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# <span id="page-1-0"></span>Smart materials in additive manufacturing: state of the art and trends

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### **ABSTRACT**

Additive Manufacturing or 3D Printing has a great potential to develop significant advances in materials, printers' technology, and processes. Thus, the layer by layer manufacturing has existed for three decades and new developments recently appeared in smart materials. Laboratories discovered ways to design and manufacture advanced structured materials and responsive materials used in multi-functional and high-performance products. The current research and development efforts will have an impact on the traditional design and manufacturing process. 4D Printing announces a major modification in the product design and manufacturing process from static structures to dynamic structures like Shape Memory Material (SMM) with integrated functionalities. This article presents a review of smart materials based on a classification of advanced structured materials and responsive materials before beginning a description of current applications. The use of multi-materials and the study of predictive models to simulate the responsive materials behaviour accelerate the smart materials development.

### ARTICLE HISTORY

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### **KEYWORDS**

Smart material; shape memory material; additive manufacturing; 4D printing; bioprinting; advanced structured materials

### Nomenclature



# 1. Additive manufacturing

Additive Manufacturing (AM) or 3D printing has been developed to support the engineers and designers throughout the product-process design to physically check some functions. The first technology was created in the 1980's to produce models and prototype parts. The additive principle is based on a layer by layer manufacturing, which begins with a three-dimensional object using computer-aided design (CAD) before slicing a STL (Standard Tessellation Language) format in layers by a specific software. Among the major advances that this process presented to product development are the time and cost reduction, human interaction, and consequently the product cycle development (Ashley [1991\)](#page-15-0). Pushed forward by a growing demand and a patent expired effect, many manufacturers and AM solutions appeared. The demand for additive manufacturing machines is increasingly growing since the 90's (Wohlers [2012](#page-18-0)). Areas of interest that have used 3D printing to create objects include aeronautics, architecture, automotive industries, art, dentistry, fashion, food, jewellry, medicine, pharmaceuticals, robotics and toys (Bourell, Leu, and Rosen [2009](#page-15-0)).

It is possible to produce complex shapes with AM technologies compared to classical manufacturing processes (ex. milling, molding, stamping, etc.). Many attributes influence the outcome and depend on manufacturing technology type. Manufacturing

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<span id="page-2-0"></span>direction, model orientation and material behaviour based on the manufacturing technology have to be studied in order to integrate them into the engineering design (Beaman et al. [1997](#page-15-0)). The use of multi-materials and different complex structures allows the addition of static or dynamic functionalities and thus enables mechanical, thermal or electrical applications.

For a decade, structural materials usable through functional and high-performance setting, multi-functional and responsive materials have enriched the AM's perspectives. Some adaptative or shape memory materials (SMMs) interest the industrials for specific applications (biomedical, textiles, electronics or nanotechnology). Although a considerable amount of progress has been made in the smart material field, there is still a lot of research work to be done (Bogue [2012\)](#page-15-0). To better understand the new development and the stakes of smart materials, this article recommends a review of different methods. First, a reminder of main AM technologies begins this work. Secondly, a classification of smart materials is submitted to perceive a set of methods before showing trends. Finally, a chapter describes the industrial and academic trends.

# 2. AM technologies

This section presents the most common technologies used in Additive Manufacturing with a classification from (Gardan [2016](#page-15-0)). Each AM technology has its own design and manufacturing constraints related to the printing method, chosen material and expectations (aesthetic, mechanical behaviour, usage, etc.).

# 2.1. Laser technologies

SLA – Stereolithography is the first of the technologies developed originally and simultaneously in FRANCE (CNRS- July 84. French Patent N′ 84 11 241) and in the USA (U.V.P- C. HULL Aug, 84. USA Patent N°45 75 330) to tackle Rapid Prototyping bottlenecks, as well as faster and better design needs (CAD induced) (Jacobs [1992](#page-16-0)). Photolithography systems build shapes layer by layer with a laser beam that scans a section over a photopolymer resin. Subsequently, a higher resolution machine was later developed and called microstereolithography. It can print a layer with a thickness of less than 10µm (Halloran et al. [2011\)](#page-16-0). In microstereolithography, an UV laser beam is focused to 1–2 µm to solidify a thin layer of 1–10 µm in thickness. Submicron resolution of the xyz translation stages and the fine UV beam spot enable precise fabrication of real 3D complex microstructures (Zhang, Jiang, and Sun [1999\)](#page-18-0).

SLM – Selective Laser Melting – The system starts by applying a thin layer of powder material spread by a roller on the building platform. A powerful laser beam fuses the powder exactly at the points defined by the computer-generated component design data. The platform moves down, and another layer of powder is applied to keep on the process. During the process, successive layers of metal powder are fully melted and consolidated on top of each other. Today, the 3D printer manufacturers offer machines with powerful double or multi-laser technology with layers from 75 to 150 µm in thickness. The material types that can be processed include steel, stainless steel, cobalt chrome, titanium, and aluminum.

DED – Directed Energy Deposition – covers a range of terminology: Laser Engineered Net Shaping (LENS), directed light fabrication (IFF – Ion Fusion Formation), Direct Metal Deposition (DMD), 3D laser cladding. It is a more complex printing process commonly used to repair or add additional material to existing components (Gibson, Rosen, and Stucker [2010\)](#page-16-0). LENS is used to melt the surface of the target point, while a stream of powdered metal is delivered onto the small targeted point.

Other laser technologies exist as SLS (Selective Laser Sintering), DMLS (Direct Metal Laser Sintering) or EBM (Electron-Beam Melting).

### 2.2. Extrusion technologies

FDM – Fused Deposition Modelling – is a layer additive manufacturing process, which uses a thermoplastic filament by fused depositing. FDM was invented in the 1980s by Scott Crump (Crump [1992,](#page-15-0) [1994](#page-15-0)) and trademarked by Stratasys Inc. The comparable term is Fused Filament Fabrication (FFF). The filament is extruded through a nozzle to print one cross-section of an object, then moving up vertically to repeat the process for a new layer [\(Figure 1](#page-3-0)). The most commonly used materials in FDM are ABS, PLA, and PC, but there are new blends containing wood and stone as well as filaments with rubbery characteristics. The support is often made of another material and is detachable or soluble from the actual part at the end of the manufacturing process (except for the low-cost solutions, which use the same raw material).

DDM – Dough Depositing Modelling – groups the marginal processes that fit different doughs. Some technologies based on FDM printers use a syringe to deposit a dough material like silicone, food dough, chocolate, etc. A syringe based extrusion tool uses a linear stepper motor to control the syringe plunger position (Malone and Lipson [2007](#page-17-0)). The medical research industry uses the deposition of biomaterial and cells to realise a

<span id="page-3-0"></span>

Figure 1. Additive manufacturing principle with the Fused Deposition Modelling (FDM) example.

tissue structure. It presents a novel method for the deposition of biopolymers in high-resolution structures, using a pressure-activated microsyringe (Vozzi et al. [2002\)](#page-18-0). Other works show applications using a piston and 3D printer head adapted on a CNC machine to deposit a pulpwood based on wood flour to create a reconstituted wood product (Gardan [2014;](#page-15-0) Gardan and Roucoules [2011](#page-15-0)). New FDM printers (Markforged, Desktop Metal, AIM3D, etc.) use metal injection molding (MIM) pellets to extrude a filament charged in metal powder for 3D metal printing. A washing and a sintering step are necessary.

# 2.3. Flash technology

DLP – Digital Light Processing – proposed by Pomerantz (Barequet et al. [1996;](#page-15-0) Bieber et al. [1990\)](#page-15-0), uses the UV photopolymerized materials. A film is coated in resin and cured by a UV flash of light from a projector for each slice of the product. The DLP projector projects the entire section of the product, and not only lines or points. This method is much quicker than other methods due to scanning time of a laser. The parts can also be pulled upward out of the resin in a DLP printer with a part attached on a build tray to prevent damage when newly formed layers are peeled from the tray after each exposure (Dean et al. [2012](#page-15-0)). The building platform can be angled upward and the light source down in some masking machines (ex. Phidias technologies, Prodways). The DLP technology is known for its high resolution, typically able to reach a layer thickness of down to 30 microns.

Close to DLP principle, the Continuous Liquid Interface Production (CLIP) is a new type of additive manufacturing that uses photo-polymerization working continuously, thanks to a projector and the ability to control oxygen levels throughout an oxygen-permeable membrane. This last process is 30 times faster than the SLS or the MJM (DeSimone [2015\)](#page-15-0).

# 2.4. Jet technologies

MJM – Multi Jet Modelling – deposits droplets of photopolymer materials with multi jets on a building platform in ultra-thin layers until the part is completed. Two different photopolymer materials are used for building, one for the actual model and another gel-like material for supporting (Singh, Singh, and Saini [2010](#page-17-0)). The photopolymer layers are cured by UV lamps and a gel-like a polymer supports the complexity of geometry by wrapping it. The soluble gel-like (support material) is then removed by a water jet. The PolyJet technique reproduced details more accurately with a very good surface finish (Ibrahim et al. [2009\)](#page-16-0) and smoothness. The accuracy of a PolyJet machine can reach thickness from 50 to 25 µm and creates high resolution parts. Polyjet dual material jetting allows for the manufacturing of more than 100 composite materials. Multi-material 3D printing combines resins simultaneously in the same printing project with predetermined mechanical properties  $(Connect500<sup>TM</sup>)$ .

Binder Jetting or Inkjet is an additive manufacturing process in that a liquid binding agent is selectively deposited to join powder particles with a variety of materials including metals, sands, and ceramics (ExOne

<span id="page-4-0"></span>and Voxeljet). The printing process occurs when a liquid binder is spurt out in jets to steel powder (Wong and Hernandez [2012](#page-18-0)). A final treatment is required to solidify the part like sintering, infiltration, and finishing processes.

3DP – Three-Dimensional Printing, also known as CJP – Colour Jet Printing, combines powders and binders. Each layer is created by spreading a thin powder layer with a roller and the powder is selectively linked by ink-jet printing of a binder. The manufacturing tray goes down to create the next layer. The thickness of layers is about 90 µm to 200 µm. This process has been used to fabricate metal, ceramic, silica and polymeric components of any geometry for a wide array of applications (Moon et al. [2001](#page-17-0)). Other powders have been tested to realise green products in wood (Gardan and Roucoules [2011](#page-15-0)). 3DP can print in multicolour directly into the part during the building process from a colour cartridge. The final model is extracted from the powder bed to realise infiltration with liquid glue.

The modelling step is important in the AM process because it shapes the product, but it must also take into account some knowledge, manufacturing constraints and design choices. The post-processing to finish the product must be to consider in the process definition.

This review suggests classifying different methods which develop smart materials through the additive manufacturing principle in order to adapt them to their environment(s), function(s) or use(s). Figure 2 shows the classification of smart materials: advanced structured materials and responsive materials (Figure 2).

# 3.1. Advanced structured materials

The advanced structured materials are based on a static definition of complex shapes or a material's combination to achieve one or more properties that respond to a predefined functionality, like a smart material without transformation over time. In many fields of application, the development of new methods and processes must be accomplished by accurate and reliable modelling and simulation techniques (Öchsner, Altenbach, and da Silva [2012\)](#page-17-0). They translate into specific requirements that include high-strength-per-mass smart materials for vehicles and large space structures, materials with designed in mechanical, thermal or electrical properties, materials for high-efficiency energy conversion, and materials with embedded sensing or compensating systems for reliability and safety (Gates [2003\)](#page-16-0).

Several studies show different methods to improve the properties of the materials structure built by 3D printing. For example, Vesenjak et al. (Vesenjak et al. [2010\)](#page-18-0) studied the effects on lightened sandwich panels produced by additive manufacturing. Other studies brought the design and the manufacturing of specific structures like curved (Galantucci, Lavecchia, and Percoco [2008](#page-15-0)), honeycomb (Abramovitch et al. [2010\)](#page-14-0),



Figure 2. Smart materials: (a) Specific lattices, (b) Metamaterials and auxetic cellular, (c) Multi-materials topological optimisation, (d) Shape Memory Material (SMM) 1D and (e) 3D in self-assembly, (f) electroactive Shape Memory Polymer (SMP), (g) Biomimetic materials with fibrils.

3. Smart materials

<span id="page-5-0"></span>or cell shapes, 'tetrachirales' (Miller et al. [2010](#page-17-0)) or 'hexachirales' (Prall and Lakes [1997](#page-17-0)) [\(Figure 2\(](#page-4-0)a)). The flatwise compressive behaviour of tetrachiral and hexachiral honeycombs is analyzed, using analytical and Finite Element simulations, and tetrachiral and hexachiral cells are composed by cylinders connected by four and six tangent ligaments respectively (Figure 3). The pattern and its cylinders act as mixed stiffeners-elastic foundations during flatwise compressive loading. The specific lattices development is also used in the nanostructures and has numerous advantages especially for the making of metal nanostructures by material adding (Haggui et al. [2012;](#page-16-0) Hubert et al. [2005;](#page-16-0) Mao et al. [2017\)](#page-17-0).

Metamaterials create complex actuations that obtain their properties from the repeating patterns of their structures. The pattern is programmed into the geometry of the cellular structures or lattices (Saunders [2018\)](#page-17-0). Auxetic structures shrink or expand along two directions. Some auxetic cellular structures with negative Poisson's ratio present mechanical properties and deformation behaviours (Babaee et al. [2013;](#page-15-0) Saxena, Das, and Calius [2016](#page-17-0); Warmuth et al. [2017\)](#page-18-0) ([Figure 2](#page-4-0)(b)). The properties of auxetics depend on their symmetries and lead to various applications.

The numerical simulation tackles the study of stresses and allows designers to suggest complex geometries suited to 3D Printing advantages. In the same way, researchers describe a new filament deposition in Fused Deposition Modelling (FDM), which reproduces through a specific pattern the principal directions of the stress into cracking specimens (Gardan, Makke, and Recho [2016](#page-15-0), [2018\)](#page-15-0) ([Figure 4](#page-6-0)). The results show that this method improves of 30% the fracture toughness. The modification of filament direction leads to 'ductile-like behaviour' in the crack extension characterised by a large deformation zone associated with a slow crack growth rate during the crack propagation.

Another challenge is to reduce weight and decrease the material volume used, while keeping the product functions (mechanical, use …). Moreover, the main and support material can be expensive in the AM technology. Topology optimisation is a mathematical approach that optimises material layout within a given design space, for a given set of loads and boundary conditions so that the resulting layout meets a prescribed set of performance targets (Bendsoe and Sigmund [2003\)](#page-15-0). Using topology optimisation, engineers can find the best concept design that meets the design requirements. The topology optimisation method can be used to find the distribution of material phases that extremizes an objective function (e.g. thermal expansion coefficient, piezoelectric coefficients etc) subject to constraints, such as elastic symmetry and volume fractions of the constituent phases, within a periodic base cell (Bendsoe and Sigmund [2003](#page-15-0); Liu et al. [2018](#page-17-0)). Multimaterial 3D-printing technologies permit the freeform



Figure 3. Geometry of a chiral cell and results. Reproduced from (Miller et al. [2010;](#page-17-0) Prall and Lakes [1997](#page-17-0)).

<span id="page-6-0"></span>

Figure 4. Improving the fracture toughness of 3D printed thermoplastic polymers by fused deposition modelling (FDM) (Gardan, Makke, and Recho [2018](#page-15-0)): (a) Finite Elements Simulation, (b) 3D Printing.

fabrication of complex spatial arrangements of materials in arbitrary geometries by topological optimisation (Hiller and Lipson [2009](#page-16-0)) ([Figures 2](#page-4-0)(c) and 5).

In this multi-material perspective, the MJM technology suggests depositing different resins having different mechanical behaviours. A multiple-material is the combination of several photopolymers in specific concentrations and microstructures to create a composite material with a range of mechanical, optical or thermal properties (ex. Connex, Stratasys 3D Printer). The SLS technology can also be used in bio fabrication (§ 4.1). Some applications in drop-ondemand (DoD) micro-dispensing system is to deposit diverse materials with a wider range of properties (Li et al. [2009](#page-17-0); Sun et al. [2010](#page-18-0)). To satisfy this requirement, multiple print heads on micro level fabrication are preferred to perform the multi-material dispensing task. The FDM technology and multiple-nozzle (MJM) systems have demonstrated their capability to print 3D electrical components, resistors and capacitors (Wu et al. [2015\)](#page-18-0).

Eckel et al. (Eckel et al. [2016](#page-15-0)) reported a novel way to fabricate 3D printed ceramic parts using specific pre-ceramics monomers mixing with UV photo-initiator as shown in [Figure 6](#page-7-0). A few curved, complex, and porous shapes, such as corkscrew, micro-lattices, and honeycomb structure, can be easily fabricated using stereolithography (SLA). These ceramic materials are of interest for thermal protection systems, propulsion components, electronic device packaging, microelectromechanical systems, porous burners (Lee, An, and Chua [2017](#page-16-0)).



Figure 5. Results for the deflected shapes of three material beams, while additionally optimizing for the lightest structure. The low-density material (yellow, shown translucent) has an intermediate stiffness between the stiff blue and flexible red materials. Reproduced from (Hiller and Lipson [2009](#page-16-0)).

<span id="page-7-0"></span>

Figure 6. Additive manufacturing of polymer-derived ceramics: (a) UV curable resin, (b) SLA technology, (c) pre-ceramic polymer, (d) polymer-derived ceramic after pyrolysis, (e), (f), (g) and (h) ceramic structures examples. Reproduced from (Eckel et al. [2016](#page-15-0)).

These materials are smart because they respond or resist one or more stimulus, however, they do not interact with their environment. The next section describes the responsive materials from 4D Printing.

### 3.2. Responsive materials

Responsive materials are based on the 4D Printing process that demonstrates a radical shift in Additive Manufacturing. It entails multi-material prints with the capability to transform over time or a customised material system that can change from one shape to another (Tibbits [2014](#page-18-0)). While 3D Printing technology has been used to make static structures from digital data in 3D coordinates, 4D Printing adds the concept of change in the printed configuration over time, depending on environment stimuli (Choi et al. [2015\)](#page-15-0). The self-material structures would transform in shape or function by accounting for any environmental stimulus, which acts as a catalyser with a time-dependent.

Thus, Shape Memory Materials (SMMs) are gathered through Shape Memory Alloys (SMAs) and Shape Memory Polymers (SMPs), which can recover their original shape from a significant deformation (among other behaviours) when an external stimulus is applied and known as Shape Memory Effect (SME).

Neri Oxman introduces the ability to dynamically mix, grade and vary the ratios of material properties to produce functional components with continuous gradients, highly optimised to fit their performance with the efficient use of materials, reduction of waste and production of highly customisable features with added functionalities (Oxman [2011](#page-17-0)). Recently, Neri Oxman and the Massachusetts Institute of Technology's Media Lab have developed a water-based digital fabrication platform using a renewable polymer from the ocean to improve objects and buildings with biological materials that can adapt, respond, and potentially interact with their surroundings (Duro-Royo, Mogas-Soldevila, and Oxman [2015;](#page-15-0) Oxman [2011\)](#page-17-0). The process combines an age-old crustacean-derived material with robotic fabrication and synthetic biology in order to use graded material properties for hydration-guided self-assembly using biodegradable composites.

Typical stimuli to trigger the SME include heating/ cooling (thermo-response), light (photo-response), chemicals (chemo-responsive) (e.g. water/moisture, ethanol, pH change), mechanical loading (mechanoresponsive), etc. (Sun et al. [2012](#page-18-0)). SMEs are a kind of phenomenon in which a SMM recovers its original shape. Multi-materials or graded materials reproduces a cellular structure that dynamically changes shape. Thus, researchers investigate the shape memory process of printed active composite (PAC) strip to use the ability of cellular structures (Ge, Qi, and Dunn [2013\)](#page-16-0). For example, a laminated sample based on layers with fibers can transform into bent, coiled, and twisted strips when it is subjected to heating [\(Figure 7\)](#page-8-0).

The SMMs are stimuli-responsive materials that have the designed properties to enable return from a deformed temporary shape to an original permanent shape via application of an external stimulus, such as temperature, magnetic fields, light and moisture (Lendlein and Kelch [2002](#page-16-0); Yu et al. [2015\)](#page-18-0). An example of SMM is used in MJM technology to develop three connected letters printed, which were heated to above a

<span id="page-8-0"></span>

Figure 7. 4D Printing with shape memory polymer. SME in the temperature range between ~15°C and ~70°C. Reproduced from (Ge, Qi, and Dunn [2013\)](#page-16-0).

specific temperature and straightened at high temperature and cooled to return to the printed form [\(Figures](#page-4-0) [2](#page-4-0)(d) and 8).

The self-evolving materials are printed through multimaterial components that would transform into their designed shapes when they are exposed to water (Raviv et al. [2014](#page-17-0)). They are based on hydrogels that swell when solvent molecules diffuse into the polymer network and used to create actuating multilayer joints (Raviv et al. [2014\)](#page-17-0). The self-evolving materials with hydrogel can be extended to a broad range of matrices and anisotropic fibers.

In the same way, some researchers describe other functional SMMs, such as self-assembly (Hartgerink, Beniash, and Stupp [2001](#page-16-0); Ikkala and ten Brinke [2002\)](#page-16-0), self-actuating (Kim et al. [2012](#page-16-0); Meisel, Elliott, and Williams [2015](#page-17-0)) or self-sensing (Park and Wood [2013;](#page-17-0) Rendl et al. [2014\)](#page-17-0). Any approaches act with external stimuli like the self-sensing dielectric system which can contract artificial muscles. Self-assembly is a process that leads to a spontaneously form ordered aggregates and involves no human intervention. The structures generated in self-assembly are usually in equilibrium states ([Figure 2](#page-4-0)) (e)).

One example of SMA, that can exhibit both SME (thermal memory) and super-elasticity (mechanical memory), is the nickel-titanium (NiTi) SMA, which is characterised by its transformation temperature (Meier, Haberland, and Frenzel [2011\)](#page-17-0). However, the transformation temperatures are very sensitive to the variation in the Ni/Ti ratio. A slight drop in the Ni content can lead to a huge increase in the transformation temperatures (Frenzel et al. [2008;](#page-15-0) Frenzel et al. [2010;](#page-15-0) Meier, Haberland, and Frenzel [2011](#page-17-0)). NiTi parts are manufactured by a SLM technology, but the lower content of Ni is a problem due to its evaporation temperature (Khoo, Liu, An, et al. [2018\)](#page-16-0). A recent study showed that the laser absorptivity and heat conductivity of materials before and after a repetitive laser scanning significantly influences the final properties of SLM NiTi (Khoo, Liu, Low et al. [2018\)](#page-16-0).

The ceramic materials open opportunities of shape memory ceramics (SMCs) thanks to their excellent thermal stability after pyrolysis at one thousand degrees Celsius (almost no shrinkage was observed).



Figure 8. Printed sample of three letter before heating (a) and after heating (b). Reproduced from (Khoo et al. [2015](#page-16-0)).

<span id="page-9-0"></span>

Figure 9. Complex flower morphologies. Simple flowers composed of 90°/0° (a) and −45°/45° (b) bilayers oriented with respect to the long axis of each petal, with time-lapse sequences of the flowers during the swelling process. Reproduced from (Gladman et al. [2016\)](#page-16-0).

Biomimicry leads these researches to new solutions because it allows the reproduction of structures that exist in nature. Emerging pathways for mimicking these dynamic architectures incorporate materials that can respond to external stimuli such as SMA (Ge, Qi, and Dunn [2013;](#page-16-0) Ratna and Karger-Kocsis [2008\)](#page-17-0) and swellable hydrogel composites (Erb et al. [2013](#page-15-0); Thérien-Aubin et al. [2013](#page-18-0)) assembled by 4D printing (Gladman et al. [2016\)](#page-16-0) and linked to self-evolving materials. Inspired by flower opening and closing, Gladman et al printed hydrogel architectures with an anisotropic swelling behaviour based on the alignment of cellulose fibrils to do a functional folding flower. They printed two lattices with the ink devoid microfibrils oriented at 90°/0° and −45°/45° and a stimuli-responsive allows reversible shape changes in water of varying temperature (Figure 9). After printing under ambient conditions, the acrylamide monomer solution is photopolymerized and physically crosslinked by nanoclay particles, or glucose and nanofibrillated cellulose (NFC), producing a biocompatible hydrogel matrix that swells readily in water (Haraguchi and Takehisa [2002](#page-16-0)).

As explained previously, SMMs are also composed of SMPs that can fix temporary shapes upon heating. [Figure 10](#page-10-0) is an example of multi-material grippers with multiple SMPs (photo-curable methacrylate) that can grab objects when heated. Studies are able to design the time-dependent sequential shape recovery (Mao et al. [2015;](#page-17-0) Yu et al. [2015\)](#page-18-0) of a structure fabricated with SMPs.

The multi-materials are a key factor to trigger the stimuli-responsive material. Conceptual design of multimaterial active structures, such as piezoelectric actuators and active vibration control structures, have been studied using multi-material topology optimisation formulations (Kang, Wang, and Tong [2011\)](#page-16-0), or integrated optimisation of structural topology and control parameters of piezoelectric structures (Zhang and Kang [2014](#page-18-0)).

# 3.3. Discussion

Smart materials manufactured by AM, either advanced structured according to a predefined function or responding to diverse external stimuli, are limited by the small manufacturing scale. SMMs are not only identified through their functions but also through their external stimuli. A programmable multi-material begins with the original shape and through a step-by-step process finds one or a few intermediate shapes to activate the SME. The predictive models to simulate the geometry of 3D printing filaments is a trend in current research to improve mechanical properties (Gardan, Makke, and Recho [2018](#page-15-0); Gleadall, Ashcroft, and Segal [2018\)](#page-16-0). Thus, the 4D Printing design problem is way more complex than a conventional one in that, it involves designing a change strategy consistent with the desired functionality, a structure which is additively manufacturable, and made (partially or not) of stimulus-responsive (Sossou et al. [2018,](#page-18-0) [2017](#page-18-0)) and a physically realistic way on a voxel representation is proposed.

To predict the mechanical behaviour of FDM parts, it is critical to understand the material properties of the raw FDM process material, and the effect that FDM builds parameters have on anisotropic material properties (Ahn et al. [2002](#page-15-0)). The determination of influential

<span id="page-10-0"></span>

Figure 10. 4D printed multi-material grippers: closing and opening of gripper after heating and cooling. Reproduced from (Ge et al. [2016\)](#page-16-0).

parameters on the quality of parts realised by FDM has been tackled in many researches (Bellini and Güçeri [2003](#page-15-0); Lee, Dunn, and Wu [2005](#page-16-0); Montero et al. [2001\)](#page-17-0) and most of them use a classic structure of layers manufacturing and mechanical tests, such as tensile and compression, through a design of experiment approach. The process parameters of FDM have been optimised for lattice structures through a concept of Manufacturable Element that proposed to link the geometrical information of lattices structures and the manufacturing process (Dong et al. [2018](#page-15-0)). Besides, the anisotropic of materials created by 3D printing affects the mechanical strength. For example, compared to a conventional material, the strength of NiTi SMA is lower than steel (Meier, Haberland, and Frenzel [2011](#page-17-0)).

In topological optimisation, many algorithms have not been closely linked to or validated by AM – e.g. the heterogeneous two-scale topology optimisation algorithm and the robust topology optimisation approach, among others (Liu et al. [2018\)](#page-17-0). Furthermore, increasingly more open problems emerge, such as the residual stress constrained topology optimisation for metal AM (SLS, SLM and DED).

To evaluate some product and process parameters, researchers work on the Design For Additive Manufacturing (DFAM) and multi-objective problems based on the analysis of the AM technologies, production time and material mass to optimise the mechanical behaviours and the roughness of product (Asadollahi-Yazdi, Gardan, and Lafon [2018](#page-15-0); Huang et al. [2017](#page-16-0); Thompson et al. [2016](#page-18-0); uz Zaman et al. [2018\)](#page-18-0). The studies also tackle the optimal part deposition orientation according to the AM technology. The formulation of the physicsbased models requires an in-depth understanding of the process and is not an easy task in presence of

partial information about the process (Vijayaraghavan et al. [2015](#page-18-0)).

Few research studies have been conducted to improve the prediction ability of the GP (Genetic Programming) and the MGGP (Multi-Gene Genetic Programming) models by hybridising them. These approaches provide a mathematical equation reflecting the relationship between the mechanical behaviours and the given input parameters to predict the performance characteristics of prototypes (Garg, Tai, and Savalani [2014\)](#page-15-0). Shape memory can be prepared for optimisation of multiple shapes through these models. Thus, the parametric optimisation methods, metamodel or genetic algorithms are research tracks in engineering design to predict the behaviour of responsive materials produced by 4D printing (Paz et al. [2017](#page-17-0)).

There are no standards in Smart Materials in AM yet, even if IEEE Standards Association (IEEE-SA) hopes to build and mature global standards related to smart materials.

To conclude, the new SMMs development can lead to programmable material for actuation or motion following a pre-determined sequence, which could be defined through a physics-based model to act accordingly of external stimuli.

[Table 1](#page-11-0) sums up some information about smart materials with their AM technology, materials used and applications.

# 4. New trends in smart material

## 4.1. Biomedical

3D bioprinting shows significant promise for creating complex tissue and organ mimics to solve transplant



<span id="page-11-0"></span>Table 1. Smart materials and applications.

needs and to provide platforms for drug testing and tissue morphogenesis studying (Derby [2012](#page-15-0)).

In the work on multi-materials, a research presents a single bio-ink method capable of producing extrudable, gel phase bio-inks from a variety of materials, both synthetic and natural. It demonstrated with 35 formulations that bio-inks can be customised with regard to composition (additives, composites), degree of cross-linking, and polymer concentration in order to optimise structural and biological performance while maintaining printability (Rutz et al. [2015\)](#page-17-0).

It was shown that anatomically shaped constructs can be successfully fabricated, yielding advanced porous thermoplastic polymer scaffolds, layered porous hydrogel constructs, as well as reinforced cell-laden hydrogel structures (Visser et al. [2013\)](#page-18-0). The anatomically shaped tissue has been built of clinically relevant sizes, which can be generated when employing multiple building and sacrificial materials in a single bio fabrication session ([Figure 11](#page-12-0)).

Lately, Kang et al described an integrated tissue-organ printer (ITOP) system that can fabricate human-scale tissue with a multi-cartridge module to extrude and pattern multiple cell-laden composite hydrogels (Kang et al. [2016\)](#page-16-0).

Some research defines organ printing as a rapid prototyping computer-aided 3D printing technology, based on using a layer by layer deposition of cell and/ or cell aggregates into a 3D gel with a sequential maturation of the printed construct into perfused and vascularised living tissue or organ (Mironov et al. [2003\)](#page-17-0). Bioprinting is an attractive method to create tissues and organs at hospitals. The success of an implantation depends on compatible materials. Prosthetic is the first biomedical area that used 3D printing and it presents several successes. Several applications combine some degradable or allogeneic scaffolding with cellular bioprinting to create customised biologic prosthetics that have the great potential to serve as transplantable replacement tissue (Giovinco et al. [2012](#page-16-0); Mannoor et al. [2013](#page-17-0); Xu et al. [2013](#page-18-0)). Metamaterials and lattices are developed to fabricate open-porous cellular structures in 3D Printing (Compton and Lewis [2014\)](#page-15-0).

The potential of 4D printing to manufacture programmable biological materials with changeable shapes and properties can be a foundation for enabling smart pharmacology, personalised medicine, and programmable cells and tissues that can precisely target treatments for diseases (Choi et al. [2015;](#page-15-0) Faulkner-Jones et al. [2013](#page-15-0); Khatiwala et al. [2012](#page-16-0); Kolesky et al. [2014](#page-16-0)). An economical article showed that the medical 3D Printing market might reach 983.2 million US dollars by the year 2020 (Meticulous Research [2015\)](#page-17-0).

# 4.2. Textile

The potential of FDM materials, PLA, for use in 4D printing and the concept of combining PLA with nylon fabric for the creation of smart textiles has been investigated (Leist et al. [2017\)](#page-16-0). The objective is to design a textile product that can adapt to heat or moisture to improve comfort and to develop new functionalities. For example, childcare products that can react to humidity or temperature, or clothes and footwear that optimise

<span id="page-12-0"></span>

Figure 11. 3D-printed complex anatomical structures based on polycaprolactone (PCL) with polyvinyl alcohol (PVA) support. Vascular tree  $((A)–(C))$  (L × W × H = 67 × 42 × 8 mm3, vessel inner diameter 2–4 mm); right ear ((D)–(F)) (L × W × H = 63 × 41 × 13 mm3). CAD designs ((A), (D), (G)) showing permanent (red) and sacrificial (gray) components; printed structures (B), (E) showing PCL in bright white and PVA in off-white; PCL scaffold after sacrificing PVA support (C), (F). Reproduced from (Visser et al. [2013\)](#page-18-0).

their form and function by reacting to changes in the environment (Raviv [n.d.\)](#page-17-0). [Figure 12](#page-13-0) shows a multimaterial that displayed a convex deformation and concave surfaces via folding deformation and ring stretching deformation (Raviv et al. [2014](#page-17-0)).

Tibbits from MIT describes how self-tuning footwear might adapt to changing performance needs. Adaptive running shoes might work by sensing changes in the running surface or environment: a change from pavement to grass, sensed by the change in impact force, might cue the shoes to grow cleats. An active self-transformable material is suggested for comprising a flexible base material with an active material disposed on or within the flexible base material in a specific pattern (Tibbits, Papadopoulou, and Guberan [2016](#page-18-0)). Further, textile-based complex and 3-dimensional interior partitions and other wall treatments will be commonly used. Thus, metamaterials and lattices will influence developments thanks to their deformation properties.

Other applications in the fashion industry include jewellry and textiles (Yap and Yeong [2014;](#page-18-0) Zarek et al. [2016](#page-18-0)). The U.S. Army has tried to adapt this technology to produce camouflage textiles that help soldier hide in certain environments by bending the light reflected

from clothing (Rubežienė et al. [2008\)](#page-17-0). The smart-clothing is a new area in Additive Manufacturing and tackles other thematic like textile sensors or integration of commercial components.

# 4.3. Electronics and soft-robotics

A combination with both conductor embedding and robotic has been made in SLA or FDM. Thus, an electronics design approach suggests an implementation of a printed circuit board (PCB) like a advanced structured material. A pre-defined volume to accomplish the layout of components with complex circuit network is presented in the [Figure 13](#page-13-0). The use of existing electronics CAD design software for layout and routing of 3D printing electronic devices is possible when the 3D shape can be represented as a flat 2D surface (Macdonald et al. [2014\)](#page-17-0).

A dielectric elastomer actuator (DEA) is a sheet of elastomer sandwiched between two compliant electrodes and known as artificial muscle for its high elastic energy density and capability of producing large strains (∼200%), is chosen as the actuator for soft robotics (Cai [2016\)](#page-15-0). Each surface layer of the 3D printed

<span id="page-13-0"></span>

Figure 12. Complex 2D multi-material component exhibiting stretching and folding from left to right, and top to bottom. Reproduced from (Raviv et al. [2014\)](#page-17-0).

silicone was coated with graphite, capable of acting as a DEA. A tubular DEA is proposed by Coulter et al through a four-axis printing system capable of spray depositing multilayer tubular silicone membranes onto an air-permeable mandrel (Coulter and Ianakiev [2015](#page-15-0)). Self-actuating materials are suitable for applications such as RFID and displays (touchscreen) ([Figure 2](#page-4-0)(f)).

An alternate manufacturing method of a SMP that can be activated by means of resistive heating (referred as electroactive SMPs) is proposed by (Garces and Ayranci [2018](#page-15-0)). FDM printers can be used to manufacture these SMPs that tailored to electrically activate and increase their load bearing capabilities. A thermoplastic SMP and a conductive filler embedded polymer, e.g. a polymer matrix such as PLA with Graphene Nano-platelet particles, can be fed into an FDM machine to produce SMP with simple or complex shapes (Figure 13). Sensoria Fitness, a company in Redmond, Wash., that manufactures artificially intelligent sportswear, has developed smart fabrics embedded with textile sensors.



Figure 13. 3D printed signal conditioning circuit (on the left) and sample with an entire layer SMP in Graphene/PLA (on the right). Reproduced from (Macdonald et al. [2014\)](#page-17-0) and (Garces and Ayranci [2018\)](#page-15-0).

<span id="page-14-0"></span>MIT engineers created magnetic 3D printed structures that can transform their shape by the wave of a magnet. They could be used to develop remotely controlled softrobotics thanks to their fast-responsive speed to do many actions in a short time (Magnetic [3](#page-17-0)-D-printed structures crawl, roll, jump, and play catch | MIT News n.d.).

# 4.4. Aerospace

NASA has used 3D printing to fabricate some rocket parts, and their tests show that 3D printing can save time and reduce costs by 60% or more (McMahan [2013](#page-17-0)). Topological optimisation implementation of a model leads to a new lighter structure, while maintaining different conditions (mechanical, design shape, functions …).

A SMP reinforced by carbon fiber fabrics developed by Xin Lan et al (Lan et al. [2009\)](#page-16-0) has demonstrated the feasibility of a deployable structure. A prototype of a solar array was fabricated and actuated by this SMP to deploy it. The goal is to develop deployable aerospace structures.

A research identified one lattice with the optimal elastic performance for deployable Unmanned Aerial Vehicles (UAV) wing design. It proposed a lattice structure that is fabricated using an Objet 350 3D printer (MJM), while the material chosen is a polypropylenelike photopolymer called Objet DurusWhite RGD430 (Moon et al. [2014](#page-17-0)).

More recently, the emergence of 4D printing, where active materials are used in 3D printing, presents a potentially powerful extension of 3D printing for active structure and device applications (Ge et al. [2016;](#page-16-0) Tibbits [2014](#page-18-0)).

# 5. Conclusions

This article aims to review the different technologies in Additive Manufacturing before presenting smart materials and other trends. Additive manufacturing principle exists for three decades and new developments have recently appeared in smart materials. Thus, the review suggested a classification. Advanced structured materials and responsive materials are described with their specificities and their ontology. Laboratories focused on 3D printing processes with multi-material printing capabilities and new internal structuration based on computer engineering. The study of metamaterials and lattices have resulted in deformable materials that seem to have potentials in textile and tissue engineering applications. 4D printing will take over a wide range of application based on the Shape Memory Materials (SMM) that have the capability to change in

shape or function over time, responding to stimuli such as pressure, temperature, wind, water, or light. etc.

Use of complex geometries and multi-materials opens many possibilities of new functionalities to improve products and processes. Topological optimisation is adapted to propose new complex geometry which is easier to produce by additive manufacturing with weight gain while keeping a high mechanical behaviour. The predictive models to simulate the geometry of 3D-4D printing filaments is currently studied to improve mechanical properties of smart materials. Engineering developments could lead to programmable material for actuation or motion following a pre-determined sequence, which could be defined through a physics-based model to act accordingly to an external stimulus. A variety of natural organs such as a plant structure responding to an environmental stimulus are mimicked through biomimetic or bioinspired composites that can be 4D printed. The multi-materials development suitable to 3D printing is the decisive point to accelerate the growth of the smart material area.

The biomedical or 3D-4D bioprinting is an emerging technology to construct artificial tissues and organs which is currently feasible, fast evolving and predicted to be a major technology in tissue engineering. Perspectives are apparent in aerospace, textile, smart clothing, electronics, robotics and beyond. The standards of smart material produce by Additive Manufacturing are in ongoing projects.

# Disclosure statement

No potential conflict of interest was reported by the author.

### Notes on contributor

Julien Gardan is assistant professor at the EPF School of Engineering in Troyes since 2013 and attached to the University of Technology in Troyes (UTT). His thesis was based on the study of reconstructed wood products in 3D Printing, and he received his Ph.D. in 2011. He is responsible for a 3D printing laboratory in Troyes and his research spans the areas of Additive Manufacturing in DFAM and Smart Materials.

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