

The status, challenges, and future in design for 4D printing

Akbar, Print Application Engineer, Provin Technos, New Delhi

Abstract

The combination of scientific developments in additive manufacturing (AM) and smart materials (SMs) has enabled the formation of a new multidisciplinary study area: 4D printing. Stimuli-responsive materials are at the heart of this technology, and they hold great promise for bringing about transformations in the functionality, shape, and properties of everyday products. Researchers from many different fields have used proof-of-concept 3D printers, either commercially available or custom-made, to explore a wide variety of stimuli-SMs correlations by taking advantage of these capabilities. Despite many efforts, advances in 4D printing are still needed to match the industry's robust system applications. The purpose of this study is to draw attention to the current state of 4D printing as well as its limitations and design-related hurdles. It begins by reviewing the basics of SMs, processes, stimuli, and AM before moving on to a synthesis of significant research works on 4D printing, with an emphasis on the technology's current state and its scientific, technical, and institutional limits. A research agenda for engineering design and cross-disciplinary design is also outlined in the article, highlighting potential and difficulties from a variety of viewpoints. The study's findings may help guide research over the next decade in the direction of creating smart products that can benefit both consumers and businesses.

Keywords: 4D printing, Additive manufacturing, Smart materials, Design, Engineering.

1. Introduction

The rationale of economic success and the mechanisms underlying value creation are both upended by Industry 4.0's new paradigm for industrial strategy. Flexible and individualized mass production, real-time coordination and optimization of value chains, reduction of complexity and costs, adaptability and the emergence of new services, and high-performance business models are just a few of the possibilities and benefits of the Industry 4.0 concept [1,2]. Flexibility and localization of manufacturing near points of demand are replacing economies of scale and volume in the industrial sector. This indicates a responsive change in response to the user. Engineering and production need to be more foresighted by shifting their rationale to focus on the user rather than the product itself. Additive manufacturing (AM) already fits nicely into this reasoning [3] since it lowers the cost of the complexity carried by the digital component of a product or machine with a standardized base. 4D printing has a place alongside the other pillars of Industry 4.0, namely soft robotics and the Internet of Things (IoT), thanks to the additive manufacturing (AM) technology's ability to create items' structure and functionality. The growth of the specific axis created by 4D printing is thus a key strategic integrating element in the technical advancement linked with the Industry 4.0 agenda.

The goal of 4D printing is to create technological inseparability between the 3D printer and the part it is making, with the 3D printer responding to inputs rather than following a predetermined sequence of steps. It is, thus, a big disruptive integration that has the potential to change the manufacturing sector if it can meet industrial criteria (otherwise, one would have to look for specialized applications, which would imply a more modest attractiveness and a less stimulating research drive). Physical products and objects, and especially design and engineering-related activities, have always been influenced by the evolution of production methods, materials development, and manufacturing techniques [4-7]. This is because AM allows for the creation of multi-functional, multi-material parts with complicated geometry, whereas conventional subtractive and formative manufacturing-oriented design techniques did not. Researchers over the past decade have opened the door towards the integration of smart or active materials, i.e. materials that are capable of changing shape, functionality, and property once stimulated by user input or autonomous sensing in their usage environment [12,13]. This manufacturing disruptive technology is primarily focused on complex shapes and high-resolution issues. Therefore, the digital printing of SMs or smart voxels leads linked study fields and scientific communities to new multidisciplinary and convergent research paradigms, namely bio-printing and 4D printing [14,15]. This study focuses on the latter topic.

Featuring AM-enabled transformation capabilities, 4D printing has gained great attention from researchers in disciplines such as chemistry, applied sciences, physics, materials science and mechanics. The proof-of-concept oriented contributions in this burgeoning research field are both intriguing and multifaceted, as they combine AM deposition techniques, stimuli, and material functionality. Between the manufacturing and usage phases (as in the case of origami-based deployable solutions using folding functions) for logistics and transport reasons, or even between two usage stages (as in the case of transformable/adaptable solutions using disparate functions), 4D printing supports multiple transformation scenarios between the two states of the object [16-19]. Such advances are appealing on paper, but they offer only a partial solution and are challenging to put into practice. This is because most studies focus on scientific disciplines that aid in technological innovation, and most studies follow an incremental research pattern that yields only partial solutions.

Problem-solving approaches (such as teleology or inverse problem strategy, as well as user-centered and systems-centered design) [20-23] are necessary for the chemistry, processes, and materials sciences domains to be appropriately aligned with end-users' expectations/needs and design to work on system/product/part levels. To be fair, not much work has been done so far [24-26] to include 4D printing into the design and development stages in order to deliver smart gadgets. Therefore, it is crucial to raise awareness of these developments among product designers and engineers so that they may solve "design for 4D printing," much as assembly, subtractive, and AM difficulties in product design have been successfully integrated in the past [27-30]. Next-generation smart items and devices will be able to meet use over time and, more crucially, will fulfill industrial needs/demands and market (i.e., productions for Industry 4.0) if 4D printing is considered at such expert and operational levels. Additionally, bringing interdisciplinary knowledge over scales of x-changing mechanisms (here x refers for property,

functionality, state, function and shape) in design and engineering would open the solutions space with SMs embedding features like sensor, actuator, transducer, etc.

Based on these core concerns, the paper can be structured as follows to bring attention to the current state of 4D printing and the inherent barriers to be addressed in terms of product/part design. The basics of 4D printing, including transformation functions, smart materials, processes, stimuli, and AM processes/techniques, are introduced in Section 2. Then Section 3 reports key research works in homogeneous (single material) 4D printing as well as heterogeneous (multi-material) 4D printing. Mechanical performances and 4D printing process limits are discussed in Section 4, along with intra- and inter-domain obstacles. Section 5 expands on this foundation by outlining the research agenda and grand problems in engineering design necessary to bring 4D printing innovations to the industrial scale via a (risky) problem-solving strategy. Indeed, existing limitations must be overcome if a desirable level of integration is to be achieved in Industry 4.0 innovations. Proofs-of-concept with improved capabilities that take into account industry requirements can be developed in response to such a research program, and designers will have greater room to experiment with new approaches.

2.Transformative functions, intelligent materials, stimuli, and amplitude modulation: the basics.

To the untrained eye, 4D printing represents a qualitative leap forward for the success of the Industry 4.0 revolution, as it brings together for the same purpose a number of different areas of expertise and, more importantly, presents a spatiotemporal dimension. This advanced techno-science and the associated spatial and temporal phenomena will be introduced in the following sections.

2.1. Functions of transformation

It is crucial to highlight the most important aspects of 4D printing. Basic transformation functions like bending, folding, twisting, expanding/contracting, and changing properties and states are sought for by the aforementioned x-changing mechanisms [31]. These functions reflect the spectrum of changes over time and space at a conceptual level to truly describe a change of shape, property, state or functionality and can also be derived to indicate complicated transformations. This is especially true for a wide variety of wave functions, including helixing, buckling, curling, and waving [32,33]. Figure 1 depicts the six fundamental alterations that can be influenced by the input of energy (stimulus) and the incorporation of both intelligent and inert materials [25,26]. For the purpose of describing and specifying smart devices, such functions and their combinations constitute a first conceptual step upon which further needs might be defined. The ideas and theories underlying transformation and reconfigurable systems [16,17,34,35] provide the theoretical underpinnings for these functions and fundamental transformations. This includes not only the required forces and torques or energy intensity over time, but also the transformation of behaviors between the initial and final states of the objects.

2.2. Stimulus-responsive materials and their functions

In recent decades, material scientists have made great strides in developing materials that function and look like those found in nature. Researchers from several fields have found much motivation in Earth's ecology (for instance, see [36]). According to the paper's argument, materials should be designed to mimic nature's innate abilities for change, transformation, self-organization, and repair [12]. Therefore, available and tailored energy has inspired the development of so-called smart/active materials or even "artificial muscles. To rephrase, SMs are stimulus-responsive materials that can be thought of here as solution/design principles to meet needs in property, functionality, condition, function, and shape changes, as expressed in the preceding section. It is possible to incorporate SMs and the proper stimulus into a design or development by first thinking about the object's context. The latter applies to either user input or autonomous sensing, or even both. All forms of electromagnetic, acoustic, thermal, chemical, mechanical, and optical energy are included.

Just like any mechatronic or mechanical system, it serves both structural and technological purposes.

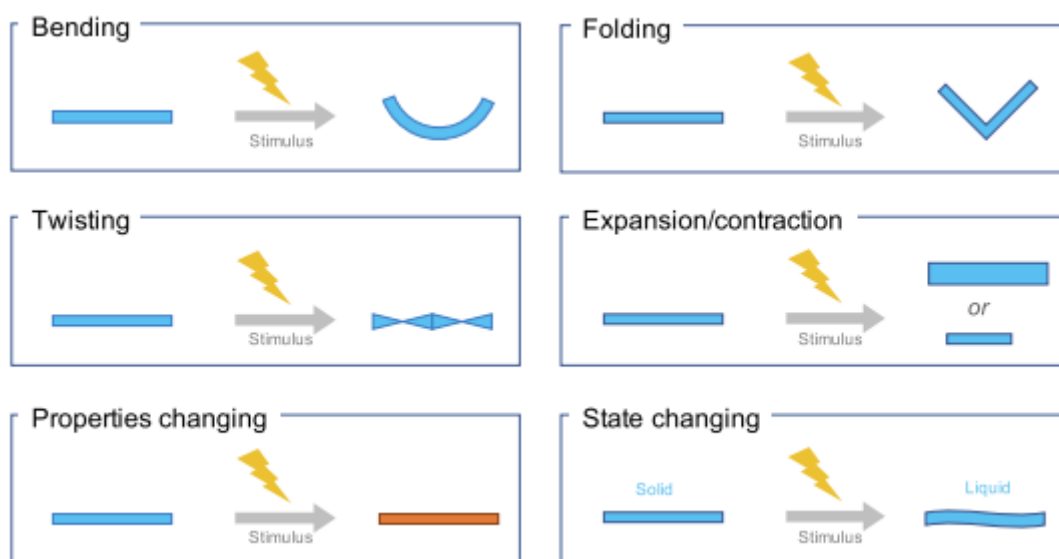


Fig. 1. Using 4D printing, it is possible to perform a variety of transformations [31] (reprinted with the kind permission of Elsevier).

Incorporating a wide range of active characteristics, stimulus-responsive materials serve an important purpose in the final product. Technical function (sensor, transducer, or actuator), stimuli, and the matter (metal, alloy, composite, polymer, or ceramic, in particular) are hence the pillars upon which SMs are founded. Fig. 2 - modified from Dimassi et al.'s classification scheme. by using functional language for design and engineering design [37,38] - displays the functional classification of the SMs (in grey boxes) and provides a peek of prospective design concepts to be understood [31]. Supporting transformation facilitators like furcation, function sharing, and shelling [16,17,34,35] complement a broader class of transformation principles

including expand-collapse, fuse-divide, and expose-cover. It gives a more complete classification than the current one offered by Grassi et. al. [39], with the exception of adhesion altering functions, which are more relevant to the microscale multi-material assembly.

Based on this functional description, which emphasizes the predicted effects and stimulation, SMs can be characterized and studied in terms of materials composition and distribution; if this is the case, the deposition mode or strategy through AM processes/techniques should also be taken into account.

2.3. Methods of additive production

Additive manufacturing (AM) is a rapidly evolving scientific and industrial field that supplements more static approaches like subtractive and formative manufacturing [10,40,41]. Whether working at the nano-, micro-, meso-, or macro-scale, the seven processes and underlying techniques can handle the deposition of ceramic, metal, alloy, composite (short fiber thermoplastics, continuous fiber thermoplastics, thermoset, etc.). and polymer layers. There aren't many tried-and-true methods for 3D printing that have been modified to work with stimuli-responsive materials. Material jetting, powder bed fusion, vat polymerization, and material extrusion are only few of the methods used to create these products. 3. Powder bed fusion and binder jetting are two further methods that have gotten limited attention in conjunction with shape memory alloys, but both have physical constraints when it comes to constructing multi-material parts [48, 50].

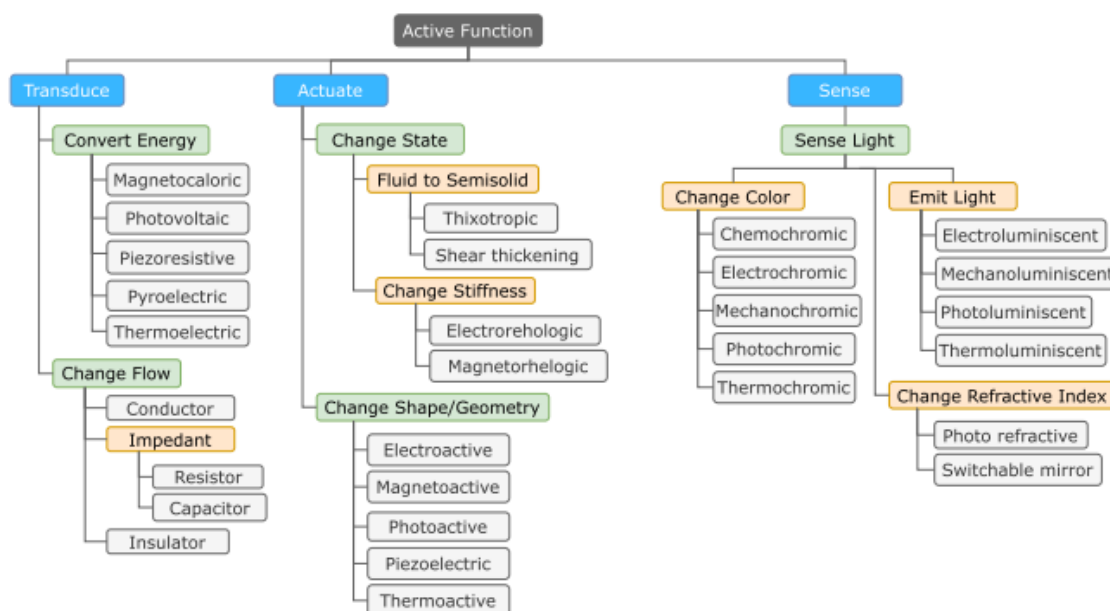


Fig. 2. Functional classification of SMs.

Fig. 2. Separating SMs by their functions.

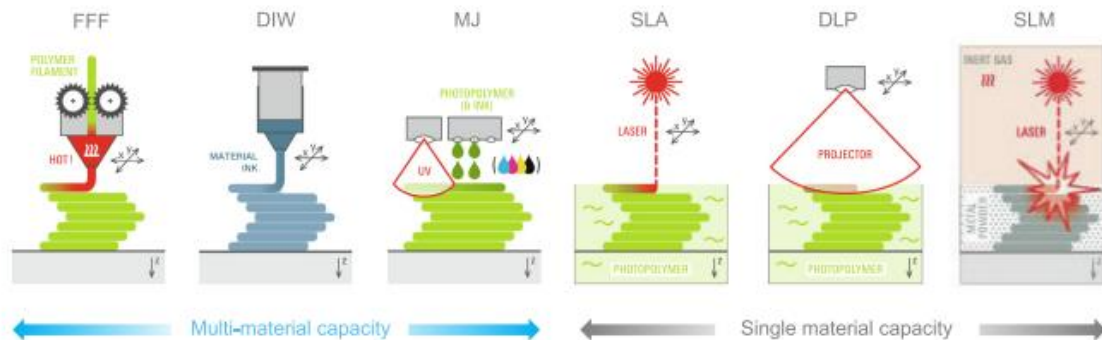


Fig. 3. Main AM techniques currently adopted by researchers to 4D printing.

Fig. 3. Researchers primarily use AM techniques for 4D printing at the moment.

Such devotion to this set of AM techniques is mainly due to the related machines' capabilities, which can (i) meet the states of the most available SMs (ink, resin or filament), (ii) be flexible to different material deposition and polymerization requirements (e.g. extrusion speed, nozzle temperature range, etc.) and more importantly (iii) tackle multi-material printing issue with fewer support structures constraints than others. Digital light processing (DLP), projection micro-stereolithography (PISL), stereolithography (SLA), and selective laser melting (SLM) have all been modified to construct multi-material parts [50,55-58], but these processes are inherently multi-material due to the use of multiple heads and reservoirs in the fabrication of fused filaments (FFF), direct ink writing (DIW), and material jetting (MJ) [51-54]. Researchers are also paving the way for hybrid solutions through the use of multi-technique, customized devices for targeted distribution and composition of materials [59,60].

3. Methods for producing four-dimensional objects

In order to accomplish the necessary transformation, the aforementioned section introduced the primary enablers of 4D printing. One of the best methods of production for tackling biomimicry is 4D printing, which adds behavioral capabilities to conventional 3D items. Researchers and designers have paid a lot of attention to nature throughout the years, using its forms and structures as inspiration for anything from architecture and art to recently, industrial items [36,61-64]. The latter seems to be legitimated with the breakthroughs of AM techniques and their ongoing use by the industry. However, another omnipresent driver, behavior [65-67], should be coupled with the AM process in order to construct biomimetic objects guaranteeing the translation of creative mechanisms to solve engineering challenges. That's why Fig. The spatial-temporal and complexity concerns [68] are illustrated in Figure 4 via 4D printing as a means to implement the biomimicry fundamentals (structure, shape, and behavior).

In the parts that follow, we'll take a closer look at the studies conducted with this goal in mind, paying special attention to the advancement of SMs, homogenous 4D printing, and multi-material 4D printing. For more information on the unique state and difficulties of AM technologies, the interested reader is directed to the aforementioned works [69-71].

3.1. To Homogeneous 4D Printing, From SMs

However, it's worth noting that ground breaking work has gone into creating shape-shifting structures with shape memory materials (SMMs) and artificial muscles with electro-active polymers.

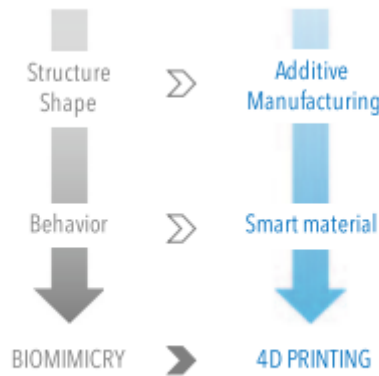


Fig. 4. 4D printing as a biomimicry-based technology.

Fig. 4. Biomimetic approaches to 4D printing.

electroactive polymers) [72] before the development of AM technology. The ultimate goal was to create actuators and sensors that could be used in a wide variety of contexts, including but not limited to organic reactions, digital composites, tissue engineering, biomimicry, and the medical field [73–75].

Among the stimulus-responsive materials, SMMs are those that can be thermo-mechanically trained to change from one permanent shape to one or many temporary shape(s) from which the original permanent shape may be regained. This was accomplished through the invention of shape memory polymers (SMPs) and shape memory alloys (SMAs). Such one-way capability for SMAs is due to the phase transformation between austenite (a phase that is stable at high temperature) and martensite (a phase that is stable at a lower temperature). For SMAs, the transformation from martensite to austenite and subsequent shape restitution requires heating to a specific temperature. A more complex training procedure or magnetic field stimulation can also achieve the reversible (two-way) effect, but both methods have their drawbacks when it comes to the repeatability required in practical applications [76–78]. For SMPs, the thermomechanical training operates differently from how the heat-induced phases change within the material guarantees the shape-changing function [79]. To deform a material into a certain shape, it must be heated above its glass transition temperature (T_g). The deformation strain is then held constant as the material is cooled. The recovery process of the permanent shape is then done by heating the material at T_g . Some important studies exist to broaden the stimulus range toward light-induced heating, light irradiation, infrared light, magnetic field, etc. [80–86] and the actuation effect toward two-way, three-way, and even

repetitive effects [87–91]. Tg, stress magnitude, and elasto-visco-plastic deformation were the focus of many investigations [83,92-94].

The first electric-coupled materials to be produced were piezoelectric ceramics. These materials can generate significant forces, but they typically only deform by a few percentage, which restricts their usefulness to a narrow range of nano- and macro-scale applications. Due to their ability to undergo up to a 380% strain when subjected to an electric field, EAPs have garnered significant attention as a potential solution to this problem [95]. Due to their unique features, which allow for substantial deformation while maintaining significant forces [75,86,89,96,97], they have sparked a widespread proliferation of reversible artificial muscles [98,99]. Table 1 provides a summary of the activation mechanisms for both electronic (activated by electric field or Maxwell stresses) EAPs and ionic (activated by mobility or diffusion of ions) EAPs.

EAPs appear to outperform SMAs in the literature in terms of reaction time, density (lower), and robustness. However, it is vital to highlight their shortcomings, which may be limiting in real applications [95]: low actuation force, mechanical energy density, and resilience. When compared to their electrostrictive counterparts, magnetic materials are appropriate for reversible effect due to their elongation along the direction of the applied magnetic field [123,124].

When triggered by a solvent, light, magnetic, or electric field, these successful advancements have been expanded to include bending, twisting, and folding [125–128]. In the context of self-healing materials [128-134], others have discussed shape memory composites and photo or pH-responsive hydrogels exhibiting self-folding characteristics beneficial for origami-based applications. Tuning constitutive behaviors, material compositions, and their couplings with the various accessible energy fields has been a common approach taken by researchers attempting to embed programmed behavior into materials [137].

Table 1
EAP materials.

EAP type	Material	Description	Author
Electronic	Electrostrictive polymers and dielectric elastomers	The actuation is caused by electrostatic forces between two electrodes that press the polymer. Dielectric elastomers are capable of withstanding very high stresses and are basically a capacitor that changes capacitance when a voltage is applied allowing the polymer to compress in thickness and expand on the surface due to the electric field.	[97,101,102]
Electronic	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.	[103-105]
Electronic	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 load and causes the assembly to rotate. This rotation causes electro-strictive stress and deformation of the polymer.	[106,107]
Electronic	Liquid crystal elastomers (LCE)	The synthesis of highly oriented elastomers leads to high stress thermal actuation in the direction of the polymer chain, with temperature variation resulting in unique mechanical properties and potential applications as mechanical actuators.	[108-111]
Ionic	Ionic/conductive polymers	Activation is caused by the displacement of ions inside the polymer.	[112]
Ionic	Electro-rheological fluids	Electrorheological 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 a low dielectric constant.	[113]
Ionic	Ionic polymer-metal composites (IMPC)	Ionic polymer-metal composites act by electrostatic attraction between the cationic counter-ions and the cathode of the applied electric field.	[114-117]
Ionic	Polymer gels	These are swellable polymer networks with a phase transition behavior in volume. These materials reversibly change their volume, optical, mechanical and other properties through small changes in certain physical (e.g. electric field, light, temperature) or chemical (concentrations) stimuli.	[118-122]

It was in 2013 that the 4D printing paradigm, and the MJ technique in particular, were used to launch an effort to study the field-coupled material behaviors associated with AM processes and techniques. Since then, there has been a meteoric rise in 4D printing research [138]. By addressing SMs in the broader context of AM, integrating behavior in issues seems to be within product designers' and engineers' reach. Homogeneous 4D printing, or 3D printing active parts out of a single material, was the first area of study [139]. This could be accomplished in two ways: (i) by programming behavior in classical polymer by tuning the AM technique and related process parameters, or (ii) by programming behavior through a single smart material. Dufaud et al. [140] introduced piezoelectric suspensions through a modified stereolithography technique that strikes a balance between rheological and photochemical characteristics in order to mold piezoelectric ceramics, although they did not refer to their work as 4D printing. Successful proposals for 4D printing have been made by Van Manen et al. [141], Goo et al. [142], and Wang et al. [143] using hobbyist 3D printers to democratize one-way actuation mechanisms of 1D/2D structures by changing the deposition orientation of polylactic acid (PLA) material with FFF technique. By taking advantage of the constraints applied during the extrusion process and the various combinations of directional strains upon exposure to temperatures exceeding the T_g (see Fig. 5a,b), the same authors have designed specific deposition-patterns to achieve self-bending/twisting and self-folding origami. Similarly, Kuang et al. [144] have modified the DLP approach to process functionally graded materials by employing grayscale light patterns and a two-stage curing ink (see Fig. 5c). This study serves as a reminder of the extensive research done on extremely stretchy and UV curable elastomers for DLP, which led to the development of initial partial solutions for use in soft robotics [145].

Mechanical metamaterials, also known as metamaterial mechanisms, are 3D-printed components that undergo deformations when subjected to mechanical strain or heat [146,147]. Soft and hard regions, as well as cells embedding bistable springs, which propagate mechanical signal in the consolidated part and lead to unobvious and non-linear behaviors [153] (see Fig. 5d), characterize the proposed cell structures, which are composed of periodically distributed patterns printed with SLA, DLP, MJ, or FFF techniques.

From a new angle, a variety of stimulus-material-process couplings have emerged to tackle active functions from 1D, 2D, and 3D structures (see Section 2.2). Water-responsive composite hydrogel structures encoded with localized and anisotropic swelling behavior have been printed by Gladman et al. [66]. These efforts are summarized in Ahmed et al. By directing the alignment of cellulose fibrils along DIW channels, they were able to achieve shape-shifting between two- and three-dimensional configurations. In a similar vein, Mulakkal et al. [155] have developed reversible water-sensitive SMMs using the DIW approach in conjunction with responsive cellulose fibers-hydrogel composite ink. The created material is the basis for the programmed behavior, rather than the printing routes. To further combine thixotropic and thermal-reversibility features, 3D-printed gel of laponite in agarose matrix has been developed. Next, a training (cooling/heating) step is performed, which is analogous to the reversible SMPs programming procedure [156].

EAPs have also been attempted from an AM perspective, this concerns more particularly liquid crystal elastomers (LCE) including light-absorption particles with photo-thermal effect bending reversible behavior [111], which can be appropriately used as strips on flexible parts [59,157] or to address shape memory reversible behavior [158]. To avoid the delamination that can occur in conventional bilayer structures during prolonged exposure to humidity, others have created single-layered humidity-driven actuators. Polymerization can be used to permanently stabilize the alignment of liquid crystals via cross-linking the liquid crystal polymers [159]. In the same way, azo-benzene embedded LCEs have been widely developed in the context of photo-responsive soft actuators [160]. These materials typically expand in a direction perpendicular to the liquid crystals, making it easy to exert control via predetermined print paths.

Furthermore, 4D printing SMPs is likely the arena where a wide range of transformation functions have been addressed, with special focus on the order of modifications [161]. For instance, Lee et al. [162] propose a sequential folding transformation by manipulating the polymer sheet's light absorption. They proposed various crease patterns that may be used to regulate the folding process based on their sensitivity. Such a desire for dominating the Hu et al. [163], van Manen et al. [164], and others have discussed the importance of planning for transition. Indeed, local stimulation is an interest-ing approach for programming the folding sequence [165,166], but tuning material response to a time-dependent global stimulus by locally controlling the material properties opens more alternatives [56,167], which require to address multi-material 4D printing issues.

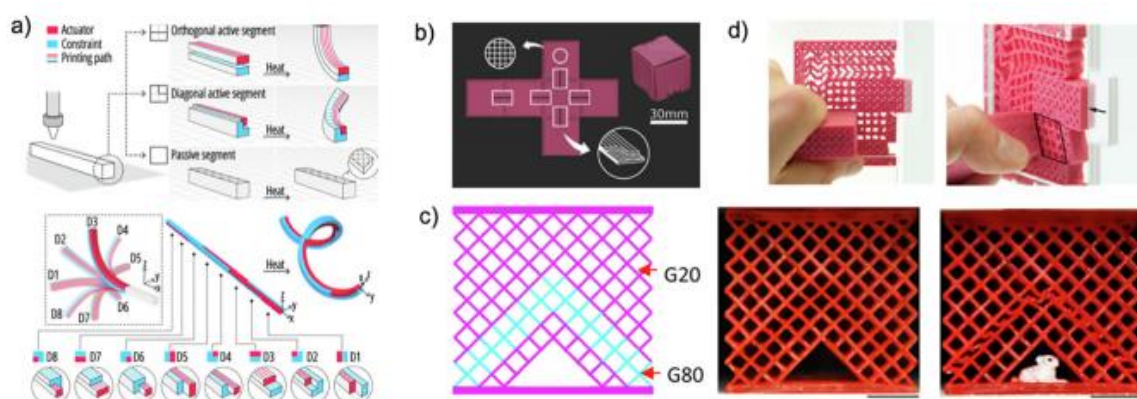


Fig. 5. Homogeneous 4D printing through tuned AM techniques, inert materials, cell structures and deposition pathways (adapted from [141] with permissions from the RSC, from [143] with permissions from the authors, from [144] with permissions from AAAS, and from [146] with permissions from ACM).



Fig. 6. Main categories of materials distributions (adapted from [31,33], with permissions from Elsevier).

3.2. Multi-material 4D printing and metamaterials

3D printing components consisting of spatially dispersed/distributed materials is an example of multi-material (or heterogeneous) 4D printing, which can be used to achieve the desired behavior. In this case, the distribution, which is influenced by the AM technique and related machines' capabilities [168], is of utmost importance and provides significant freedom in the design and engineering of active structures, where efforts in the development of multi-layers (sandwich and bilayer) structures, functionally graded materials (FGM), metamaterials, or even structures with computed materials distributions have been led so far (see Fig. 6).

For example, artificial muscles and origami-based applications can both benefit from the multilayers category, which describes an alternative deposition of layered materials. In order to introduce SMPs with inert materials or to train elastomeric materials with residual shape memory properties to operate bending and folding functions with a one-way effect when stimulated by heat [54,169,170] and/or color-dependent selective light absorption [171], researchers have tried out multi-material FFF and MJ-based techniques. Baker et al. [172] have designed a sequential actuation combining ionoprinting and redox chemistry of 2D hydrogel membranes into 3D folded architecture. Raviv et al. [173] have also shown that multi-material printing may be used to create a linear stretching structure (from 1D to 1D), a 2D folding and stretching structure, and a 3D topographical surface modification. Ding et al. [174] have also looked into using several materials in a cross section to regulate bending in wired constructions. Poly Jet printing allows elastomer and glassy polymer to be printed in a variety of orientations, allowing for the subsequent training of thermo-mechanical shape memory effects, as demonstrated before in [54].

In addition, with the advent of digital materials [175,176], FGMs have received a lot of attention in order to produce parts with adapted mechanical performances in the right place or to program active behavior (for instance, by using adapted DLP technique [144], customized PLA-PEG filaments with FFF technique [177], etc.). FGMs are composites with gradual property variation over one or multiple directions.

An example of a special pattern is the work being done on 4D printed gels that can dramatically and reversibly alter their volume in response to external changes, beginning with water and thermo-active hydrogels [119] and their AM implementation by means of the DIW technique and alginate/PNIPAAm ICE gel inks to construct an active valve [178]. Periodic microstructures of material and void, or even multi-material and void [179], as seen in studies of active metamaterials, promote many desirable properties, including low weight, high stiffness and strength, a negative Poisson ratio, and energy absorption [180]. Metamaterials, in a nutshell, are special structures at the nano and micro scales that may manipulate wave propagation and mechanical properties through the integration of a number of transformations. Composite hydrogels or anisotropic hygroscopic composite materials activated in water have been the focus of early attempts in heterogeneous active metamaterials to cover reversible contraction/expansion (i.e. swelling/deswelling) functions. In order to create consecutive self-folding structures, Mao et al. [181] have used the MJ Poly Jet approach with hydrogels. The

lattice structure undergoes reversible shape-shifting when submerged in water (as seen in Fig. 7a,b), thanks to a bilayer structure made of active hydrogel and inactive polymer created by Zhang et al.[182]. Thermo-responsive metamaterials have also been addressed using DIW method and glass fiber-reinforced poly(dimethylsiloxane) multi-materials to produce shape-shifting lattice structures [183], as demonstrated in Fig. 7c,d. To the same active metamaterials, while Roach et al. [184] have employed DIW with shape memory composites.

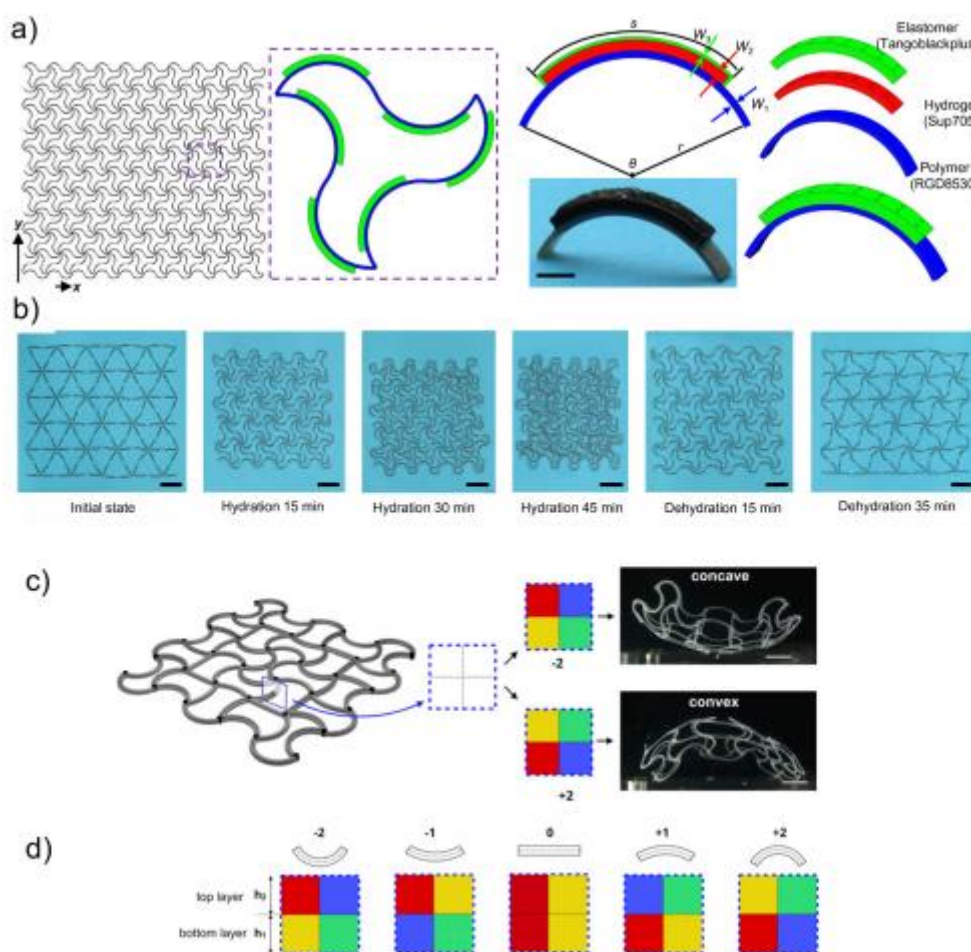


Fig. 7. Example of soft active metamaterial with large negative swelling ratios and tunable stress-strain curves highlighting (a) multi-material patterns build from MJ (Polyjet[®]) technique and (b) water-based actuation [182] (Reproduced with permissions from AAAS), and (c) DIW printed multi-material section structures to control (d) bending orientation in 2D-to-3D transformation lattice-based applications [183] (Reproduced with permissions from PNAS).

Active LCE has been paired with thermally inert materials to generate metamaterials that can change shape [185]. A more recent proposal for printing multi-material lattice structures with controlled buckling and crack propagation pathways is a DIW method based on a dynamic photo mask, as described by Chen et al.[186]. Researchers have developed magnetoactive metamaterials, which can quickly and reversibly change shape by adding hard-magnetic particles in a soft matrix material, similar to the work done on water- and temperature-responsive structures. In this regard, the responsiveness of active materials has been extended toward multi-responsive (both magnetic and thermal stimuli) magnetic SMPs by Ze et al. [187] and subsequently by Ma et. al.

Researchers have recently addressed multi-material distribution computation by employing a voxel-based modeling approach [25,26,190] (see Fig. 8) made possible by the development of connectionist AI techniques and building off of earlier efforts in material properties exploration in solid part [189]. By considering both the intended form and an initial one - whether in 2D or 3D state -, their generative algorithm is able to find counter-intuitive distributions in solid sections that go beyond the well-known patterns exhibited in Fig. 6. However, AM processes and machines that are compatible with digital materials are required for such a distribution category.

4. Intra- and inter-domain barriers

This review does not aim to replace in-depth analyses that have been published over the past eight years on various aspects of 4D printing. As a rule, include methods for 4D printing (191-197), smart materials (198-204), active structures inspired by origami (205), kirigami structures (206, 207), and 4D printing applications (85, 99, 208-215). It is a formidable design and engineering challenge to create artificial versions of natural phenomena [216]. The literature study is organized in a way that makes it easy to see the development, current state, and breadth of the numerous research initiatives, all with the goal of elucidating the obstacles that must be surmounted before the design and engineering process can even begin to take place. One would expect that there would be domain-specific (intra) barriers and limitations related to domain interface (inter) issues, given the nature of the scientific journals studied and the main pillars of 4D printing (i.e., transformation functions, SMs, AM processes/techniques, stimuli, and geometry). The quality and performance of the developed proofs-of-concept in the literature should be analyzed first, and any gaps between the literature and industrial applications should be identified.

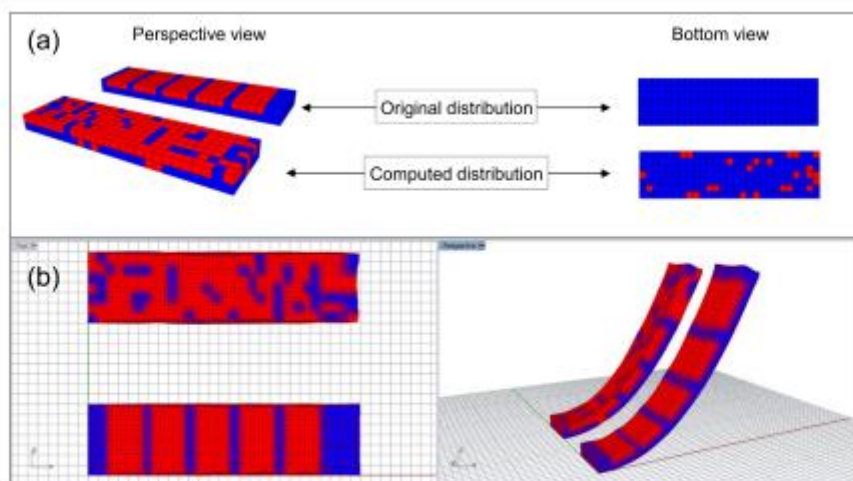


Fig. 8. Voxel-based computation of smart and inert materials distribution and simulation with VoxSmart, a developed Rhinoceros Grasshopper plugin [25,26]: (a) comparison of original and computed distribution and (b) their simulation in conceptual design using Bernoulli-Euler beams.

4.1. Material performances

Smart (typically soft) actuators with sensing and shape-changing capabilities are the primary focus of 4D printing research; these actuators must be monitored via key performance indicators (KPIs) and mechanical properties. Some attributes, such as actuation stress, actuation strain, actuation modulus, work output, work capacity, actuator efficiency, stroke work coefficient, etc., may have special attention from designers and engineers, in accordance with the conventional actuators classification proposed by Zupan et al. [217], in order to define the industrial specifications. Attributes like Young's modulus, response time under stimulation, the force generated, and actuation efficiency (i.e. the ratio of mechanical work output to energy input during a complete cycle) can be derived and expressed in terms of material properties [218] to measure and monitor the performances of 4D printed actuators with SMs. That's why Fig.

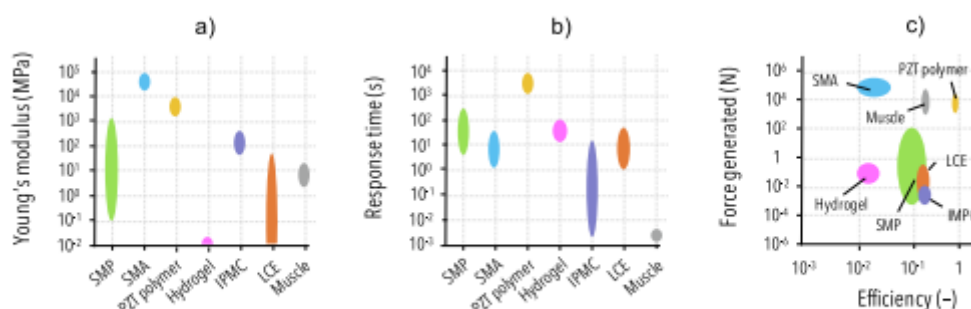


Fig. 9. Main performances of the main SM categories used in 4D printing : (a) Young's moduli, (b) response times once stimulated, (c) forces generated and actuation efficiencies.

Table 2
4D printing process limitations for an engineering and industrial adoption.

4D printing facet	Limitations	Origin
SMs elaboration	Availability in terms of quantity and type (powder, resin, filament, fiber and pellet). Need a tuning step. Degradation of change capacity over multiple cycles (if doable) Induce multi-scale behaviors.	Inter-domain
AM processes and techniques	Material/process couplings limited. Type of raw material (resin, pellet, filament, fiber and powder)/process couplings. Few process/technique exhibits multi-material capacity. Lack of SMs compatibles with AM technologies. Lack of process-compatible SMs. Anisotropy induced by the layer-by-layer deposition mode.	Intra-domain
Stimulation	Most techniques require a post-processing step. Temporal application of the stimulus and its propagation in the material (e.g. conduction/diffusion) within the structure. The stimulus needs to be maintained during the stimulation. External and global application of the stimulus to the whole structure. Volume of the stimulation device. Energy consumption of the stimulation leading to weak actuator efficiency. High amplitude of the signal required.	Inter-domain

Numerous studies [219–224] have centered on SM characterization to provide useful information for material selection. There can be huge discrepancies in the calculated/measured indicators, making it tough for laypeople to use them. This is due, in large part, to the fact that, over the past two decades, many advances have been made in areas such as material customisation and distribution, actuator size, and form-factor diversity. Current PKIs are still low compared to the performance of actuators (such as hydraulic, pneumatic, etc.) utilized in

the industry (Fig. 9). Mechanical stiffness, actuation forces, actuation/recovery responses (e.g., thermo-active materials with important heating/cooling times due to thermal conduction Fourier's law), change capacity, and even material stability over multiple cycles are all areas in which most 3D printed SMs fall short.

4.2. Process limitations

Fabrication, training, stimulation, actuation, and recuperation are just few of the many processes involved in creating smart things through homogeneous or heterogeneous 4D printing. These steps can be performed in any order or nested within one another. Indeed, the training step can be operated either after [169] or during [141,225] the material deposition phase, making it a pre-stimulation and actuation process. Some restrictions can be identified at the level of processes/techniques and stimulation phases that integrate materials and physics concerns, but these have a direct bearing on the performances of the final printed items through the actuation and recovery steps. Printing single SMs has been the focus of study, however due to their limited transformation functions and mechanical performances, this work has mostly been extended to multi-material active structures. The current limits of the process are presented in Table 2 and need to be taken into account in the context of this complicated transformation process. Advanced 4D printing techniques based on heterogeneous principles make it possible to print in several materials and voxels with just one or a few 3D printers, which could eventually lead to the kind of performance levels required for industrial applications. Associating requestor and designer in a more top-down sophisticated logic of request will necessitate breaking out of a bottom-up logic offer. This is unquestionably the "price to paid" if top-tier manufacturing output is the goal.

From this set of performances and technical limitations as well as the maturity of the technology (which can be estimated at TRL 1-3 scales), it becomes obvious that integrating such an interdisciplinary research issue in engineering design is a long-term challenge and is also necessary to reach application sectors with concrete solutions. Since there is a gap between the substantial progress made in designing SMs and the transition to designing using SMs and AM technologies [226], the following part will discuss research measures to fill that gap in knowledge, models, methods, and tools.

5. Opportunities and challenges in engineering and interdisciplinary design: A strategic research roadmap

5.1. Main scientific and industrial stakes

Both the breadth and depth of current research into 4D printing show that (i) the field is thriving thanks to a wealth of proofs-of-concept and interest in making active structures with AM technologies, and (ii) the field has interdisciplinary aspects that cannot be addressed in silos. Over the past few decades, researchers and designers have confronted interdisciplinary challenges in product development related to the various technologies embedded in goods and

objects. In fact, the manufacturing sector has been under increasing pressure to boost product/system capabilities and shorten lifecycles by guaranteeing a certain level of flexibility and efficiency as competitive edges, particularly in the product development phase where design activities and decisions have a significant impact on downstream processes (i.e., manufacturing, assembly, maintenance, etc.) [227]. This has been accomplished by taking into account (i) the integration of other disciplines such as electronics in mechatronic system design; (ii) the integration of computing capability in the age of the Internet of Things (IoT); and (iii) the capture and integration of lifecycle constraints and knowledge in product design (e.g. design for X - DFX spectrum approaches, advanced Computer-Aided Design - CAD) in mechanical product design to deliver lifecycle friendly products. Ad hoc (1), repeatable (2), defined (3), managed (4), and optimizing (5) are the first five levels.

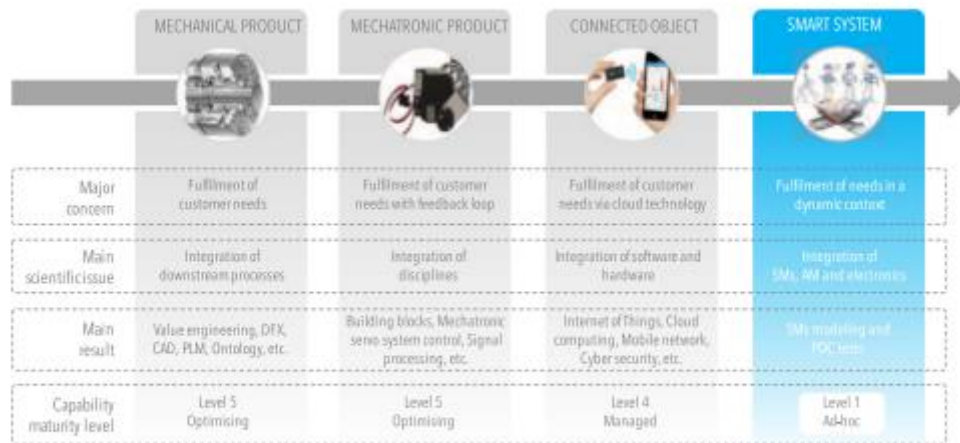


Fig. 10. Evolution of product technologies and related scientific challenges, results and maturity levels.



Fig. 11. Key ingredients to be considered for building the design for 4D printing roadmap.

Research Association [229], Manufuture [230], NASA [226], and ESA [194] to name a few - entails increasing the lifetime of systems/products by providing appropriate usage

configuration in a specific use context and environment (see the blue box in Fig. 10) [20-2] and making them more responsive and smart after they have been manufactured in light of user needs or customer expectations. Research projects like this are part of the larger context of "industry 4.0," which includes AM, soft robotics, and 4D printing. As smart products and systems (i.e. products and systems that can adapt to their environment, self-assemble, repair themselves, sense their surroundings and react accordingly in a programmable manner, etc.) have emerged over the last decade, it has become necessary to significantly revise current well-established design and engineering models and approaches in order to deliver dynamic product definitions (i.e., product geometry embedding functional m) using AM technologies. 4D printing needs to be studied from a design and development standpoint in order to cross the operational and industrial threshold, where the inverse problem is mostly accepted by product architects and mechanical designers in their daily activities. Driving this manufacturing paradigm shift in the product development process, with an emphasis on a multi-material printing strategy, presents significant challenges and opportunities. In the following sections, we will attempt to outline the primary lines of inquiry that must be pursued if a "design for 4D printing" strategy is to be effectively applied to the creation of intelligent systems.

5.2. Design for 4D printing roadmap

Multi-X is a roadmap that aims to map all aspects of 4D printing in order to provide products-systems that are meaningful, useful, and sustainable. In order to achieve this far-reaching goal, it is important to take into account the many different types of materials, domains, processes, techniques, plans, functions, physics, worlds, representations, sectors, niches, scales, and times that will need to be factored into the creation of systems, products, and objects. This can be accomplished with the help of AI techniques like connectionist and symbolic reasoning, biomimetic computing, and parametric/topology optimization. Figs. 11 and 12 summarize these three primary lines of inquiry. It reveals the hidden depths of 4D printing's numerous facets, as well as many potential avenues and obstacles for further study.

5.3. Design framework of stimulus-responsive structures

Considering a general design for 4D printing framework that encourages an inverse problem approach to building innovative solutions is crucial in light of the widespread creation of proofs-of-concept to exhibit stimulus-responsive behaviors in objects and structures. The inherent spatiotemporal behaviors of active materials, as well as the complexity level induced by the multiple involved stakeholders (i.e. product architect, product designer, material expert, AM process planner, mechanical engineer, etc.), necessitate a progressive integration of the 4D printing constraints, whether at the functional, logical/behavioral, structure, or geometric levels, compared to well-adopted design for AM, manufacturing, and assembly methods [That's why Fig. Top-down definition of stimuli-responsive product-systems and concurrent consideration of stimuli, SMs, and AM knowledge are highlighted in the process flow presented across the conceptual, embodiment, and detail design stages in 13. The incorporation of a design innovation process flow allows for a more adaptable method of continuously

focusing on the desirability of all involved stakeholders, as well as the possibility of infusing creative, inspired, and user-centered ideas into the design stages [239].

By evaluating such a broad framework with AM-enabled design theory and methodology [238], new crucial steps have been presented and well-adopted ones now exhibit extra consideration and actions [22,239]. Decision-making aids, heuristics, guidelines, and rules are needed for each step/component of the design framework, which brings its own unique opportunities and challenges. There is enough flex-ibility to incorporate any reasoning procedures [240], and the main steps can be processed proactively to involve SMs and AMs in early design stages critical decisions leading to a multi-perspective development (i.e. structure, geometry, and properties).

5.4. 4D printing knowledge capture and reuse (design and material informatics)

Top-down design methodology is used when the inverse problem method is followed. This expansive field of study necessitates the development of a solid foundation of knowledge concerning stimuli, SMs, products, and manufacturing/stimulation/actuation process models. In order to define 4D printing knowledge at the product/part level, the HERMES domain ontology [31] was developed, taking into account basic formal ontology (BFO) and 3- and 4-dimensionalism theories. The BFO upper ontology can be used to construct a thriving research domain ontology, as shown in Fig. 14 - guarantees its compatibility with any other domain ontology, including those advocated for by the IOF (Industry Ontology Foun-dry) effort [245].

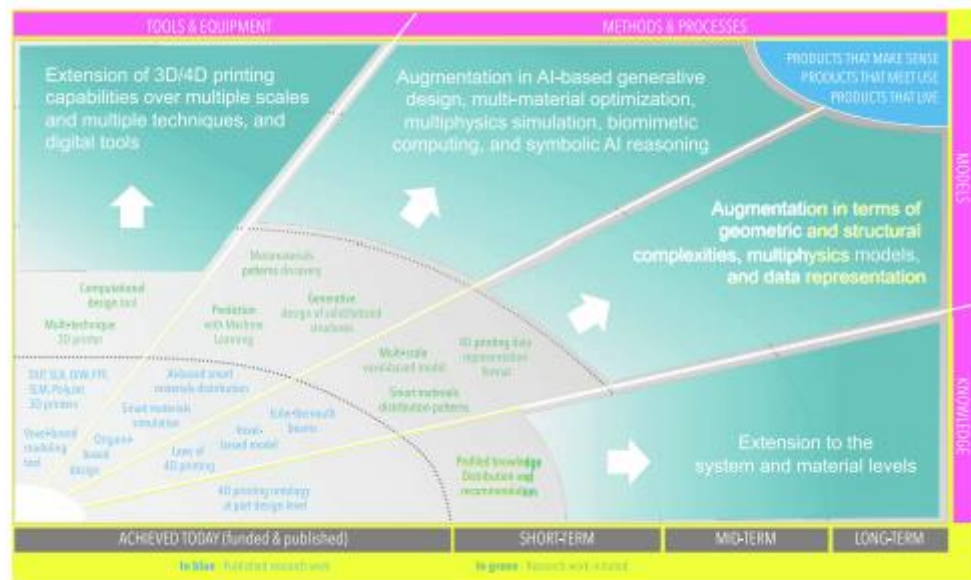


Fig. 12. Research roadmap of design for 4D printing.

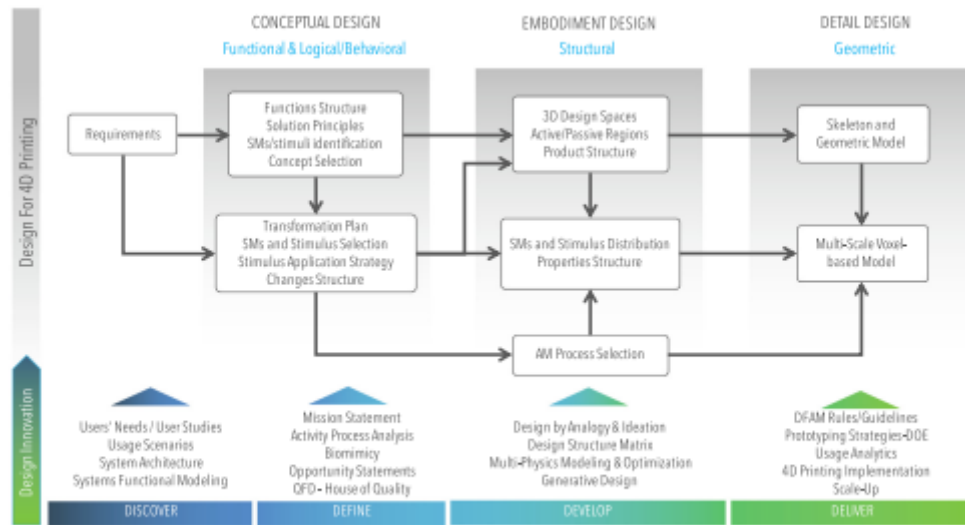


Fig. 13. General design for 4D printing framework.

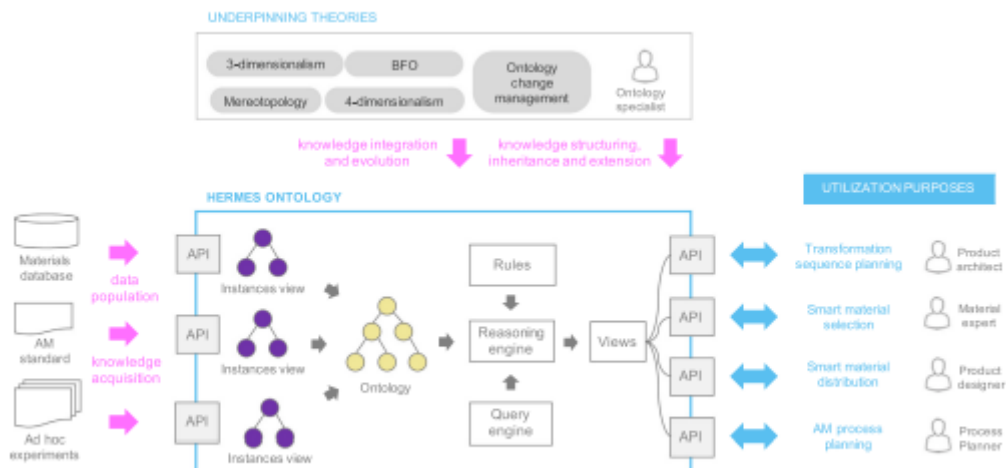


Fig. 14. HERMES ontology strategy to formalize 4D printing knowledge for multiple reuse perspectives [31] (with permissions from Elsevier).

Knowledge capture in this setting needs to take into account not only knowledge reuse and reasoning situations, but also existing literature and standards that are being continuously refined and updated (see Fig. 14). Next research efforts have to solve this issue and extend 4D printing knowledge whether at system and materials levels or even to cover soft robotics [246] and structural electronics while maintaining their semantic alignment. Research in this area would pave the way for the creation of symbolic AI reasoning techniques as a qualitative decision-support tool for use in a variety of selection problems and design suggestions. It can also play a pivotal part in giving context to connectionist AI-based design strategies [247,248].

5.5. Functional modeling and design principles

When designing deployable, reconfigurable, or transformable systems, developers must account for x-changing requirements, which means introducing new transformation functions

or dynamic functions to accommodate a variety of states (from manufacturing to usage phases or just during the usage stage) [16-19,38,249]. It is possible to directly relate such functions to solution principles based on SM/stimulus couplings if sufficient detail is provided. Since SMs have a practical application, the latter can be implemented now. Technical functions, materials, signals, and information must be mapped in order to create a functional model of a system's response to a stimulus. Therefore, the Black Box concept advocated in [4-6,37,38] could be a viable option; nonetheless, there are certain obstacles to this approach, including those listed below.

- streamlining technical processes with things that serve several purposes;
- combining user-centric, user-interface, user-experience, and system tasks with technological and multi-material functionality;
- by using AM techniques for shape-shifting, material-hopping, and function-doubling, 4D printing has advanced design for AM and design innovation [22,233,234].
- developing AM, creative, and technological design concepts based on biological inspiration that pair form/behavior/structure with SM/stimulus distribution patterns [62,249-258]
- setting up tools for design principles recommendation.

In order to develop design concepts from a technology semantic network, as advocated by Sarica et al. [259], it would be difficult to reconcile 4D printing expertise with natural elements. The development of new ideas and concepts may benefit greatly from this course of action.

5.6. Smart materials/stimuli selection and transformation planning

Comparative study of the created solution principles in light of end-user needs and functional specifications must precede the selection of the best design concept to be developed. The idea may originate from a single design principle or from the synthesis of several. However, guidance in the selection of active materials and stimuli is necessary to enable engineers and designers to comprehend and make judgments. To solve this problem, a transformation plan outlining the needed adjustments can be implemented. To do this, one must determine the stimulation mode (autonomous with external environment or regulated with the integration of a stimulator into the structure) and the corresponding stimulation application strategy (global, local, internal, external, or a combination of these). Once the technique for transformation has been established, the desired SMs or stimuli can be processed. Unfortunately, product designers lack a reliable SM selection tool at this crucial stage [260]. This is mainly owing to the complexity of the ad hoc experiments discovered in literature that highlights a wide spectrum of parameters (stimulation intensity, stimulation flow, material composition, size, form and distribution, 3D printing parameters, etc.). Currently, there exists no well-adopted database to compare SMs/stimuli. In particular, the number of times the 4D printing effect may be achieved by the employed SMs, or the recovery % that is attained after a set number of cycles, is not described. The widespread use of SMs in engineering design and design education could be accelerated by developing a standardized process for testing SMs. A first problem consists in defining SMs KPIs similar to those available for measuring performances of actuators and sensors as indicated in Section 4.1. As mentioned in Section 5.4, there is also the possibility of

using AI methods to extract relevant facts and knowledge from existing literature in order to create a 4D printing ontology. The end goal is to pick the right combination of SMs and stimuli to bring about the desired behavioral changes. The latest standards of the American Society for Testing Materials (ASTM) should be used to this line of inquiry.

5.7. Design spaces definition and CAD representations

As shown in Fig. 15, the CAD representation of the solution being created must be handled at various abstraction levels, perspectives, and scales as part of the proactive design for 4D printing process. In Fig. In order to specify the overall geometry of the product/part and the active/inert regions, as well as the properties, materials, and stimulus, the top-down design has highlighted CAD modeling strategies [261,262] for behavior specifications with geometric skeletons (i.e. line, point, plane, sketch, etc.) or 3D design spaces (i.e. functional surfaces, volumes-envelope, etc.). These methods are required to deal with the inverse problem methodology of 4D printing design, and they provide a variety of abstraction levels in which to introduce complex reasoning processes. A primary difficulty at the level of the design space is the allocation of active and inert regions requiring special computational attention, as well as the construction of geometric skeletons to govern the shape-changing behavior of the structure. In order to establish the right qualities in the right place for the multi-material distribution, based on such geometric input, a specific CAD representation is needed, which satisfies both the intended transformation at the digital level and multi-material 4D printing. To facilitate both material design and design using the material, multi-scale voxel-based modeling appears to be highly important [226].

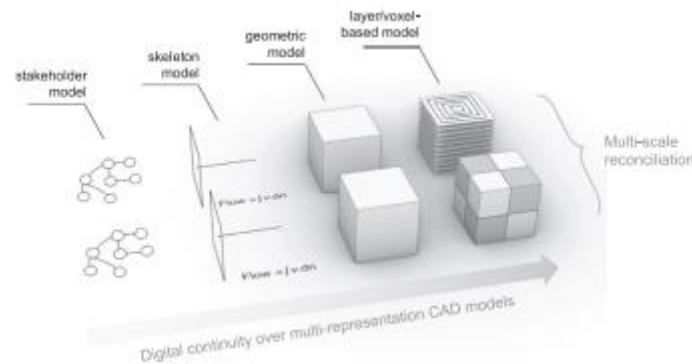


Fig. 15. Top-down construction of the design spaces [240] (with permissions from Elsevier).

Another problem involving embodiment and detail design stages is to align and reconcile the aforementioned set of CAD representations [240]. Suitable CAD representations for knowledge reuse are provided by the gradual rise in abstraction layers and the creation of composite solids and lattice solutions. The intricacy of the resulting 3D models, however, necessitates the use of a standardized data interchange protocol for the purposes of capturing data at various scales and passing it on to the manufacturing phase [262]. Standard tessellation language (STL) [264], wavefront object (OBJ) [265], additive manufacturing file (AMF) [266], and 3D manufacturing format (3MF) [267] are some of the most used data representations for

AM. Academic and industrial communities share an interest in AMF because of the structure of the representation, which makes it ideal for taking into account data from multiple scales and materials.

5.8. Smart material/stimuli distribution vs. Topology optimization

Distributing several materials and their associated qualities in the correct location inside the final geometric specification of the product or part is a challenge posed by the design and realization of the transformation function and its needed performances. Using the well-known distribution patterns shown in Fig. 1, we may implement a primitive behavior in a primitive form. 6. Only by taking into account AM process and planning limits in advance can a digital specification of the multi-material structure be generated. There has been a lot of study done on the topic of choosing and designing AM processes [268,269], but the emergence of multi-material AM processes and techniques (shown in Fig. Deposition capacities (either layer-by-layer or volume-by-volume) bring up compatibility concerns with materials and distribution patterns. This choosing process should be guided by the various methods of stimulation. For instance, if a stimulator needs to be built into the structure to generate online and offline stimulation, the production strategy will need to account for the stimulator's integration either during the AM process or afterward by an assembly operation. Therefore, the design decisions regarding the materials and their distribution in the structure are determined by the AM technique chosen.

Several difficulties in addressing the distribution of several materials and stimuli can be derived from this scenario, particularly if a multi-scale modeling approach is used.

Spatial-temporal phenomena in hybrid materials require not only multi-scale modeling (bottom-up, top-down, and scale-bridging techniques), but also multi-physics simulation. Particle-based and continuum-based models can be coupled by extending scale-specific models with scale-bridging models, which account for the diverse dynamics and temporalities at play in 4D printing's various application scales. Getting closer to this long-term goal will aid in the development of real-time simulation-driven design of SMS/stimuli with quantitative predictive capabilities of their structure and properties [270].

- In order to encourage an inverse approach, we compute the distribution of materials in 3D design spaces (where active zones have been defined) on a voxel/octree basis at various sizes (macro, micro, nano);
- Combining parametric optimization with multi-scale (continuous or discrete) topology optimization in multi-material structures [271,272];
- Use of connectionist AI (e.g., machine learning, deep neural network, etc.) to uncover distribution patterns in materials, metamaterials, and multi-scale systems;

When defining a voxel's structure, it's important to keep in mind bio-inspired generative design methodologies. The development of the multi-scale geometric definition might be guaranteed by pursuing potential research lines in the direction of voxel automata, L-system, digital morphogenesis, etc.

5.9. Digital thread for design and 4D prototyping of smart systems

Design methodology implementation also calls for a separate digital thread [273-275]. Fig. Systems engineering, product design, multi-physics modeling and simulation, process planning, and process engineering are all links in the digital chain depicted in Figure 16 for constructing intelligent systems using 4D printing technology. When paired with a 4D FabLab value chain beginning with raw materials and progressing via combined manufacturing, assembly, 3D printing, training, stimulation, and actuation, this chain has potential. By linking together relevant data systems and technologies, including those for digital and physical models, the digital thread can be established. Incorporating a robust and dynamic knowledge base associated with a digital twin in such a way as to (i) enhance theoretical and simulation models from physical data, (ii) guarantee cyber-control of physical systems, and (iii) enable a seamless reconciliation between the two worlds via feed-back and feed-forward information flows represents a significant scientific challenge.

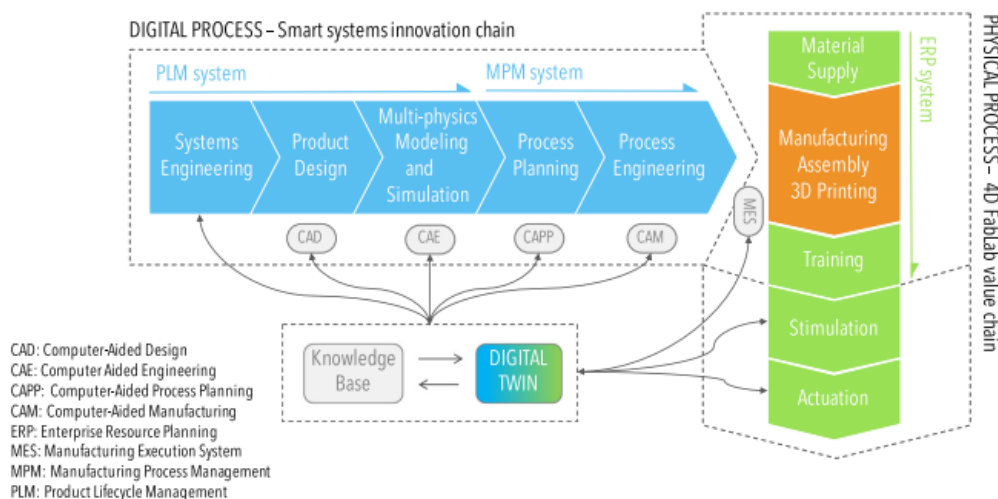


Fig. 16. Envisioned digital thread dedicated to design and prototyping of smart systems.

6. Conclusion

Academe has taken a growing interest in 4D printing during the past decade, initially from the fields of material science, process science, and mechanics. Numerous proof-of-concept demonstrations show that this highly multidisciplinary technology allows for more flexibility in the construction of dynamic structures that respond to applied energy. This observable benefit, which highlights the functional enhancement over time via integrated elements, reveals complexity and paradigm shifts in design. From smart materials to homogeneous and heterogeneous 4D printing, a comprehensive literature review has been done for this paper in an effort to reveal existing restrictions and underlying inter- and intra-domain hurdles. A lengthy research path for engineering design to address "design for 4D printing" is based on this foundation. Everything that can be done to bring this developing technology over the hump into the industrial realm has been done. The current form of 4D printing shows promising capabilities to play a pivotal role in the era of Industry 4.0, but it also brings new usage

scenarios that merit investigation in terms of human aspects and experience for the acceptance of stimulus-responsive structures in society.

The use of 4D printing techniques, technological solutions, and 4D printing applications is moving forward in industry and research societies with the backing of international and national research organizations. BMW's Vision Next 100 [276] is a concept car that has a flexible body and a flexible cockpit that can be rearranged depending on the driving style. Hexonic, a wheel concept invented by Hankook, adapts its size and surface roughness to the conditions it encounters [277]. Calls for research with such possible uses can be found in the scope of European programs, ESA programs, and even in nations that encourage the growth of novel and potentially dangerous scientific issues (such as the United States, Singapore, France, and others). The creation of high-throughput (smart) materials for use in commercial or even hobbyist 3D printers is currently seen as one of the most important areas of research and development.

4D printing needs to go beyond connecting AM machines with SMs and requires ground-breaking innovations to solve multi-material manufacturing. In the context of a CNRS (French National Center for Scientific Research) maturation program, startups like GenIsis are emerging to create hybrid 4D printing solutions (in terms of tools and machines) for use in industrial settings [278].

Declaration of Competing Interest

No authors have disclosed any conflicts of interest that would have been perceived as having influenced the work presented in this paper, either financially or otherwise.

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