHI 99] showed that the irradiation of hanging drops may lead to their fall. From there, envisaging a principle of rapid matter deposition using this process, only involves one step (see Figure 1.22 which represents the developed formula for surfactants).

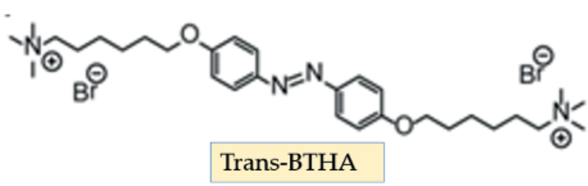

Figure 1.22. *Formula developed for surfactants*

1.3.3. *Distortion of metal parts*

1.3.3.1. *Drops*

By way of an initial example, we quote the article of 3ders.org [3D 14], a sign of the possibility of rotating and moving the droplets of an alloy Ga/In/Se melting at 10°C, floating (due to surface tension) on the water via a magnetic field. The Chinese authors Sheng, Zhang and Liu [SHE 14], of the Chinese Academy of Sciences in Beijing, also succeeded in altering the shapes and sizes of the droplets. It is a matter of seeing how we can access the third dimension. Figure 1.23 shows the effect of an electric field [NEW 15].

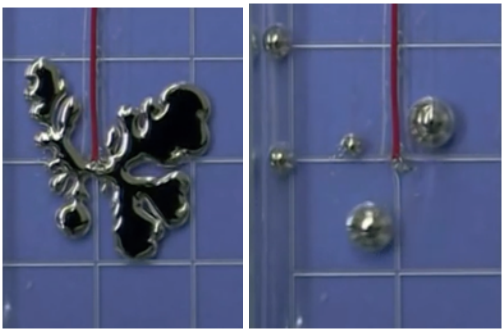

Figure 1.23. *Effect of an electric field (left) on the shape of an alloy droplet*

1.3.3.2. *Magnetic-oriented architectures*

Recently, Martin, Fiore and Erb [MAR 15] linked with photopolymerizable resins for magnetic particles, which can be specifically oriented, by altering the range and the orientation of the given field. This achievement, where two forms of energy are coupled together, enables the production of singular magnetic property structures, at the same time as a possible strengthening of the material by the increase in the sequence of elongated particles in the polymeric matrix. Catheters having neonatal applications are thought to have been produced using this principle.

1.3.3.3. *Shape-memory alloys*

Lastly, shape-memory alloys may be used [BAL 96]. These alloys have several novel properties among metallic materials: the capacity to remember an initial shape and to go back to it even after distortion. There is the possibility of alternating between two previously memorized forms, when its temperature varies around a given critical temperature. Moreover, superelastic behavior enables elongation without permanent distortion greater than that achieved with other metals. Among the main shape-memory alloys, we find a variety of alloys made of nickel and titanium as principal constituents, in almost equal proportions [MEI 12, DAD 14]. Although the name "nitinol" is in fact only the name of one of these "quasi-equiatomic nickel-titanium alloys", this designation is currently used within works in the field to denote all of these alloys, with very similar properties. To a lesser extent, some copper–aluminum alloys also have shape-memory properties [WIK 15]. Although these materials have not actually been used in additive manufacturing, their spheres of application today cover medicine, the aerospace industry, the car industry, women's clothing, the spectacle trade, security and research [NIM 17]. This development was for a long time limited by low durability and their capacity to regain their initial form [CHL 15]. However, everyone remembers an example which is now classic, that of a demonstration by Uri Geller, before an astounded audience of the distortion of spoons produced by such alloys using a thermal effect (L'arrêt public gouverne MENT, [LAR 10]). Several applications in additive manufacturing can be envisaged (by remembering low-volume variations, and also shape changes which may be reasonable). In particular, as Figure 1.24 indicates, metal shape-memory alloys have a specific advantage (relative to polymers). This is their weight/power correlation (hence their use as actuators). This has the potential for exploitation in 4D manufacturing.

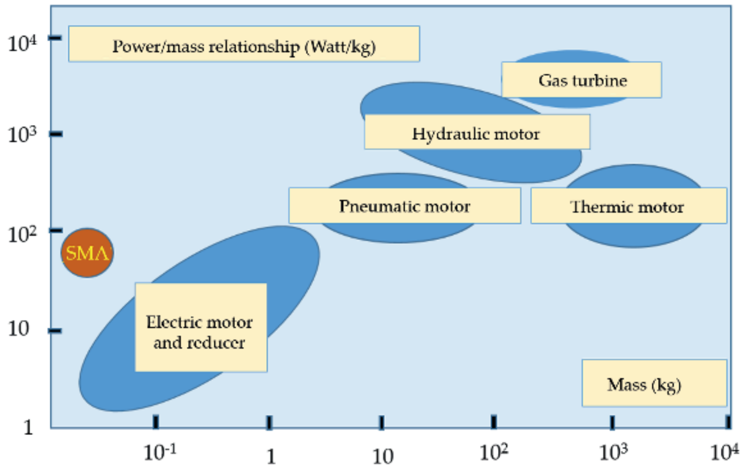

Figure 1.24. *Power–mass relationship of shape-memory alloys (SMA) versus motors according to Patoor [PAT 06]. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

1.3.4. *Conclusion*

The original component of this recently emerged sphere for additive manufacturing resides in the principle of "smart" matter to extract spatial functionality from it (rather than falling within spontaneous self-assembly). At this stage, although "photochemical muscles" have already been the subject of works, to the author's knowledge, the idea which consists of starting from a simple shape, which may easily be produced by traditional techniques (molding and traditional machining) for post-treatment by light (or all other forms of localized energy in space and time) and the idea of obtaining a complex object has not been put forward. It poses the question of having available materials with a high scalability (we recall that for shape-memory alloys, the gap between both forms is generally of the order of a few percent), of being capable of processing the inverse issue. Starting from the basis of achieving a given form, what may in fact be the initial form?

However, although this route appears interesting, we should tackle studies of very simple objects to infer given laws. Evidently, it may be a question of working on surfaces rather than volumes to simplify the situation. For all that there is a risk of the number of influencing parameters to take into consideration being high, leading not to a unique solution for a given spatial resolution, but to a set of solutions. At the same time, we run the risk of the absence of solutions for forms, which are too complex. Thus, starting from a given continuous material, the formation, for example, of a percolating environment could be problematic. These means are explored below.

1.4. A transition to 4D printing: swimming robots

In section 2.2, the possibility of moving a droplet of matter by the provision of external energy was shown. On this principle basis, minuscule swimming robots containing particles of iron oxide linked by chemical bonds are able to move, thanks to a magnetic force [CHE 16]. In this paper, these particles are not intended to self-assemble to create a given object, but from a size of the order of a nanometer, this object should be able to circulate within the vascular system from an external magnetic field ([RAO 15], Figure 1.25). Indeed, the swimming robots' swimming is well controlled, as they move at a low Reynolds numbers: "Indeed the Reynolds number, in respect of flow is very small, of the order of 0.00001, whilst within usual flows (for example that we ought to consider for a human swimming in water) this number is typically of the order of 100 000" [ALO 12]. A swimming robot made up of three spheres, placed side by side, was studied by Alouges, De Simone and Lefebvre [ALO 08]; Naja and Golestanian [NAJ 04] and Garcia [GAR 13] where it is shown that it can effectively swim (slowly) within a three-dimensional flow with a low Reynolds number.

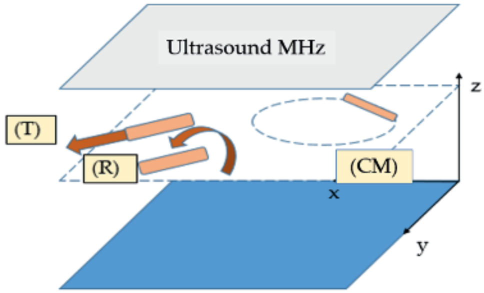

Figure 1.25. *Principle of robot-swimmer movement (various types: translation (T); rotation (R) and circular movement (CM)). For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

Although the researcher perspective is to carry medicines (cargo effect) or to use swimming robots for emitting obstructed vessels (icebreaker effect), it is envisaged to use for a given voxel or convertible matter which, in a state of weightlessness, may be guided upon a given object undergoing construction. The principle of this construction necessitates accurate guidance of each robot, which is considered as isolated from other swimming robots. Otherwise, apart from the feasibility of a precise command, these entities behave according to their density and flow as Lushia and Peskin [LUS 13] and Liebchen, Cates and Marenduzzo, [LIE 16] showed. Yet, in Volumes 1 and 2, the question of voxel size was widely mentioned as it was linked to manufacturing time. Within this system, by using variable-sized particles, it is possible to envisage the construction of nanometric objects or a more significant size using a "multi-voxel" principle, indeed, multi-materials with cargo voxels of varying sizes (see Figure 1.26).

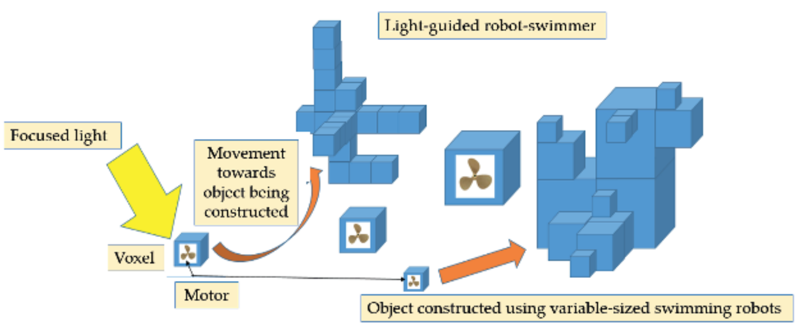

Figure 1.26. *Application of swimming robots to additive manufacturing, for example, light-induced polymerization. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

Various guidance techniques may be envisaged. These are presented in Table 1.3, largely stemming from the works of Rao *et al*. [RAO 15].

Locomotion principle	Materials	Commentary	Bibliographical references
Electrophoresis	Microspheres, bimetallic rods, fibers	Known mechanisms, self-generated electric field, sensitivity to the given medium conductivity, high speed > 100 $\mu m.s^{-1}$	Paxton *et al*. [PAX 05]; Wheat *et al*. [WHE 10]
Diffusiophoresis	Heterogeneous voxel	Self-generated concentration slope with enrichment of fluid support in substances dissolved in the process (difficult to apply in additive manufacturing)	Hong *et al*. [HON 10]; Ibele *et al*. [IBE 09]; Howse *et al*. [HOW 09] and [HOW 07], Clément *et al*. [CLÉ 16]; Clément [CLÉ 16]
Bubble propulsion	Elongated forms	Catalysis for the decomposition of a reagent producing bubbles, with an uncertain trajectory	Wilson, Nolte and van Hest, [WIL 12]; Gibbs and Zhao, [GIB 09]; Solovev *et al*. [SOL 09]; Zhang *et al*. [ZHA 15a]
Electric field	Microparticles	Localized electrolysis in water producing bubbles, accuracy difficult to achieve and slow movements	Calvo-Marzal [CAL 10]; Loget and Kuhn, [LOG 11]
Magnetic rotation	Flexible stretched out shapes	Swimmer rotation within a magnetic field inducing locomotion, accuracy possible but slow speed	Gao *et al*. [GAO 10] and [GAO 11]; Ghosh and Fischer [GHO 09]; Tottori *et al*. [TOT 12]; Peyer *et al*. [PEY 12]; Li *et al*. [LI 16a]
Ultrasound and acoustic waves	Metallic particles	Conversion of moving energy, uncontrolled behavior, high speed	Wang *et al*. [WAN 12]; Ahmed *et al*. [AHM 16]
Photochromism	Elongated form	Spiropyranes or azobenzene compounds are likely to have light-induced Trans—Cis transitions leading to the movement of flagellum	Li *et al*. [LI 16]; Huang *et al*. [HUA 15] Stanton, Trichet-Paredes and Sanchez [STA 15]

Table 1.3. *Various forms of propulsion of swimming robots*

For magnetic propulsions, Li *et al*. give some indications as to the speed of movement according to means of action. This data is shown in Table 1.4.

Nature of magnetic stimulation	Speed in μm.s^{-1}	Commentary
Internal particle oscillation	30.9	Movement as if a fish
Oscillation	6-14.4	Boat (scull) movement
Rotary feed	18-40-180-250	Screw movement

Table 1.4. *Speeds of movements obtained by Li et al. [LI 16a]*

In the case of azobenzene Trans–Cis linked to a light-induced transformation, the motor is linked to the voltage induced by the change in form of azobenzene within a sandwich structure, such as that shown in Figure 1.27. There is, with this type of mechanism, a means to control partial rotation of a fixed flagellum on this motor as shown in Figure 1.28 ([HUA 15], see also [EGU 16]). Although the constraints are not homogeneous within the different layers, a moment of forces appears and the thin layers may then curve spontaneously, subject to the action of external forces. Thus, by exploiting these constraints, a new "self-winding" material produced with cross-linked polydimethylsiloxane (PDMS), a transparent and biocompatible elastomer and silicone oil was able to be produced.

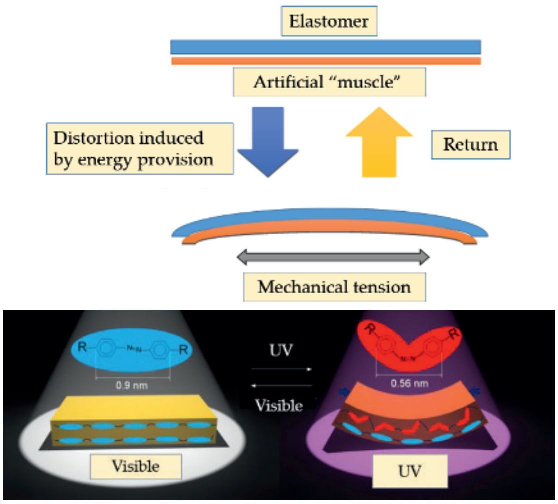

Figure 1.27. *Principle of operation of photochemical motor. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

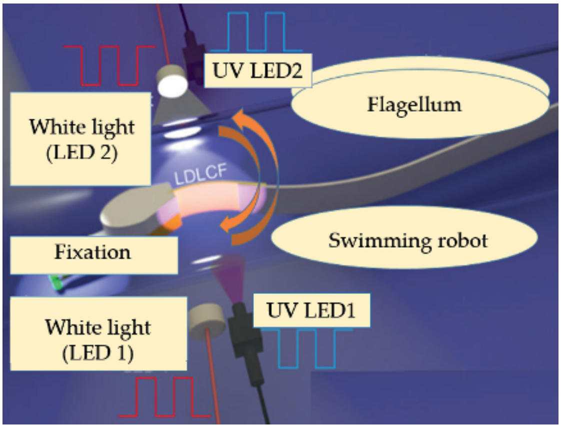

Figure 1.28. *Relationship between photochemical motor and its flagellum. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

In their publication, Khoo *et al.* [KHO 15] have produced a synthesis of applicable processes in terms of "smart" matter. A summary of their conclusions is comprised in Table 1.5.

Materials and structures	Approximate resolution	Commentary
Piezoelectric nanocomposites	5 µm	Production of electric charges when a given constraint is both applied (and reciprocated). The support material is a polymer such as the PEGDA (Poly(ethylene glycol) diacrylate)
Memory-shape alloys (NiTi)	/	Change of shape induced by temperature (problem of hysteresis to take into consideration)
Memory-shape polymers	20 µm	Distortion of a bi-component system with various properties (bimetallic strip effect) induced by the temperature or by the light (heating)
Bimaterial system including a dielectric elastomer	30 +/- 10 µm	Electric activation and bimetallic strip effect
Polymeric composite and origami	45 +/- 15	Movements induced by temperature

Table 1.5. *Several examples of applied smart matter in additive manufacturing*

1.5. 4D Printing

A simple means to produce a given object sensitive to stimulation of energetic origin is that of the rubber balloon, the shape of which increases in size as the pressure of the gas filling it increases. Another example which is even better adapted to the demonstration comes from Tibbits at MIT [MIT 16]. He places objects in a flask which after shaking gather together to create another object (the traditional problem of creation of order from chaos, which will be widely mentioned in the part devoted to bio-printing). Figure 1.29 inspired by Tibbits and Thomas [TIB 13], and published by Leist and Zhou [LEI 16], shows this.

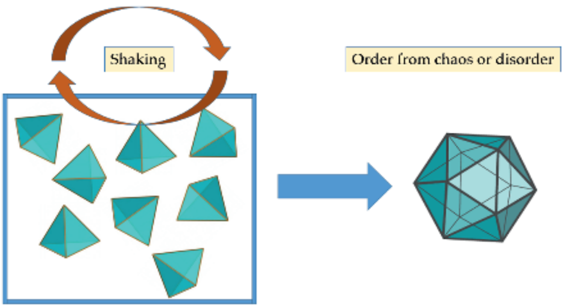

Figure 1.29. *Creation of order through disorder (general concept according to [LEI 16], citing Tibbits and Olson). For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

From there producing more sophisticated components such as the actuators shown in the previous chapter or folding systems including internal expansion enables interesting developments of shapes, such as those in Figure 1.30 by Yuk *et al.* [YUK 17] which is an object produced by additive manufacturing, the shape of which may be altered by the effect of hydraulic pressure. There are only extensions of this concept. It is the same, for example, with the works of Ballandras *et al.* [BAL 96] where a structure produced by 3D printing was connected to a structure-memory alloy to manufacture an actuator. According to Pimenta [PIM 15], a humanoid on a genuine human scale may be directly printed at home, thanks to a 3D printer. All the parts have been designed to print without any particular difficulty and self-assemble either by using screws or clamps. Other examples of the same type exist (see, for example, [DEM 14, ATL 15, MAR 12, STR 13, MEH 14, YIM 07,

KOT 98, KEA 13, YIN 16, HAR 16, PRA 16, ENG 17]). However, is this 4D printing?

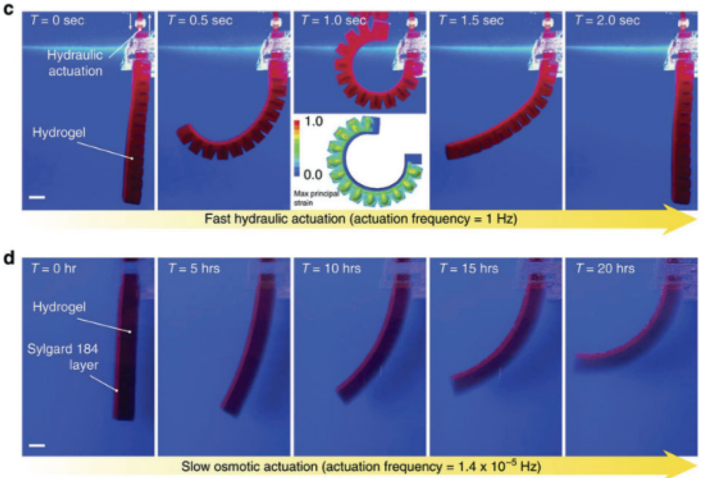

Figure 1.30. *Hydraulic actuator. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

The concept of 4D printing hypothesizes that the given object is adaptive through integration and less through the juxtaposition of the various components, even when produced through 3D printing. The fourth dimension is time and/or the given functionality [PIC 17]. The 4D technology concept enables the printing of objects which themselves change shape and self-assemble with time. The dividing line is therefore not so simple to find especially around the first aspect of subject framework: changing shape by itself, as every robot does, even every machine does. Moreover, the German firm BMW [BMW 16] in its research on vehicles of the future has developed a prototype relying upon the concept consisting of spatial conversion of the vehicle structure according, for example, to its speed. The idea of 4D printing tackles the assembly and the installation of products/objects which may be able to self-assemble and their implementation in environments facilitated by "Programmable Matter". As has already been suggested, programmable materials which build themselves thus make superfluous the need for assembly plants and heavy industrial installation procedures. The robotization, the core of productivity gains of the 20th Century, would thus be integrated to the products themselves. Nowadays, even Figaro Magazine (a French Newspaper) is speaking of 4D printing [4DP 15]!

Independently of this problem of definition, it appeared useful to investigate, through the library site of the University of Lorraine, what is produced on the emerging theme. Figure 1.31 shows the results from the key words "4D Printing". Approximately 200 publications make up the available "stock", but what this figure shows is that it is an over-exponential development, indeed illustrating an emergence, with the initial publications in the sphere being around 10 years old or more. Moreover, Leist and Zhou [LEI 16] estimated that the 4D printing market will be worth US$63 million in 2019 and nine times more in 2025 (values that it is appropriate to consider relative to 3D printing estimates of US$5 billion to US$30 billion/year on this latter date).

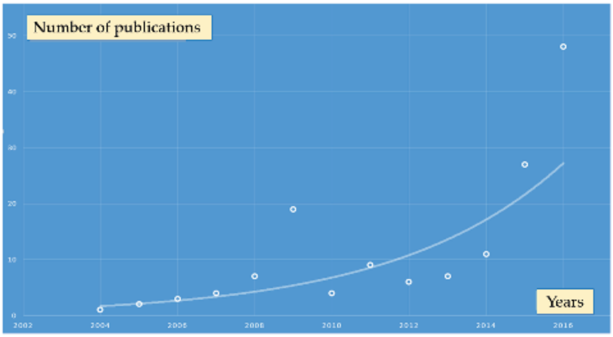

Figure 1.31. *4D printing publications (curve: exponential through given optimal points)*

1.5.1. *Automation and robots*

D. Rus of MIT succeeded in demonstrating the capacity for robotized voxels to self-assemble in a given form, by using a type of distributed algorithm, without a central server, in the manner of cellular automation. His team developed "m-cubes" liable to roll on each other and also to climb on each other [SUS 14]. In the first stage, it is possible to envisage, as MIT [MIT 13a] showed, articulating on the basis of origamis, sets of voxels placed among each other to enable the introduction of temporal development in the overall geometry of a given object. The photographs in Figure 1.32 give examples of such developments.

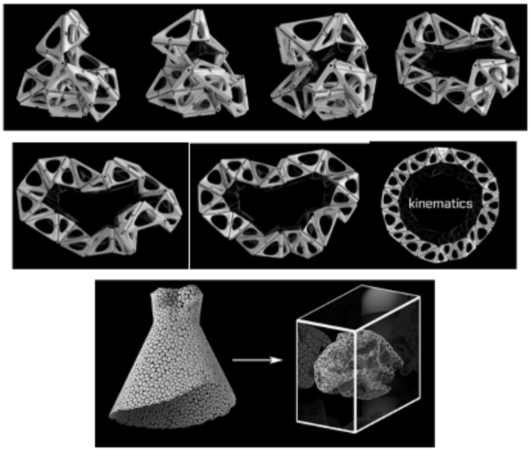

Figure 1.32. *Distortions from movable voxels (with the consent of the organization Nervous System; http://nervo.us)*

The same institute [MIT 13] shows that using simple examples, it is possible to "control" not least by visual recognition. This idea, validated by Figure 1.33, relies upon the movement of cylinders by using, on a two-dimensional basis, a means to take an impression of a cornice or like certain toys which permit three-dimensional construction. If we add a digital command to the movement of each cylinder, there is a means of making matter directly or indirectly "programmable". However, in these two examples, which serve to introduce the subject, matter is inert. The idea consists of going slightly further with the approach by "introducing the 'smart' element" directly into the matter, at finer scales than this figure.

Figure 1.33. *Reversible sculpture toy [CDI 17]*

Figure 1.34 from MIT [MIT 13a] shows the intrinsic complexity of such a system which will not be able to be used at lower scales due to the high number of actuators which it would be necessary to put together within a finished volume. There is also the issue of the size of voxels to take into consideration.

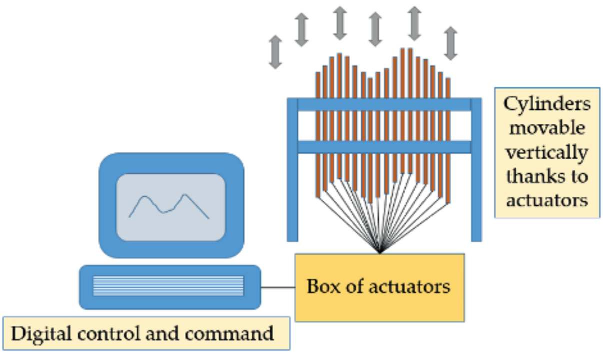

Figure 1.34. *Experimental set-up of the MIT [MIT 13]. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

A 4D technology still in the process of assembly of programmed development of robots [GE 13, GE 14, THI 14, BOU 14, LI 15, BAK 15, CRA 15, SAV 14, THO 14, FIS 14, BÜC 15, JIN 11, AZN 12, MAC 10, MAC 12] and other references) may consist of creating "catoms", capable of self-assembling magnetically to produce any object [GIL 12, KNA 10]. From 2007 onwards, researchers at the MIT had created prototypes from cylinders approximately five centimeters in diameter, which have subsequently been somewhat miniaturized. They have manufactured cylindrical "catoms" of around 1 mm in diameter, capable of communicating and receiving energy. There is one small problem, they do not yet move [SUS 14, HAR 12, HAR 15], but the cubes from the team Rus at MIT meanwhile move to create genuine three-dimensional structures, however, having a number of voxels which are very limited, as Figure 1.35 shows [GIL 12].

In this project, a new "smart" system manufactured from a component material and a software approach enables the creation of a set of programmable objects (taking a "smart" collective shape). The material component is a micro-robot manufactured in series which uses the same IT mechanism, the same energy, communication, and adhesion and locomotion distributions. The software approach enables reconfiguration of a set of micro-robots for a given end.

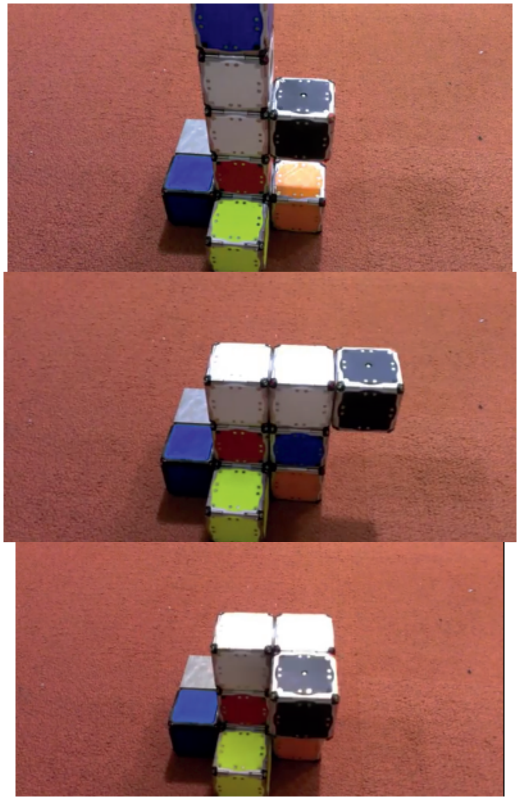

Figure 1.35. *Example of 4D printing using cubes [SUS 14]*

A demonstration was carried out with the assistance of a limited number of programmed mechanisms. To go further, it extends to the resolution of the following scientific and technological challenges:

1) The change of scale, while maintaining the same levels of movement and adhesion performance, to achieve compatible sizes with a given applicable need, although the number of robots becomes important. We can cite the project of the research unit FEMTO-ST in Besançon (France), which is engaged in this research direction. “For the material, we will create a genuine 3D micro-robot through LSI/MEMS integration between the chip and a distortable substrate. Our main manufacturing process will concern the actuator, which groups together all of the necessary micro-robot functionalities” [BOU 16].

2) The control of the cooperation of movements of a set of robots moves in an autonomous way, without affecting other robots. However, to produce a complex object with an "acceptable" accuracy, it is necessary to have a large number of "nano-robots" engaged within collective manufacturing, with the difficulties of multiple interactions between a multitude of micro-robots before avoiding one another, as the following Figure 1.36 shows [YEO 15].

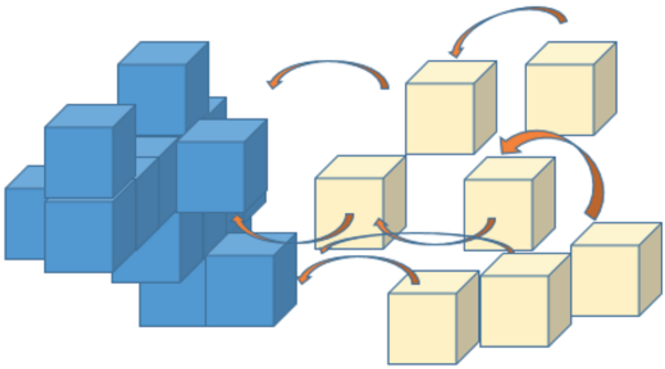

Figure 1.36. *Collective approach to interdependent movements in object production and movements which may be concerted*

What is shown on these figures is a form of tangled causality which may be observed in a number of mechanical systems [SÈV 05], with a nonlinearity and largely unpredictable effects which may appear, especially if the number of independent voxels at the start has increased. "For software the challenge is to intensify the number of components which may be efficiently controlled with a single easy-to-understand program" by the user. "We are going to create Foxel, an invariant descriptive recursive language implemented within a logical programming language. In addition, we will devise 'Meld', to create programs which are naturally distributed, tolerate breakdowns, and which can also progress towards clear evidence. This project follows the 'Claytronics' project initiated by Intel and the University Carnegie Mellon, then co-directed with FEMTO-ST" [BOU 16].

3) The energy supply in increasingly small objects, by reducing the size, we reduce the space devoted to energy storage, with a charge that varies with the cube of the average dimension of the object.

1.5.1.1. *Concluding comments*

"Although the necessary progress as regards hardware is yet to be accomplished, it does benefit from a fairly simple road map: the strategy of miniaturizing! The creation of distributed software is, for its part, even more uncertain. We have been working on this subject for more than two decades. There have been an increasing number of experiments, as much in the real as the virtual world. However this type of process is still an economic study without practical applications" [SUS 14]. Nevertheless, going from a few centimeters to a few hundred µm corresponds to change in scale of approximately 100, or for a three-dimensional object having a factor of 10^6. It is nowadays difficult to imagine a process involving energized micro-robots and having a collective assembly program and moving without causing collisions. Perhaps it is necessary to investigate the movements of a given number of social insects (bees, ants and termites) who know how to move within a complex environment asking the following: "Why is the group moving coherently whilst each individual seems to be autonomous? How are the activities of all individuals coordinated without supervision? Behaviorists studying the behavior of social insects observe cooperation within colonies as being subject to self-organization: often, it results from interactions between individuals" [BON 00]. That which nature knows how to do with ants, man begins to know how to do. However, the application phase, within 3D or 4D additive manufacturing, is only taking tentative steps. Nevertheless, although we can content ourselves with less precise forms, it is not impossible to envisage a very flexible process to produce "smart" objects. It would certainly be useful to monitor this issue.

1.5.2. *Origami*

Sheets of origami when the author was young generally had a square shape (but this was not always the case). The various shapes are obtained by successively folding the given sheet. In theory, it should not be cut. The given origami can represent an animal, a plant or an object, but can also represent geometric forms which are simple or complex: these are the so-called "modular" origamis. Although this Far Eastern expression is introduced into this chapter (apart from DNA origami), it is in order that we can create a three-dimensional shape with an initially flat structure.

For centimeter-sized entities, it is possible to position the traditional actuator at given folds. Their rotation enables the movement of given planes and, consequently, the movement of origami which can, through an adapted control, become a robot-origami. Nisser *et al*. [NIS 16] used origami techniques to create and deploy four robots constructed from a single sheet. These were made from several layers of different materials. This process would enable facilitation of the deployment of robots in swarms for various applications. Shape-memory polymers and the origami principle are used to create robots from a single sheet, enabling precise folding control thanks to mounted sensors. For the moment, the capacities of small origami robots are fairly limited. Their top speed is approximately 10 mm per second and their autonomy approximately a few minutes [ACK 16]. This coupling example between robotics and "smart" matter is indeed not unique.

Very recently, researchers at MIT presented a "robot miniature" prototype capable of adopting various shapes, of moving and of transporting loads by pushing them. This small ultra-light robot, which is remotely controlled, can close in on itself to adopt different postures. These include walking, swimming and moving heavier loads than the robot itself (before finally self-destroying within the acetone substance). What is interesting is that to produce this movable 3D object, it is sufficient to print programmable matter using a traditional 2D printer.

"This machine is only made up of two extremely thin layers of polystyrene (or paper) between which are a magnet and a sheet of PVC measuring with a side measuring 1.7 cm in length. These relatively simple materials enable the inventor to produce remarkable feats when working within a suitable environment. Thus, when the device is placed upon a heated area at the right temperature, the sheet of PVC of which it is made folds in less than a minute according to pre-determined structural lines. Then, thanks to the combined action of four solenoid mechanisms arranged below the surface, the "robot" is able to move in any direction at an average speed of 3 to 4 cm per second. Moreover, it is capable of carrying a weight of 0.6 grams, climbing a slope and cutting a chosen path through various obstacles" [RAN 15].

In the same spirit, other works have been published: Rus at the MIT shown by Hardesty [HAR 15] on origamis; see also Khoo *et al*. [KHO 15], Ge, Qi and Dunn, [GE 13, GE 16a], Ge *et al*. [GE 14]; Hawkes *et al*. [HAW 10]. The following Figure 1.37 shows the potentialities of these microelements distortable due to one or more localized magnet(s) localized around the given object, having cell components with the Peltier effect and controlled movement by magnetization determined at the surface. Optic or thermic control can be used.

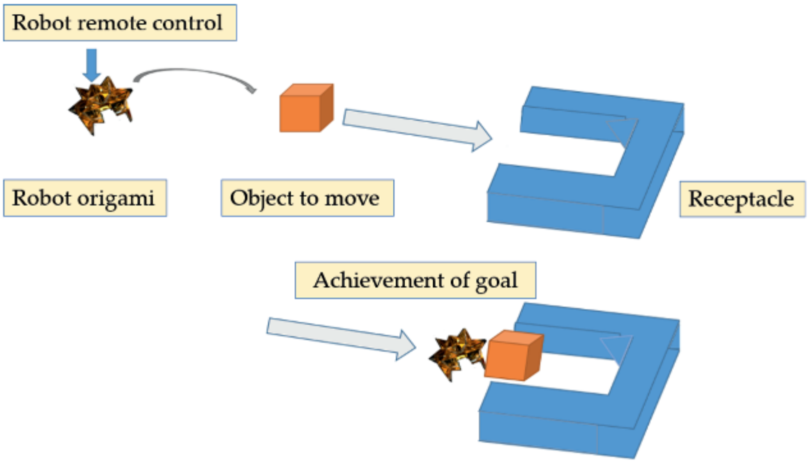

Figure 1.37. *Robot-origami moving a given object (from the start to reaching the objective). For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

Using this system (which has been subject to other publications by Daniela Rus' team such as: Felton *et al*. [FEL 13, FEL 13a], An and Rus, [AN 14], Hawkes *et al*. [HAW 10], Onal, Wood and Rus, [ONA 13], we find ourselves in the same difficulty of self-assembly collective production, having a three-dimensional object responding to instructions. However, it is possible to use light in Figure 1.7, the possibility of controlling micro-robots to move voxels. This enables the manufacture of an object or its completion using the matter of an existing object. This system might be used advantageously in areas which are difficult to access or even dangerous.

Figure 1.38 shows the principle which stems from Ge *et al*. [GE 14] resumes the same idea with thermomechanical systems. In their publication, the relaxation time is of the order of 0.4 seconds. For an adapted design, there is the possibility of achieving more complex forms provided that the object can be described on a plan before being folded (see Figure 1.40 stemming from Kimionis *et al*. [KIM 15], see also Tenzeris *et al*. [TEN 16].

Examples stemming from Py *et al*. [PY 07] use the influence of water and surface tension illustrating the concept within another framework, that of altering surface tension (see Figure 1.39). Others use porous high-power displaced polymer materials distorted by an electric field [HAN 04].

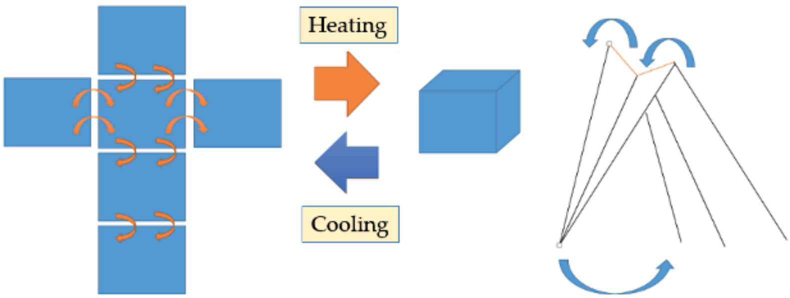

Figure 1.38a. *Principle of operation of a given thermomechanical origami. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

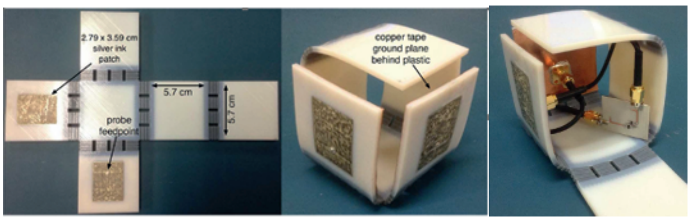

Figure 1.38b. *RF antenna produced by additive manufacturing and multi-materials producing origami to achieve a 3D structure (with the permission of M.M. Tenzeris). For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

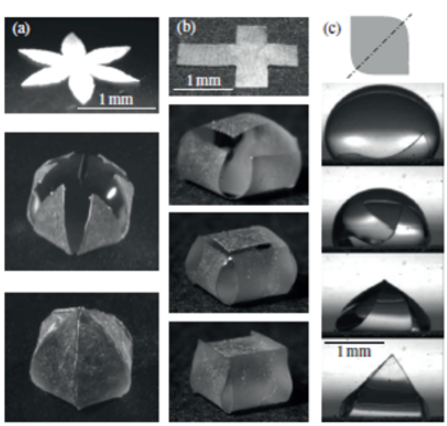

Figure 1.39. *Development of PDMS membranes in the presence of water per the initial template produced as flat (with the permission of Benoît Roman)*

1.5.3. *Octobot*

Mazzolai and Mattoli [MAZ 16]; Wehner *et al*. [WEH 16] printed the body of a silicone octopus in 3D. They used tanks of oxygenated water and catalytic reactors which store localized hydrogel-based inks in their tentacles. These contain particles of platinum, which catalyze the controlled decomposition of oxygenated water which frees the oxygen serving as a thruster. Pistons and gates regulate the movements of gas to propel the robot. "Being less than 2 centimeters in thickness and almost the size of a human palm, the Octobot can operate for 4 to 8 minutes before getting to the end of its supply of oxygenated water. Larger versions of this robot (shown in Figure 1.40), may be capable of remaining active for longer time periods upon each replenishment" [DEV 16].

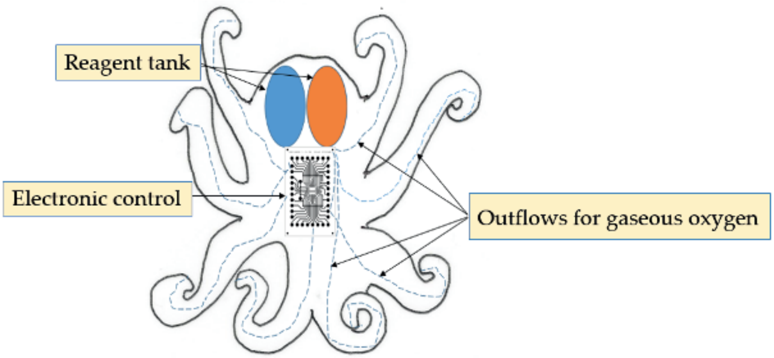

Figure 1.40. *Approximate drawing of an "Octobot". For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

1.5.4. *Massive objects*

Figure 1.41 stemming from an idea of Yang *et al*. [YAN 16] and Ge *et al*. [GE 16a] highlights the aptitude of memory-shape polymers involved in additive manufacturing to produce actuators (see also [SEN 16, LU 17, ZAR 15, YAN 16]). This is completed by the actual demonstration of the idea on the figure as shown at the bottom.

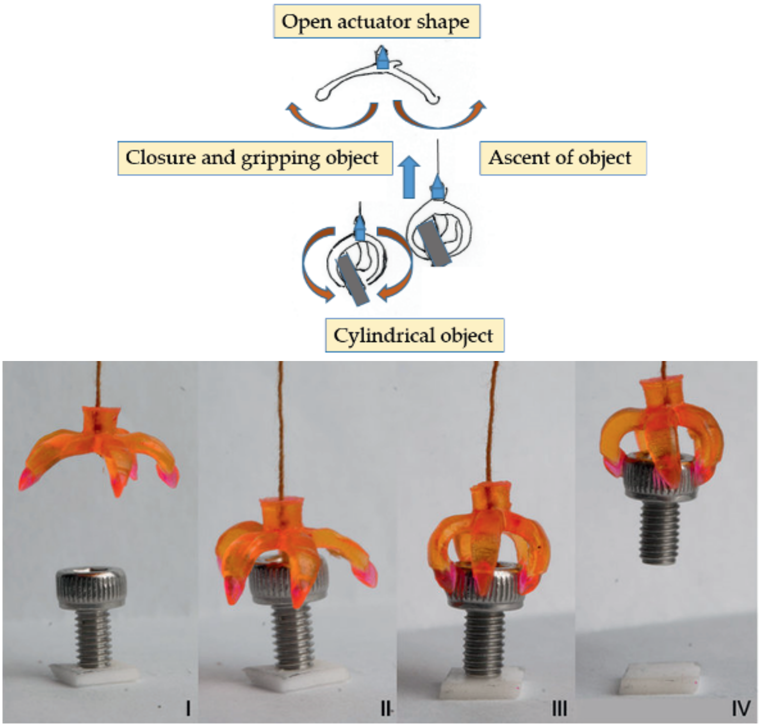

Figure 1.41. *Actuator thermically agitated by a shape-memory polymer (image supplied by Kevin Ge [KEV 16]). For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

Although it is possible to calculate the shape of a 3D object fairly easily and to envisage it developing, the experiments shown to illustrate the significance of this emerging domain essentially concern elongated objects or those in the form of layers. Concerning massive objects, such as that of the chosen provocative example in Figure 1.42 to illustrate the idea, the problem to process becomes more delicate, as it involves:

– a radical change of shape;

– a radical change in functionality.

Since it does not appear possible to the author to resolve the complex problem of the conversion in this figure, posing questions linked to the nature of the material (so that it becomes distorted and that it has good machining properties), energy provision (within the particular material) and the process, it appears at this stage necessary to commit to a "minimalist" demonstration. This happens by choosing an apparently simple example of that of the passage from a sphere to a cube, as shown in Figure 1.42. To simplify the problem, we could content ourselves with a form of surface modeling of these two entities (which are then assumed to be defined by their surface). In making up the base object with line segments (actuators, the length of which may be activated by the provision of remote energy), it is already possible to consider a given number of interrelationships to take into account between these various segments. This is because the reduction of a segment (pixel) affects others, being those which are the closest and less and less the others which are the furthest away.

In a recent article [ALA 16], the conversion to apply to each pixel of a cube to achieve a sphere, or the inverse conversion, can be calculated fairly easily using morphing methods which the authors have developed. "The work carried out shown in this publication indeed enables us to go from one shape to another using the same meshing: we mesh the so-called 'reference' shape. They calculate the conversions to apply to it to obtain the final desired form. The advantage from a digital viewpoint is that both forms have the same meshing, that is to say the same number of nodes and the same connectivity matrix, which enables us to deal with a digital problem without having to re-input the same data each time" [BEL 16]. From a principle viewpoint where there is the preservation of volume, the work of mathematical modeling around the relevant distortion to apply to each surface component was produced by Rohmer [ROH 14] and Rohmer, Hahmanna and Cania [ROH 15]. Sophistication linked to the non-interpenetration of surfaces (the example of an arm folding) has been developed by Vaillant *et al*. [VAI 12]. This scientific activity takes its origin, at the time of the animation of virtual characters of the calculation of a credible skin distortion at the joints. The production phase of a distortion is usually called "skinning".

There is thus, for this "very simple" example, the possibility of modeling the local energy path (for each pixel) to achieve a given conversion (with at the end of the chain, the possibility of analyzing the feasibility of an object from a given form).

This work is to be undertaken before addressing massive objects, which may however, as a first approach, be represented by a meshing close to that proposed on the surface of the object, by using a three-dimensional network.

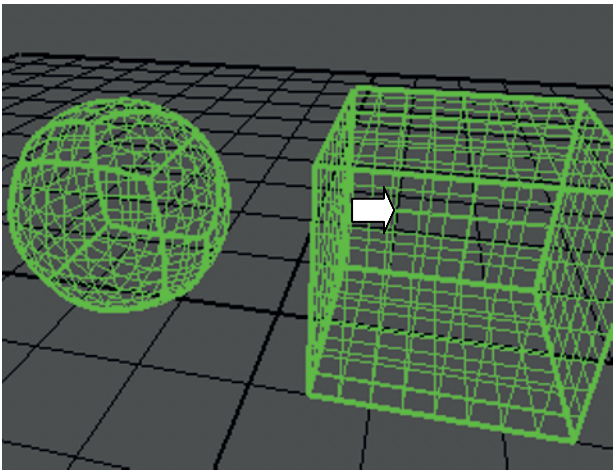

Figure 1.42. *Desired transition from a sphere to a cube. For a color version of the figure, see www.iste.co.uk/andre/printing3.zip*

1.6. Conclusion

The original component of this emerging sphere lies in the principle of voluntarily making the matter “smart”, so as to extract a spatial functionality (rather than falling, at the present time, however exciting these concepts are within robotic self-assembly. This poses the question of the size of voxels involved and their cooperativeness in the given movements). At this stage, if “photochemical muscles” have already been the subject of works (which may be advantageously considered in depth), to the knowledge of the author, the concept which consisted of starting from a simple shape, easily produced by conventional techniques (for example, molding and conventional machining) so as, after processing by localized energy in time and space (for example, with light), to achieve a complex object. This is yet to be explored. It poses the question of having available materials with a high expandability (we recall that for shape-memory alloys, the gap between both forms is generally of the order of a few percent). It is then a matter of being able to process the converse issue. Starting from the basis of achieving a given shape, what might the initial shape actually be?

However, although this route appears interesting, we might acknowledge the need to tackle studies of very simple objects to infer given laws. Seemingly, it may initially be a question, as has been suggested, of working on surfaces rather than volumes to simplify the situation. For all that, the number of influencing parameters to take into consideration risks being higher leading not to a unique solution to a given spatial resolution, but indeed a set of solutions (and, at the same time, the absence of solutions). Thus, starting from a continuous material, the formation, for example, of a percolating environment may be problematic. However, it should be studied.

Table 1.6 collates all of the elements to master to develop this process (classified as + for "easy", = "feasible", - "existence of an impediment", -- "sticking point") with commentary.

Issue	Attainability	Commentary
Processes	??	Spatial transformation of matter induced by energy (electromagnetic, photochemical, heat and other forms of energy). Ideas are yet to be researched, the sphere is practically uncharted (outside of aspects of robot self-assembly) with the emergence of work on the so-called "swimming robots" (micrometric space and biomedical applications)
Flexibility	-	Possibility of using reversible reactions functioning by analogical means (photo-mechanic effects around polymers) or digital (shape-memory alloys)
Materials	=	Function of process (or processes)
Modeling	--	Outside of author competence (see the inverse problem)
Converse problem	--	Complex research avenue to explore, probably starting by studies of surface distortion rather than volume. Ideas are sought in this area
Interdisciplinarity	-	Collaborative encounter between photonics, photochemistry, mechanics, automation engineers, and other domains to move towards applications (or other processes)

Table 1.6. *Elements to bring under control for development of the 4D process*

To reach the objective of a good control of this futuristic field, there is a need for individuals originally from unconnected disciplines to explore these new boundary objects which appear capable of a genuine future application. The expected convergence (for which associated lines of questioning will be subject to discussion in Part 1 of Volume 3) necessitates that we depart from "conventional" means of innovation research. In exploring this axis, "accurately", nowadays, capable of illustrating its capacity for proofs of creative concepts, but still limited in number, these methods appear to have reached their limits. Around such an original and key theme, still uncharted, it may be more interesting to consider what enables "disruptive" innovation. This is the only way to think otherwise about scientific developments around new applications. Nowadays, we are therefore witnessing types of research "Uberisation" and 4D printing deserves better.

France Stratégie has stated that "A wave of innovations is materializing which is creating new markets, and is overturning the economy in numerous sectors. Faced with these disruptive innovations, the state must define its position: a wait-and-see stakeholder possibly having a considerable economic and social cost which could lead to a loss of sovereignty" [CHA 17]. However, within too many management teams, risk-taking is too often perceived as an interesting topic of conversation, not often appropriate, even esoteric, "particularly for researchers and academics, but with no real significance on daily activities in terms of strategies, and without any impact on their decision-taking processes" [LOU 17]. This idea is confirmed by R. Quirion, the President of the Comité de l'Évaluation (Assessment Committee) of CNRS who wrote in [CNR 16]: "At the CNRS, as elsewhere, interdisciplinarity is a genuine paradox. Although it is one of the most pluridisciplinary institutions to exist, [CNRS] must nevertheless make substantial efforts to stimulate interdisciplinary research. This indicates that there exists even within the institution, barriers which must be lowered if we wish to take advantage of the wealth and diversity of science generated".

To conclude for the time being, let us leave the responsibility to Alain Le Méhauté [LEM 14] to provide his contribution for this chapter (which actually he is not aware of as such). He wrote: "We estimate the economic potential of this future technology to be several hundred billion dollars. We know that hundreds of billions of dollars equates to between 1 and 4 million jobs… besides knowing that the control of this technique induces multiple derivative creations, including conceptual ones. We cannot regret the intellectual poverty of clichés, and the absence of ambition which we make clear in our public and private research through result-based programs. It is often very sad to read the content of guidelines into which the research will fall within and compromise the future, if it wishes to be financed".

It is a question of creating a little piece of nature to do so, and then to do so with the so-called "programed" objects, rather than remaining does so, by only operating in respect of opportunities (even if that only gets a foot in the door). Upon an open passionate subject, French academic research cannot set aside this accessible domain which bears a number of applications that are as yet unknown. Let us therefore try to prove Alain wrong.

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