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

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Abstract

The combination of scientific advances in additive manufacturing (AM) and smart materials (SMs) has enabled the development of a new interdisciplinary research area: 4D printing. This technology offers – via stimuli-responsive materials – promising transformation capabilities to objects whether at the functional, shape, or property levels. By considering such capabilities, researchers from multiple disciplines have investigated a large spectrum of stimuli-SMs associations with proofs-of-concept built from either commercial or custom 3D printers. Despite the abundant initiatives, 4D printing requires additional developments to meet robust system applications for the industry. The paper aims to highlight the status, inherent barriers, and challenges of 4D printing to be addressed from a productsystems design perspective. It firstly reminds the fundamentals of SMs, processes, stimulus, and AM to which a synthesis of significant research works related to 4D printing highlighting the current status as well as scientific, technical, and organizational limitations is provided. Beyond this comprehensive study, the paper emphasizes opportunities and challenges from multiple perspectives and draws a research roadmap for engineering design and cross-disciplinary design. The outcome of the work tends to structure research efforts for the next decade towards the development of smart products that meet use for humans and the industry.

Keywords: 4D printing; Additive manufacturing; Smart materials; Design; Engineering

1. Introduction

With Industry 4.0, the entire paradigm of industrial strategy is transformed, including its logic of economic performance and the mechanisms of value creation that underlie it. The possibilities and potential benefits of the Industry 4.0 concept are multiple: flexible and customized mass production, real-time coordination and optimization of value chains, reduction of complexity and costs, adaptability and emergence of new services, and high-performance business models [1,2]. Industrial production relies less and less on scale and volume effects, but more and more on flexible and localized production close to demand. It marks a dynamic adaptation to the user's needs. By placing usage and no longer the product at the heart of its logic, engineering and production require to be more predictive. By reducing the cost of the complexity carried by the digital component of a product or machine with a standardized base, additive manufacturing (AM) already fits perfectly into this logic [3]. But, with a complementary opportunity of creating objects 'shape and functionality of the objects via the additive manufacturing (AM) technology, 4D printing has a role to play with the other pillars of Industry 4.0, i.e., soft robotics and the Internet of Things (IoT). The development of the specific axis constituted by 4D printing is thus an important strategic integrating element in the technical progress associated with the Industry 4.0 initiative.

By design, 4D printing generates technological inseparability when it comes to producing a part within the 3D printer, made active by stimuli, rather than by activities organized in different successive steps. It is therefore a major disruptive integration that can revolutionize the manufacturing domain if it can satisfy industrial specifications (otherwise, one would have just to look for niche applications, corresponding to a more modest attractiveness and a less exciting research drive). Evolution in production strategies, materials development and manufacturing techniques has always influenced the way people think and perceive physical products and objects, and mostly design and engineering-related activities [4-7]. This is true with AM since this technology reconsiders traditional subtractive and formative manufacturing-oriented design practices by paving the way for multi-functional and multi-material parts with complex geometry [8-11]. Built upon such a manufacturing disruptive technology that is mainly focused on complex shapes and high-resolution issues, researchers over the last decade have opened the door towards the integration of smart or active materials, i.e. materials that have capabilities of changing shape, functionality and property once stimulated by user input or autonomous sensing in their usage environment [12,13]. Printing SMs or smart voxels from a digital point of view therefore leads related research domains and scientific communities toward new interdisciplinary and converging research paradigms, namely bio-printing and 4D printing [14,15]. The latter is the subject of this paper.

Featuring AM-enabled transformation capabilities, 4D printing has received huge attention from researchers in domains such as chemistry, applied sciences, physics, materials science and mechanics. By combining AM deposition processes, stimulus and material functions, the proof-of-concept oriented contributions in this booming research field are promising as well as multifaceted. 4D printing supports multiple transformation scenarios between two object's states, whether between manufacturing and usage phases (e.g. case of origami-based deployable solutions using folding functions) for logistics and transport reasons, or even between two usage stages (e.g. case of transformable/adaptable solutions using disparate functions) [16-19]. Such developments are extremely attractive from conceptual point of view but only provide a partial solution that is difficult to implement in real application cases. This is due to that most of the research efforts follow an incremental research leading to partial solutions and are mainly addressed by scientific disciplines supporting technological innovations.

To reach an industrial maturity threshold, chemistry, processes and materials sciences domains should be appropriately aligned with end-users expectations/needs and design to work on system/product/part levels, which consists in applying problem-solving approach (i.e., teleology or inverse problem strategy, as well as user-centered and systems centered design) [20-23]. Indeed, few efforts have been made towards the integration of 4D printing in design and development phases to deliver smart devices so far [24-26]. As a consequence, it becomes urgent to bring such advances to the attention of product designers and engineers to tackle "design for 4D printing" as previously successfully achieved to integrate assembly, subtractive and AM issues in product design [27-30]. By considering 4D printing at such expertise and operational levels, next-generation of smart products and devices will be enabled to meet use over time, and more importantly, will fulfil industrial needs/demands and market (i.e. Industry 4.0 productions). Additionally, bringing interdisciplinary knowledge over scales of x-changing mechanisms (here x stands for property, functionality, state, function and shape) in design and engineering would open the solutions space with SMs embedding functionalities like sensor, actuator, transducer, etc.

Built upon these main stakes, the paper aims to highlight the status and the inherent barriers of 4D printing to be addressed from a product/part design perspective, and can be organized as follows. Section 2 presents the fundamentals of 4D printing in terms of transformation functions, smart materials, processes, stimuli and AM processes/techniques. Then Section 3 reports significant research works in homogeneous (single material) 4D printing as well as heterogeneous (multi-material) 4D printing. Section 4 addresses intra and inter-domain barriers through mechanical performances and 4D printing process limitations. Built on this, Section 5 presents a research agenda and grand challenges in engineering design showing an articulation/integration across scientific domains and scales to bring 4D printing advances to the industrial scale by following a (risky) problem-solving approach. Indeed, current constraints must be circumvented if a satisfactory integration objective is to be met in Industry 4.0 developments. Such a research agenda provides key drivers to develop proofs-of-concept with better capabilities respecting industrial requirements and will give more creative space to generate innovative design solutions.

2. The fundamentals of transformation functions, smart materials, stimulus and AM

At first glance, 4D printing can be seen as a sophisticated manufacturing paradigm for scientists, engineers and designers, representing a very important qualitative breakthrough for the success of the Industry 4.0 revolution, since it gathers, for the same goal, multiple expertise areas and more importantly it inherently presents a spatiotemporal facet. The following sub-sections aim at introducing the key ingredients of this high-level techno-science and the related spatiotemporal phenomena.

2.1 Transformation functions

Putting forward the key factors with 4D printing is something of utmost importance. The aforementioned x-changing mechanisms aim at fulfilling basic transformation functions such as bending, folding, twisting, expansion/contraction, property and state changing [31]. These functions represent the spectrum of changes over time and space at a conceptual level to actually describe a change of form, property, state or functionality and can also be derived to specify complex transformations. This is particularly the case with helixing, buckling, curling curving, waving functions to name a few [32,33]. Figure 1 illustrates the six basic transformations to be controlled by energy input (stimulus) and embodied smart and inert materials [25,26]. Such functions and their combinations represent a first conceptual step for the description and specification of smart devices, on which additional requirements can be expressed. These functions and basic transformations are supported by the principles and theories of transformation and reconfigurable systems [34-37]. This encompasses the transformation either one way or two-way (reversible) behaviors between the original and final states of the objects but also the needed forces and torques, or even energy intensity over time.



Fig. 1. Transformation functions enabled by 4D printing [31] (Reproduced with permission from Elsevier)

2.2 Stimulus-responsive materials and their functions

Over the last decades, material science has put forward tremendous efforts to develop materials that mimic nature in terms of behavior and structure. Mother Earth is an ecosystem that has been offering great inspiration for researchers in a wide spectrum of domains (see for example [38]). In the context of the paper, it seems that nature presents a flair for adaptation, transformation, self-organization, and self-healing, which deserves to be engineered in materials [12]. Thus, the so-called smart/active materials or even "artificial muscles" have been developed in parallel with available and adapted energy as trigger. In other words, SMs are stimulus-responsive materials, which can be considered – in the present context – as part of solution/design principles to fulfil needs in property,

functionality, state, function and shape changes, which can be expressed according to the previous section. By considering the surrounding of the object to be designed and developed, it is then possible to embody SMs and appropriate stimulus accordingly. The latter refers to either user input or autonomous sensing, or even both. It includes light, magnetic and/or electric fields, moisture, pH, temperature, solvent and mechanical energy.

Similar to any mechanical or mechatronic systems inherently fulfilling both structural and active technical functions, stimulus-responsive materials encompass a panel of capabilities under the active functions category and henceforth play a functional role in the object, product or system. The nature of SMs is therefore built upon the association of the technical function – sensor, transducer or actuator -, the stimulus and the matter or the material itself (metal, alloy, composite, polymer and ceramic especially). Figure 2 – adapted from the taxonomy proposed by Dimassi et al. [31] – shows the functional classification of the SMs (in grey boxes) by using functional language for design and engineering design [39,40], and provides a glimpse of potential design principles to be comprehended. These are supported by a broader class of transformation principles, including expand-collapse, fuse-divide, and expose-cover, as well as supporting transformation facilitators such as furcation, function sharing, and shelling [16,35-37]. It presents a more comprehensive classification than the recent one proposed by Grassi et al. [41], except for adhesion changing functions which are more applicable to the multi-material assembly at the microscale.



Fig. 2. Functional classification of SMs

From this functional description highlighting the nature of expected effects and the stimulus, SMs can also be described and analyzed in terms of materials composition and distribution, if so the deposition mode or strategy through AM processes/techniques should also be considered.

2.3 Additive manufacturing processes

AM refers to a dynamic scientific and industrial domain, which is complementary to traditional subtractive and formative manufacturing [42,43,10]. It is spread over seven processes and underlying techniques to tackle ceramic, metal, alloy, composite (short fiber thermoplastics, continuous fiber thermoplastics, thermoset, etc.) and polymer deposition in a layer-by-layer manner whether at nano-, micro-, meso- or even macro-scale [44-49]. Among such well-adopted and developed processes, few have been used or customized to be suitable for 3D printing stimuli-responsive materials. These mainly include material extrusion, material jetting, vat polymerization, and powder bed fusion and more particularly their operational techniques illustrated in Fig. 3. One can also state that other processes like

powder bed fusion and binder jetting have also received reserved attention with shape memory alloys but presents – in their actual shape – limitations to build multi-material parts [50-52].

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. Although fused filament fabrication (FFF), direct ink writing (DIW) and material jetting (MJ) have intrinsically multi-material capacities through the usage of multiple heads and reservoirs [53-56], digital light processing (DLP), projection micro-stereolithography (P μ SL), stereolithography (SLA) and selective laser melting (SLM) techniques have also been customized to build multi-material parts [52,57-60]. Beyond these techniques, researchers are also paving the way towards hybrid strategies implemented through multi-techniques customized machines for specific material distribution and composition [61,62].



3. A review of 4D printing processes

The aforementioned section has introduced the key enablers of 4D printing to fulfil the desired transformation function. By extending traditional 3D objects with behavioral capability, 4D printing is one of the most appropriate manufacturing technologies to address biomimicry. Over the years, nature has received great attention of researchers and designers to design and build original shapes and structures whether in architecture, art and more recently in industrial products [38,63-66]. The latter seems to be legitimated with the advances of AM techniques and their ongoing adoption by the industry. However, to fabricate biomimetic objects ensuring the transposition of innovative mechanisms to solve engineering problems, the AM process should be associated with another ubiquitous driver, behavior [67-69]. As such, Fig. 4 illustrates 4D printing as a way to realize the biomimicry basics (structure, shape, and behavior), therefore introducing spatiotemporality and complexity issues [70].



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

The forthcoming sub-sections will investigate the research efforts towards this aim, with a particular emphasis on SMs development, homogeneous 4D printing and multi-material 4D printing. As for peculiar status and challenges in AM technologies, the reader may refer to relevant existing studies [71-73].

3.1 From SMs to homogeneous 4D printing

Nevertheless, it should be noted that pioneered efforts have been dedicated to shape-morphing structures with shape memory materials (SMMs) and also artificial muscles via electro-active polymers (EAPs) before the rise of AM technologies [74]. The long-term objective was to develop actuators and sensors for different application domains such as organic reactions, digital composites, tissue engineering, biomimicry and biomedical to name a few [75-77].

Among the stimulus-responsive materials, SMMs - through the development of shape memory polymers (SMPs) and shape memory alloys (SMAs) - can be thermo-mechanically trained to shift from one permanent shape to one or several temporary shape(s) from which the permanent shape can be recovered. 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). SMAs need to be heated at a certain temperature to shift from martensite to austenite and then to shape recovery back to their initial shape. The reversible (two-way) effect can also be realized via a more sophisticated trained process or through magnetic field stimulation but presents limitations for repeatability needed applications [78-80]. As for SMPs, the shape-changing function is ensured by the heat-induced phases change within the material but the thermomechanical training works in a different way [81]. It is needed to heat the material above a glass transition temperature (T_8) and then deformed it into the desired shape. Afterwards, it is cooled by maintaining the deformation strain. The recovery step of the permanent shape is then performed by heating the material at T_s . Some relevant works can be found to extend the actuation effect towards two-way, three-way and even repeatable effects [82-88] and the range of stimulus towards light-induced heating, light irradiation, infrared light, magnetic field, etc. [89-93]. Specific attention was paid to behavioral studies on T_{s} , stress magnitude and elasto-viscoplasticity deformation of materials [94,95,85,96].

As for electric-coupled materials, piezoelectric ceramic materials have been first developed. Although these materials are capable of generating large forces, they generally deform only a fraction of a percent; therefore limiting their applications to specific niches at nano- and macro-scales. To overcome this limitation, EAPs – which fulfil shape-changing functions (including size-changer) when stimulated by an electric field – have received huge interest as they can exhibit up to a 380% strain [97]. Their specific characteristic properties enable a large amount of deformation while maintaining significant forces [77,88,91,98,99]; then leading to the mass development of reversible artificial muscles [100,101]. One may make the distinction of two activation mechanisms: electronic (stimulated by electric field or Maxwell stresses) EAPs [102] and ionic (stimulated by mobility or diffusion of ions) EAPs [97], for which Table 1 gives a synthesis.

EAP type	Material	Description	Author
Electronic	Electrostrictive	The actuation is caused by electrostatic forces	[99,103,104]
	polymers and	between two electrodes that press the polymer.	
	dielectric	Dielectric elastomers are capable of withstanding	
	elastomers	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.	

Table 1. EAP materials

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 nanometers in size.	[105-107]
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.	[108-109]
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.	[114-117]
Ionic	Ionic/conducti ve polymers	Activation is caused by the displacement of ions inside the polymer.	[114]
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.	[115]
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.	[116-119]
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.	[120-124]

Based on the literature, EAPs seem to have better performances compared to SMAs in terms of response speed, density (lower) and resilience. However, it is important to emphasize their limitations in terms of low actuation force, mechanical energy density and robustness which may be constraining in practical applications [97]. Considered as electrostrictive materials counterparts, magnetic materials exhibit elongation along the direction of the applied magnetic field and are suitable for reversible effect [125,126].

Such fruitful developments have then been extended in terms of shape-changing functions to cover bending, twisting and folding especially once stimulated by solvent, light, magnetic or electric fields [127-130]. Others have addressed shape memory composites and photo or pH-responsive hydrogels presenting self-folding capabilities useful for origami-based applications [130-136] and in the context of self-healing materials [137,138]. In a general way, researchers have tried to embed programmed behavior into materials by tuning constitutive behaviors, material compositions and their couplings with the multiple available energy fields [139].

Such field-coupled material behaviors endeavor associated with AM processes and techniques have been initiated in 2013 under the umbrella of 4D printing paradigm, more particularly by using MJ technique [13,56]. From then on, research interest in 4D printing has seen an exponential increase [140]. By considering SMs in the broader context of AM, embedding behavior in matters seems to be within product designers' and engineers' reach. First research efforts were dedicated to homogeneous 4D

printing, i.e. 3D printing active parts made of a single material, whether by (i) programming behavior in classical polymer by tuning the AM technique and related process parameters or (ii) programming behavior through a single smart material [141]. Although Dufaud et al. [142] did not claim their contribution as part of 4D printing, they have introduced piezoelectric suspensions through a customized stereolithography process balancing rheological and photochemical properties to shape piezoelectric ceramics. Van Manen et al. [143], Goo et al. [144] and Wang et al. [145] have successfully proposed a 4D printing approach by tuning the deposition orientation of polylactic acid (PLA) material with FFF technique using hobbyist 3D printers to democratize one-way actuation mechanisms of 1D/2D structures. The same authors have designed specific deposition patterns to achieve selfbending/twisting and self-folding origami (i.e. ensure 1D/2D to 3D shape transformation in an autonomous and one-way manner), which take advantage of the constraints applied during the extrusion process and the different combinations of directional strains upon exposure to temperatures exceeding the T_{δ} (see Figs. 5a,b). In the same way, Kuang et al. [146] have adapted DLP technique using grayscale light patterns and a two-stage curing ink to process functionally graded materials (see Fig. 5c). This work reminds us that prior research efforts have been dedicated to highly stretchable and UV curable elastomers for DLP, then opening partial solutions for soft robotic applications [147]. Another example can be found in metamaterial mechanisms or mechanical metamaterials, which are 3D printed assembly-free mechanisms exhibiting deformations once stimulated by mechanical strain [148,149] or heat [150-154]. The proposed cell structures – which are composed of periodically distributed patterns printed with SLA, DLP, MJ or FFF techniques - exhibit soft and hard regions and also cells embedding bistable springs, which propagate mechanical signal in the consolidated part, then leading to unobvious and non-linear behaviors [155] (see Fig. 5d).



Fig. 5. Homogeneous 4D printing through tuned AM techniques, inert materials, cell structures and deposition pathways (adapted from [143] with permissions from the RSC, from [145] with permissions from the authors, from [146] with permissions from AAAS, and from [148] with permissions from ACM)

From a different perspective, a wide spectrum of stimulus-material-process couplings has emerged to address active functions (see Section 2.2), whether from 1D, 2D, or 3D structures. One may cite efforts towards 4D printed hydrogels as reviewed in Ahmed et al. [156], especially Gladman et al. [68] who have printed water-responsive composite hydrogel architectures encoded with localized and anisotropic swelling behavior. They have controlled the desired behavior via the alignment of cellulose fibrils along DIW pathways, therefore enabling shape-shifting from 2D to 3D structures. Similarly, Mulakkal et al. [157] have used DIW technique with responsive cellulose fibers-hydrogel composite ink to develop reversible water-responsive SMMs. Here the programmed behavior is based on the developed material and does not depend on printing pathways. Additional efforts have been made in 3D printed gel of laponite in agarose matrix combining thixotropic and thermal-reversibility properties respectively. The thermal actuation is then made via a training (cooling/heating) step that is similar to reversible SMPs programming process [158].

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 [113], which can be appropriately used as strips on flexible parts [61,159] or to address shape memory reversible behavior [160]. Others have fabricated single-layered humidity-driven actuators to go beyond typical bilayer structures, which may suffer from delamination after long-term exposure to moisture. It can be addressed via the control of the alignment of liquid crystals with cross-linked liquid crystal polymers that can be then be fixed by polymerization [161]. In the same direction, azo-benzene embedded LCEs have been widely developed in the context of photo-responsive soft actuators [162]. In general, such materials swell along to the direction perpendicular to the liquid crystals, and then the control is made possible through the printing pathways.

Besides, 4D printing SMPs is probably the field where a huge variety of transformation functions have been addressed with particular attention to the sequence of changes [163]. For example, Lee et al. [164] have proposed a sequential folding transformation by controlling the light absorption on a polymer sheet. They proposed different crease patterns with different sensitivities enabling the control of the folding sequence. Such interest in controlling the transformation planning has also been addressed by Hu et al. [165] and van Manen et al. [166]. Indeed, local stimulation is an interesting approach for programming the folding sequence [167,168], but tuning material response to a time-dependent global stimulus by locally controlling the material properties opens more alternatives [58,169], which require to address multi-material 4D printing issues.

3.2 Multi-material 4D printing and metamaterials

Multi-material (or heterogeneous) 4D printing can be seen as the mean of 3D printing parts made of spatially scattered/distributed materials to reach the desired behavior. The distribution here – influenced by the AM technique and related machines capabilities [170] – is of utmost importance and provides significant freedom in 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).



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

The multi-layers category relates an alternative deposition of layered materials, therefore generally leading to 2D-to-3D transformation dimension such as used in artificial muscles and origami-based applications. Researchers have tested multi-material FFF and MJ-based techniques to introduced SMPs with inert materials, or trained elastomeric materials with residual shape memory properties to operate bending and folding functions with one-way effect when stimulated by heat [56,171,172] and/or color-dependent selective light absorption [173]. Baker et al. [174] have programmed a sequential actuation combining ionoprinting and redox chemistry of 2D hydrogel membranes into 3D folded architecture. Moreover, Raviv et al. [175] have also demonstrated a linear stretching structure (from 1D to 1D), a 2D folding and stretching structure and a 3D topographical surface change through multi-material printing. Similarly, Ding et al. [176] have addressed cross-sectional multi-material to control multiple bending in wired structures. By exploiting PolyJet® technique capabilities, it is then possible to print elastomer and glassy polymer according to different orientations and afterwards train thermo-mechanically shape memory effects like previously done in [56].

Also, FGMs – seen as composites with gradual property variation over one or multiple directions – have recently received attention with the rise of digital materials [177,178], whether to produce parts

with adapted mechanical performances at the right place or to program active behavior (for example via adapted DLP technique [146], customized PLA-PEG filaments with FFF technique [179], etc.).

In the special pattern category, one may cite efforts towards 4D printed gels that can drastically and reversibly alter their volume in response to external changes, starting from water and thermo-active hydrogels [121], and their AM implementation by using DIW technique and alginate/PNIPPAAm ICE gel inks to build active valve [180]. We can also find research works in the field of active metamaterials as they exhibit periodic microstructures composed of material and void, or even multi-material and void [181], therefore promoting a wide range of desirable properties such as low weight, high stiffness and strength, negative Poisson ratio and energy absorption [182]. In short, metamaterials highlight unique structures currently limited to nano- and micro-scales that can control the propagation of waves and mechanical properties via the consideration of multiple small transformations. Among efforts made in heterogeneous active metamaterials, first attempts have been focused on composite hydrogels or anisotropic hygroscopic composite materials stimulated in water to cover reversible contraction/expansion (i.e. swelling/deswelling) functions. For instance, Mao et al. [183] have utilized MJ PolyJet® technique with hydrogels to realize sequential self-folding structures. Zhang et al. [184] have developed a bilayer structure composed of active hydrogel and inactive polymer, which enables the reversible shape-shifting of the lattice structure once dived in water (as shown in Figs. 7a,b). Thermo-responsive metamaterials have also been addressed using DIW technique and glass fiberreinforced poly(dimenthylsiloxane) multi-materials to build shape-shifting lattice structures [185], as illustrated in Fig. 7c,d. In the same way, Kuang et al. [186] have used DIW with shape memory composites to develop active metamaterials, whereas Roach et al. [187] have combined active LCE with thermally inert materials to realize shape-shifting metamaterials. More recently, Chen et al. [188] have proposed a customized DIW technique based on dynamic photo mask to print multi-material lattice structures with controlled buckling and crack propagation pathways. In addition to the developments made in water and thermo-responsive structures, researchers have built magnetoactive metamaterials by employing DIW technique and introducing hard-magnetic particles in a soft matrix material to enable quick and reversible shape-shifting behavior. In such a context, Ze et al. [189] followed by Ma et al. [190] have extended active materials responsiveness towards multi-responsive (both magnetic and thermal stimuli) magnetic SMPs.



Fig. 7. Example of soft active metamaterial with large negative swelling rations and tunable stress-strain curves highlighting (a) multi-material patterns build from MJ (PolyJet®) technique and (b) water-based actuation [184] (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 [185] (Reproduced with permissions from PNAS)

More recently, researchers – through the advances of connectionist artificial intelligence (AI) techniques and based on previous initiatives in material properties exploration in solid part [191] for multi-material printing – have addressed multi-material distribution computation by applying a voxel-based modeling approach [25,26,192] (see Fig. 8). By considering both the desired shape and an initial one – whether in 2D or 3D state –, their generative algorithm is able to find counter-intuitive distributions in solid parts that go beyond the well-known patterns illustrated in Fig. 6. However, for such distribution category, it is needed to use AM techniques and machines that are compatible with digital materials.



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. Intra- and inter-domain barriers

This aforementioned synthesis on significant 4D printing-related researches does not intend to replace insightful reviews that have been published over the last eight years. Most of them are dedicated to 4D printing processes [193-199], smart materials [200-206], origami-inspired active structures [207], kirigami structures [208,209] and 4D printing applications [87,210-218]. Mimicking nature with engineered materials and products is indeed a challenging task in design and engineering [219]. The literature review has been structured in a way to show the evolution, the status, and the spectrum of the abundant research efforts so as to clearly identify barriers to overcome to be potentially considered in design and engineering. Due to the nature of the studied scientific journals and the main pillars of 4D printing (i.e. transformation functions, SMs, AM processes/techniques, stimuli, and geometry), one naturally suspects that there will be domain-specific (*intra*) barriers and limitations related to domain interface (*inter*) issues, thus associated with the inherent interdisciplinarity of the research theme. But before addressing the subsequent barriers, it is relevant to conduct a preliminary analysis on the quality and the performances of the developed proofs-of-concept in literature, and then identify potential gaps with industrial applications.

4.1 Material performances

Most of the research efforts in 4D printing are focused on smart (often soft) actuators with sensing and shape-changing capabilities and need to be observed through key performance indicators (KPI) and mechanical properties. By following the traditional actuators classification proposed by Zupan et al. [220] and more especially their derived characteristics, some attributes may have specific attention from designers and engineers, such as actuation stress, actuation strain, actuation modulus, work output, work capacity, actuator efficiency, stroke work coefficient, etc., in order to define the industrial specifications. In the present analysis, to measure and monitor the performances of 4D printed actuators with SMs, such attributes can also be derived and expressed in terms of material properties [221], such as 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). As such, Fig. 9 reports KPIs ranges of the main SM categories (such as introduced in Section 3) used in 4D printing processes.



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.

Numerous studies have focused on the characterization of SMs to provide relevant elements for the selection of materials [222-227]. However, large disparities can be reported between the calculated/measured indicators which makes it difficult for non-experts to exploit them. This can be explained in particular by the different scales of measurement highlighting different temporalities, the materials customization and distribution, the size of the actuators as well as the different material forming processes over the last two decades. However, it should be noted current PKIs (illustrated in Fig. 9) are still low when they are compared to the performance of actuators (e.g. hydraulic, pneumatic, etc.) used in the industry. Most of the 3D printed SMs exhibit notable weaknesses in terms of 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.

4.2 **Process limitations**

Homogeneous or heterogeneous 4D printing is a complex process involving multiple steps to build smart objects such as fabrication, training (if necessary), stimulation, actuation and recovery (if possible), that can be carried out either sequentially or nested. Indeed, the training step can be considered as a pre-stimulation and actuation step that can be operated after [171] or during the material deposition step [143,228]. Actuation and recovery steps are directly related to the performances of the final printed objects, some limitations can be noted at the level of processes/techniques and stimulation stages that integrate materials and physics issues. To build the desired behavior, researches towards printing single SMs have shown limitations in terms of transformation functions but also of mechanical performances and have been largely extended to multi-material active structures. In this context of a complex transformation process, Table 2 presents the current process limitations to be considered. However, by using more sophisticated methods of 4D printing with heterogeneous principles, the realization of multi-material and multi-voxel systems through one or more 3D printers becomes conceivable, which could allow, in the future, to reach performances of industrial applications. The activity to be carried out will have to go out of a bottom-up logic offer and will have to associate requestor and designer in a more top-down complex logic of request. This is undoubtedly the "price to paid" if high industrial performances are targeted.

4D printing facet	Limitations	Origin
SMs elaboration	Availability in terms of quantity and type (powder, resin,	Inter-domain
	filament, fiber and pellet).	
	Need a tuning step.	
	Degradation of change capacity over multiple cycles (if doable)	
	Induce multi-scale behaviors.	
AM processes and	Material/process couplings limited.	Intra-domain
techniques	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.	
	Most techniques require a post-processing step.	
Stimulation	Temporal application of the stimulus and its propagation in the	Inter-domain
	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.	

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

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. Therefore, the next section will address research actions to cover the lack of knowledge, models, methods, tools to reconciliate great achievements made in designing SMs towards designing with SMs and AM technologies [229].

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

5.1 Main scientific and industrial stakes

The wide spectrum of research initiatives and the current limitations of 4D printing demonstrate that (i) it is a research area in full expansion due to the abundance of proofs-of-concept and the interest related to making active structures with AM technologies, but also (ii) it exhibits interdisciplinary facets that cannot be addressed in silos anymore. Interdisciplinary issues in product development have been tackled over the past decades concerning the types of technologies used within products and objects. The manufacturing industry has been indeed progressively forced to increase product/system capabilities and compact its lifecycles by ensuring a certain level of flexibility and efficiency as competitive edges, especially in the product development phase where design activities and decisions have a great impact on downstream processes (i.e. manufacturing, assembly, maintenance, etc.) [230]. This has been successfully achieved by considering (i) 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, (ii) the integration of other disciplines such as electronics in mechatronic system design and (iii) the integration

of computing capability in the era of the Internet of Things (IoT) for connected objects (see the grey boxes in Fig. 10) [231].



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

Figure 10 shows that such kinds of system have generated interesting and fruitful results in terms of theories, models, approaches and tools with a high level of capability maturity¹ (mostly from levels 4 to 5). One promising and emerging trend in engineering and interdisciplinary design – such as promoted by the European Factories of the Future Research Association [232], Manufuture [233], National Aeronautics and Space Administration (NASA) [229] and European Space Agency (ESA) [196] to name a few - consists in augmenting systems/products responsiveness and smartness once manufactured regarding user needs or customer expectations, and then increasing their lifetime by providing appropriate usage configuration in a specific use context and environment (see the blue box in Fig. 10) [20-23,234,235]. Such research actions fall under the broader context of industry 4.0 and its components like AM, soft robotics, and 4D printing. Over the last decade, the emergence of smart products/systems (i.e. products/systems that are capable to adapt to their environment, self-assemble, repair themselves, able to sense their environment and react accordingly in a programmable manner, etc.) and innovative AM technologies has required to deeply revise current well-established design and engineering models and approaches so as to deliver dynamic product definitions (i.e., product geometry embedding functional material and a transformation logic) at various levels of description (i.e., mechanical assembly, part, material, etc.). To pass to an operational and industrial threshold - where the inverse problem is mainly adopted by product architects and mechanical designers in their daily activities – 4D printing needs to be considered from a design and development perspective. Driving this manufacturing paradigm shift – with a focus on multi-material printing strategy – in the product development process poses grand challenges and opportunities for the industry. The following sections aim at drawing the key research directions to be tackled to successfully implement/apply a "design for 4D printing" vision for the development of smart systems.

5.2 Design for 4D printing roadmap

The roadmap, called Multi-X (X stands for different 4D printing components), seeks to map all 4D printing facets to deliver products-systems that make sense, meet use and live. To address such a long-term vision, it is relevant to consider multi-material, multi-domain, multi-process/technique/planning, multi-function, multi-physics, multi-world, multi-representation, multi-sector/niche, multi-scale and related temporality to which knowledge, models, methods and tools have to be setup in the systems/products/objects development process. This can also be done with the support of reasoning

¹ Level 1: Ad-hoc; Level 2: Repeatable; Level 3: Defined; Level 4: Managed; Level 5: Optimizing.

procedures like parametric/topology optimization, biomimetic computing, and connectionist and symbolic artificial intelligence (AI). Figures 11 and 12 provides an overview of the aforementioned research directions. It also shows the underlying complexity of the 4D printing facets and exhibits many research opportunities and challenges if they are associated each other.



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



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

5.3 Design framework of stimulus-responsive structures

With the mass development of proofs-of-concept to illustrate stimulus-responsive behaviors in objects and structures, it becomes important to think about a general design for 4D printing framework that promotes an inverse problem way to build innovative solutions. Compared to well-adopted design for AM, manufacturing, and assembly methods [28-30,236-240], the complexity level induced by the

multiple involved stakeholders (i.e. product architect, product designer, material expert, AM process planner, mechanical engineer, etc.) and the inherent spatiotemporal behaviors of active materials require a progressive integration of the 4D printing constraints, whether at the functional, logical/behavioral, structure, and geometric levels, and also the introduction of user-centered design innovation mechanisms. As such, Fig. 13 presents a process flow over conceptual, embodiment, and detail design stages, which highlights the top-down definition of stimuli-responsive products-systems and also the concurrent consideration of stimuli, SMs, and AM knowledge. The introduction of a design innovation process flow provides a flexible way to continually focus on the desirability of all involved stakeholders and the opportunity to inject creative, inspired, and user-centered inputs into the design stages [242].



Fig. 13. General design for 4D printing framework

By considering such a general framework with AM-enabled design theory and methodology [241], new critical steps have been introduced and well-adopted ones now exhibit additional consideration and actions [22,242]. Each part/step of the design framework represents in their own a set of opportunities and challenges and requires decision supports, heuristics, guidelines and rules. In its actual shape, the main steps can be processed in a proactive manner to involve in the early design stages SMs and AM critical decisions leading to a multi-perspective development (i.e. structure, geometry and properties), and offers enough flexibility to incorporate any reasoning procedures [243].

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

Following an inverse problem approach leads to apply a top-down design strategy. To do so, this broad research area requires the establishment of a strong knowledge base relating to stimuli, SMs, product and manufacturing/stimulation/actuation process models. Among the current efforts in material formalization [244,245] and AM knowledge capture [246,247], the HERMES domain ontology has been built to formalize 4D printing knowledge at the product/part level by considering basic formal ontology (BFO) and 3- and 4-dimensionalism theories [31]. Building a booming research domain ontology with the BFO upper ontology – as illustrated in Fig. 14 – ensures its accessibility for any other domain ontology such as promoted in the Industry Ontology Foundry (IOF) initiative [248].



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

In such a context, the knowledge capture should be thought accordingly with the knowledge reuse and reasoning scenarios, but also with published research works and standards from real-time updates (see Fig. 14). Next research efforts have to address this issue and extend 4D printing knowledge whether at system and materials levels or even to cover soft robotics [249] and structural electronics while ensuring their semantic alignment. If this research direction is appropriately addressed, then symbolic AI reasoning procedures can be developed as qualitative decision support in different selection issues and design recommendations. It can also have a critical role in providing meaning in connectionist AIbased design approaches [250,251].

5.5 Functional modeling and design principles

Considering x-changing requirements to develop deployable, reconfigurable or transformable systems will introduce additional transformation functions or dynamic functions to cover different states (whether from manufacturing to usage phases or simply during the usage stage) [16,252-256]. If enough detailed, such functions can directly be related to solution principles based on SM/stimulus couplings. The latter can be incorporated at this stage as SMs exhibit a functional facet. The functional modeling of stimulus-responsive systems requires mapping technical functions, materials, signals, and information. As such, using the Black Box model promoted in [4-6,39,255] could be a suitable strategy, to which challenges consists in :

- composing transformation functions with multiple basic functions (for instance, a bending can be decomposed into several expansion/contraction functions);
- consolidating technical functions with multi-material and multi-functional objects;
- integrating technical and multi-material functionality with user-centric, user-interface, user-experience, and systems functions;
- advancing design for AM and design innovation with AM approaches for transformation, multi-material, and multi-functional capabilities through 4D printing [22,236,237];
- elaborating bio-inspired AM, creative, and technical design principles that match shape/behavior/structure with SM/stimulus distribution patterns [256-266];
- setting up mechanisms for design principles recommendation.

By following the initiative promoted by Sarica et al. [267] to generate design ideas from a technology semantic network, it would be challenging to reconciliate 4D printing knowledge with natural elements. This direction could largely contribute to the emergence of innovative principles.

5.6 Smart materials/stimuli selection and transformation planning

Selecting the best design concept to be developed requires beforehand a comparative analysis of the generated solution principles in regards to the end-user requirements and functional specifications. The concept can emerge either from a single design principle or a concatenation of multiple design principles. However, in order to enable engineers and designers to understand and make decisions, it is important to provide assistance in the selection of active materials and stimuli. This can be addressed through the establishment of transformation plan describing the desired changes over time. To do so needs to define the stimulation application strategy, whether global, local, internal, external or a combination, accordingly with the stimulation mode (autonomous with external environment or controlled with the integration of a stimulator in the structure). Once the transformation strategy defined, the selection of SMs/stimuli can be processed. However, such critical step suffers from a lack of SM selection tool in product design [268]. This is mainly due to the complexity of the ad hoc experiments found in literature that highlights a wide spectrum of parameters (stimulation amplitude, stimulation flow, material composition, size, shape and distribution, 3D printing parameters, etc.). Currently, there exists no well-adopted database to compare SMs/stimuli. Especially since there is no description of the number of times that the 4D printing effect can be achieved by the used SMs, nor the recovery percentage that is attained after a predefined number of cycle. Building a standard protocol for testing SMs would drastically increase their adoption in engineering design and design education. A first challenge consists in establishing SMs KPIs similar to those existing for measuring performances of actuators and sensors as mentioned in Section 4.1. Another opportunity is to use AI techniques to subtract data and information from published literature and populate a 4D printing ontology (as introduced in Section 5.4). The ultimate objective is to appropriately select SMs/stimulus able to fulfil desired change performances requirements. This research direction should be addressed in consistency with the American Society for Testing of Materials (ASTM) current standards.

5.7 Design spaces definition and CAD representations

Along the proactive design for 4D printing process, the CAD representation of the solution being designed needs to be addressed according to different abstraction levels, viewpoints and scales (see Fig.15). In Fig. 13, the top-down design has highlighted CAD modeling strategies whether for behavior specifications with geometric skeletons (i.e. line, point, plane, sketch, etc.) [269,270] or 3D design spaces (i.e. functional surfaces, volumes-envelope, etc.) with geometric modeling to specify the overall geometry of the product/part and the active/inert regions, or even for properties, materials and stimulus with multi-scale voxel based modeling. Such strategies are necessary to tackle the inherent inverse problem approach to designing for 4D printing, and offers different abstraction level spaces to introduce advanced reasoning procedures. At the design space level, a first challenge consists in building geometric skeletons to control the shape-changing behavior of the structure and also allocating active and inert regions where specific computational attention is needed. Depending on such geometric input, the multi-material distribution requires a specific CAD representation in order to determine the appropriate properties at the right place, therefore fulfilling both a desired transformation at the digital level and multi-material 4D printing. Thus, multi-scale voxel based modeling seems to be extremely strategic to enable both the design of the material and the design with the material [229].



Fig. 15. Top-down construction of the design spaces [243] (with permissions from Elsevier)

Another challenge covering embodiment and detail design stages is to align and reconciliate the aforementioned set of CAD representations [243]. The progressive evolution of abstraction levels offers suitable CAD representations for knowledge reuse and generation of multi-material solid and latticed solutions. However, the complexity of the generated 3D models demands the adoption of an appropriate data exchange format whether to capture critical information at different scales or to exchange the information to manufacturing stage [271]. Among the most used data representations for AM purposes, one may identify standard tessellation language (STL) [272], wavefront object (OBJ) [273], additive manufacturing file (AMF) [274], 3D manufacturing format (3MF) [275], to which important industrial attention are paid. In terms of structure, AMF seems to be the most suitable representation format to consider multi-scale and multi-material data and exhibits common interest for academic and the industry.

5.8 Smart material/stimuli distribution vs. topology optimization

The transformation function to be designed and realized as well as its required performances lead to the distribution issues of multiple materials and related properties at the right place in the final geometric definition of the product/part. Implementing a primitive behavior in a primitive shape can be done through the usage of well-known distribution patterns illustrated in Fig. 6. However, such a digital definition of the multi-material structure can only be established by considering beforehand AM process and planning constraints. AM process selection and planning are a well-known and well-addressed research issue [276,277], but the multi-material AM processes and techniques – such a highlighted in Fig. 3 – introduce compatibility issues regarding materials and distribution patterns that are intrinsically related to deposition capacities (layer-by-layer or even volume-by-volume). This selection step must be carried out on the basis of the stimulation strategies. For example, if a stimulator must be integrated into the structure to produce an internet and local stimulation, the manufacturing plan will have to take into account the integration of this element during the AM process or afterwards by an assembly operation. The selection of the AM process therefore guides the design choices in terms of materials and their distribution in the structure.

In such a context, many challenges can arise to tackle multi-material/stimuli distribution, especially if a multi-scale modeling strategy is applied, and can be developed as follows:

• Multi-scale modeling (bottom-up, top-down and scale bridging approaches) and multiphysics simulation of spatio-temporal phenomena in multi-material structures. The applications scales of 4D printing exhibit different dynamics and temporalities, therefore extending scale-specific models with scale-bridging models will play a role in coupling particle-based and continuum-based models. Addressing this long-term objective will contribute to the needed real-time simulation-driven design of SMs/stimuli with quantitative predictive capabilities of their structure and properties [278];

- Computing materials distribution in 3D design spaces (where active regions have been defined) at different scales (macro, micro, nano) on a voxel/octree basis in a way to promote an inverse approach;
- Integration of multi-scale topology optimization (continuous or discrete) in multi-material structures coupled with parametric optimization [279,280];
- Discovery of materials distribution patterns, metamaterials and multi-scale mechanisms based on connectionist AI (e.g. machine learning, deep neural network, etc.);
- Consideration of bio-inspired generative design strategies for voxels structure definition. Possible research directions towards voxel automata, L-system, digital morphogenesis, etc. would provide means to ensure the growth of the multi-scale geometric definition.

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

Applying a design method also requires a dedicated digital thread [281-283]. Figure 16 illustrates a digital chain to build smart systems with 4D printing technology that is composed of systems engineering, product design, multi-physics modeling and simulation, process planning and process engineering. This chain has merit if appropriately coupled with a 4D FabLab value chain starting from raw materials to a combined manufacturing, assembly, 3D printing steps followed by training, stimulation and actuation. The digital thread can be set up through the connection of the related information systems and tools covering both digital and physical models. To do so, a scientific challenge consists of introducing a strong and active knowledge base associated with a digital twin in a way to (i) improve theoretical and simulation models from physical data, (ii) ensure the cyber-control of physical systems, but also (iii) enable a seamless reconciliation between both worlds through feed-back and feed-forward information flows.



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

6. Conclusion

Over the last decade, 4D printing has received growing academic interest, initially from material science, process science and mechanics. Given the large number of proofs-of-concept demonstrations

proposed in the literature, it is obvious that this technology – which is highly interdisciplinary – provides greater freedom to build structures capable of changing over time under the effect of energy stimulation. This tangible advantage – featuring the functional augmentation over time via consolidated parts – brings out complexities and paradigm shifts in design. This article has been conducted towards a wide analysis of the published research ranging from smart materials to homogeneous and heterogeneous 4D printing to identify current limitations and underlying *inter-* and *intra-*domain barriers. Built on this, the authors have drawn a long-wind research roadmap for engineering design to tackle "design for 4D printing". As a whole, opportunities and challenges have been highlighted and developed in a way to increase this emerging technology to an industrial threshold. In its actual shape, 4D printing exhibits useful capabilities to play a crucial role in the era of Industry 4.0 but also introduces new usage scenarios that deserve to be investigated in terms of human factors and experience for the adoption of stimulus-responsive structures in society.

Industry and research societies with the support of international and national research agencies are currently going ahead to embrace 4D printing processes, technological solutions, and 4D printing applications. Among the latest industrial concepts is BMW's concept car called Vision Next 100 [284], which features a deformable body and a cockpit that can be reconfigured according to driving mode. The Hankook company has also developed a wheel concept called Hexonic that can change size and texture depending on the weather conditions [285]. Such potential applications trigger calls for projects within the framework of European programs, ESA programs, and even in countries supporting the development of emerging and risky scientific themes (e.g., USA, Singapore, France, etc.). Current expectations are also focused on the high-throughput (smart) materials research and development for commercial 3D printers or even hobbyists ones.

4D printing demands to go beyond coupling AM machines with SMs and requires groundbreaking innovations to address multi-material manufacturing. As an example, this trend leads to startup initiatives like GenIsis in the context of a CNRS (French National Center for Scientific Research) maturation program that aims to develop hybrid 4D printing solutions (in terms of tools and machines) dedicated to industrial applications [286].

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