Smart Materials and Structures

TOPICAL REVIEW

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Topical Review

On 4D printing as a revolutionary fabrication technique for smart structures

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Abstract

Since the inception of additive manufacturing (AM) which is colloquially known as 3D printing, tremendous technological advancements have been achieved over the years thanks to the unwavering research efforts. Particularly, the progress in the 3D printable materials to include stimuli-responsive materials has been hailed as one of the most significant breakthroughs in AM. The combination of the smart materials and the AM's ability to fabricate intricate geometries has given rise to an emerging fabrication method which has been dubbed 4D printing due to its ability to imbue the printed structure with dynamic capabilities (4th dimension). 4D printing has received unprecedented research interest owing to its potential applications in a myriad of fields including medical, aerospace, soft electronics, morphing structures, and even fashion wear. In the spirit of furthering the concept of 4D printing; this review presents its general background, recent advancements, various methods of 4D printing, applications, current challenges, and future possibilities.

Keywords: additive manufacturing, 4D printing, smart materials

(Some figures may appear in colour only in the online journal)

1. Introduction

Since the antiquity, humankind has devised methods and means of converting the formless raw materials into precise and useful structures for their day to day applications ranging from household to industry. Over time, tremendous advances in such techniques have been achieved culminating into the well-acclaimed first industrial revolution of the 18th and 19th centuries. At the time, a massive transition from hand-based manufacturing methods into machines led to unprecedented growth in every aspect of the general population. It led to a birth of an era of advanced techniques aided by machines which we now term them as traditional manufacturing processes. In the said methods, the desired geometry of a structure is achieved by successive removal of material from a workpiece either manually or automatically using Computer Numerical Control (CNC) based machines hence termed as 'subtractive processes' and includes methods such as milling, turning, and drilling, etc. Forwards to about three decades ago, another revolutionary manufacturing technique emerged thanks to the advances in computer software and hardware technology. In contrast to the traditional methods, the emerging technology achieves the desired shape of the structure through successive deposition of extremely thin layers of the parent material one after the other and has been termed as 'additive manufacturing'.

At the time of its inception; additive manufacturing (AM) which is known to the general populace as three-dimensional (3D) printing and sometimes called rapid prototyping (RP) presented both a futuristic and a revolutionary method of fabrication [1] applicable to a wide variety of industries from aerospace, biomedical, to fashion and jewelry [2]. In fact, it has been hailed as one of the most innovative inventions of the 21st century. It possesses numerous advantages compared to the conventional methods with the topmost being its ability to fabricate parts with complicated geometries which would otherwise be impossible to achieve with other techniques. Indeed, 3D printing is now ubiquitous in the manufacturing world and has attracted the interests of both the industry and academia. A well-presented review of the status and challenges of 3D printing can be found by the work of Gao *et al* [3].

At its initial development, 3D printing was anticipated to bring a new paradigm in the way engineering structures are designed and manufactured. Campbell and Ivanova [4] named it as a disruptive technology that would have significant economic impacts. Undoubtedly, the overall mechanical behavior of a 3D printed structure depends on the properties of the base material. This has motivated researchers to focus on advancing the printer's material capabilities on top of other equally important research areas in AM as illustrated in figure 1. Particularly, the ability to print metallic parts such as those made of titanium makes 3D printing full of future promises [4]. Despite the myriad of advantages and possibilities of AM, there exist some limitations. In some applications such as in the medical industry, the printed parts lack the mimicry to the human tissue due to their static nature [5]. In other words, the printed structure for medical applications should be able to respond to the dynamic environment presented by a living body so as to achieve the intended function. The same is true for other applications that require the 3D printed structure to respond to some stimulus and perform certain functions such as self-morphing [6].

The necessity to have 3D printed dynamic structures for various applications has led to a whole new concept arising under the umbrella of AM and has been dubbed 4D printing [7]. Since its first introduction about half a decade ago, 4D printing has caused unparalleled attention from researchers all over the world because of its immense possibilities in the manufacturing world. In a nutshell, 4D printing encompasses the use of AM technology to create a structure that can change shape, property, or functionality over time in a predictable way when a specific stimulus is applied [8]. According to the original conceptor, the 4th dimension is the time factor required by the additively manufactured structure to change shape, property, or functionality when exposed to a stimulus [9]. The targeted change in shape, property, or functionality of a 4D-printed structure can be achieved in three possible ways. The first and the most common method is the AM of stimuli-responsive materials with the best example being the shape memory polymers (SMPs). As it is well known, SMPs are polymeric materials that can significantly change their properties such as shape in a controlled fashion when a specific stimuli e.g. heat is applied. SMPs stand out amongst other stimuli-responsive materials thanks to their unrivaled properties such as high recoverable strains, easy processing, and multiple activation methods [10]. In selecting the AM method for 4D printing, researchers should put into consideration the process parameters that affect the properties of SMPs such as curing behavior [11, 12]. The second is the AM of a multi-material structure where several materials are combined into one single structure [13]. The difference in material properties such as thermal expansion coefficient or swelling ratio will result into the desired shape, property, or functionality change. The third involves the utilization of deformation energy that relies on stress mismatch between two layers in the additively manufactured structure [14]. This is where mathematical modeling plays a crucial role in the design of the structure. The models help in the design, evaluation, and the adjustment of the parameters in order to realize the targeted transformation of the structure. Figure 2 presents a schematic illustration of the difference between 3D and 4D printing.

4D printing possesses a combination of two indispensable traits in manufacturing. The first one is the capability of the AM to accomplish intricate geometries whereas the second one is the capability of the printed structure to transform upon introduction of a trigger or stimulus hence the 4th dimension. The two mentioned traits of 4D printing make this technology open to a myriad of applications. As an example, the most noticeable advantage of 4D printing is its ability to significantly reduce the storage volume which comes in handy for space applications. The structure could be stored in its contracted shape which may be in 2D then deployed to the required shape upon exposure to a stimulus. The current widespread popularity of 4D printing in numerous research groups worldwide can be attributed to its increasing potential applications. Particularly, the pioneering work of Ge *et al* [15] demonstrated the possibility of utilizing 4D printing to realize self-assembling structures based on active composite materials. Moreover, the work of Gladman *et al* [16] introduced the concept of biomimetic 4D printing wherein the printed structure is inspired by the biological systems such as plant motions in response to stimulus like humidity or sunlight. They were able to design hydrogel-based composite structures with anisotropic swelling behavior when immersed in water. At the time of this writing, their paper remains the most cited in 4D printing papers. With the progress in research, advances in AM equipment, software, and material capabilities will make 4D printing the fabrication technique of choice for smart structures. In the same manner that the AM has revolutionized the manufacturing sector, 4D printing is envisioned to have an even more impact in the near future. Recognizing the potential of this disruptive technology, the current review aims to present a concise yet complete introduction of 4D printing that we hope would be highly informative especially to the ever-increasing number of upcoming researchers. Unlike most reviews in the open literature, this work presents on 4D printing as a fabrication technique with a focus on its process, different classifications, applications, challenges, and future directions.

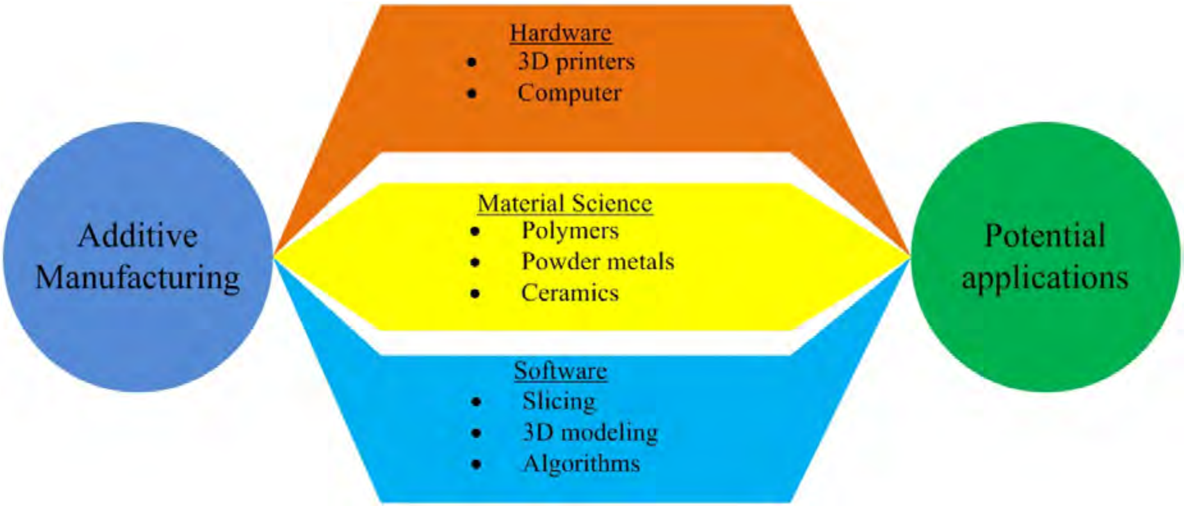

Figure 1. Schematic illustration of the main research directions in additive manufacturing.

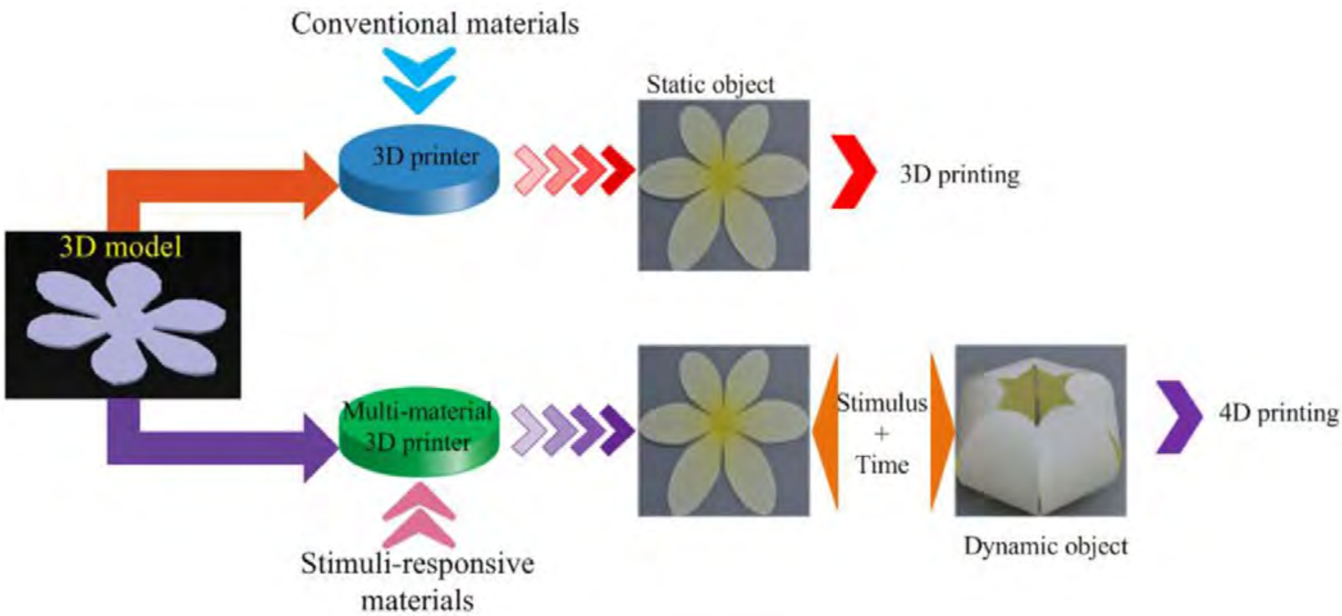

Figure 2. Diagrammatic presentation of the basic differences between 3D and 4D printing.

1.1. 4D printing process

Like many other modern technologies, 3D/4D printing would be non-existent without the technological advancements in other sectors. These include computer hardware and software. As for the former, the significant improvements in computer memory capacity and the reduction in cost made it possible to process huge amounts of data as in the case of Computer-Aided Design (CAD) models. The developments in computer software saw the unveiling of powerful 3D graphics and numerous professional CAD programs [17]. In principle, the AM process entails the realization of a physical model from virtual CAD data. Both 3D and 4D rely on AM to achieve the physical models and, therefore, their initial processes from CAD modeling to the point of printing are identical. Their main difference is on the material employed with the former using conventional materials such as aluminum whereas the latter requires smart materials such as SMPs [18].

For convenience and easy understanding, we would like to categorize the 4D printing process into three main steps. In terms of the process steps, 4D printing has one extra step compared to the generic AM process which the programming of the printed part. This, however, would be different in the case that programming is incorporated into the printing process. The generic 4D printing process is explained as follows:

1.1.1. Design. The first step in any fabrication process is the conceptualization of the product. Before proceeding, one should have a clear idea of the product he/she intends to achieve. This is mostly determined by the intended application. 3D/4D fabrication processes have the product concept represented in digital data using 3D-CAD software [19]. There are dozens of 3D modeling software all of which can accomplish this step. The user's choice depends on their expertise and also the part to be modeled as some are suited for certain geometrical designs. The creation of the 3D model data is not exclusive to the CAD software only but also reverse-engineering software such as laser scanning.

The CAD modeling software offers a number of file extension options to save the 3D model part. These include IGES, STEP, STL, etc. Most of the AM technologies work with an STL extension. STL (Standard Tessellation Language or Standard Triangle Language) file format goes back to the advent of the first commercial AM technology in the 1980 s (Stereolithography) and has become the de-facto file standard for describing the CAD model geometry data [19]. The second part of the pre-processing step, therefore, is to convert the 3D model into STL format.

1.1.2. Manufacturing. Since the AM fabrication technique forms the overall part in a layerwise manner, the first step in machine-processing is to sub-divide the STL file to form thin cross-sectional layers in a process known as slicing. There are several slicing software available both free and commercial. It is worth to note that slicer settings such as the layer thickness affect the overall quality of the printed part. Once slicing is done, the information is bundled into a *G-code* file which contains information about the model understandable to the printer. The file is then fed into the 3D printing machine.

Depending on the type of machine used, some settings are necessary before the actual printing. These include information such as the printing speed, the nozzle temperature, .etc once settings are completed, the printing process commences.

1.1.3. Post-processing. Most of the printed parts can be used directly upon removal from the machine. Depending on the complexity of the printed part, printing supports are required as part of the structure. This will necessitate further processing including the cleaning and removal of the supports before the component is used for a specific application. As already mentioned, 4D printed parts whose programming is not incorporated into the printing process require a programming step. Taking an example of the heat-activated material; heating of the printed component past the glass transition temperature then deforming to the temporary shape and cooling to the room temperature will be required to achieve the 4D functionality.

Generally, the number of steps required in 4D printing is more than that of their corresponding 3D printing. This has raised concerns with some researchers pointing that it makes the process complicated and less distinctive from the conventional fabrication of smart materials. The work of Ding *et al* [20] proposed the integration of the programming step into the printing process thereby reducing the number of steps from five to only three. As the research progresses, future 4D printed parts would be removed directly from the printing machine and utilized without further manipulations.

1.2. Pre-requisites for 4D printing

Unlike the conventional AM process, 4D printing ought to achieve the transformation of the printed part in terms of shape or functionality [21]. This increases the complexity of the process with some extra conditions to be met in comparison to 3D printing. The first requirement is on the printer itself. The printer should be compatible with the material intended for the structure. This is true for most of the cases where the dynamism of the structure is achieved by an introduction of a stimulus that will trigger shape or functionality change courtesy of its shape memory behavior. In some other cases, the mechanisms of achieving the dynamism of the part is the deformation mismatch induced by the difference in the properties of the material such as the coefficient of thermal expansion [20, 22, 23]. In such a case, 4D printing will require a multi-material printer in order to achieve part with different material type compositions. In contrast to a 3D printed structure which is static, a 4D printed structure is dynamic and it is crucial to predict the behavior of the part upon stimulus. Specifically, mathematical models are required to predict the desired final shape.

Without a specific stimulus, 4D printing would not be a success. There are dozens of stimulus methods applicable to smart materials such as heat [24], light [25], microwaves [26], water/moisture [27], magnetic field [28] etc. Most of the stimulus responses are not intrinsic to the material and, therefore, are tailored by incorporating fillers such as carbon nanotubes (CNTs) [29], magnetite [30, 31], silver nanowires (AgNWs) [32], Gold nanoparticles (AuNPs) [33], etc.

1.3. Advantages of 4D printing

Since 4D printing is based on AM technology, it inherits all the advantages of 3D printing and adds the advantage of having a dynamic structure. AM in itself has been hailed as one of the most promising and innovative technologies of our time that will lead to a new industrial revolution [34]. This is because of its remarkable advantages over the traditional manufacturing processes as has been reported by numerous researchers including Attaran [35] and Ford [36].

Once a product has been conceptualized and designed, it is the duty of the designer to opt for a manufacturing process that produces the part as efficiently as possible. Subtractive processes involve numerous steps from cutting of the bulk material to obtaining of the required geometry. These may include cutting, welding, drilling, and polishing, etc. In contrast, AM produces the part in a single step as soon as the 3D CAD model of the part has been completed and uploaded to the printing machine. The single-step fabrication eliminates the need for extra machinery, labor, costly tools, and saves on time leading to a significant reduction in the overall fabrication cost. Additionally, there is no material wastage unlike in the subtractive processes thereby saving much on the cost and enabling re-usability [36].

One of the most obvious advantages of AM technology is the ability to produce parts with complex geometry. In most applications such as in the medical industry, parts with complicated geometries are required which are not feasible with

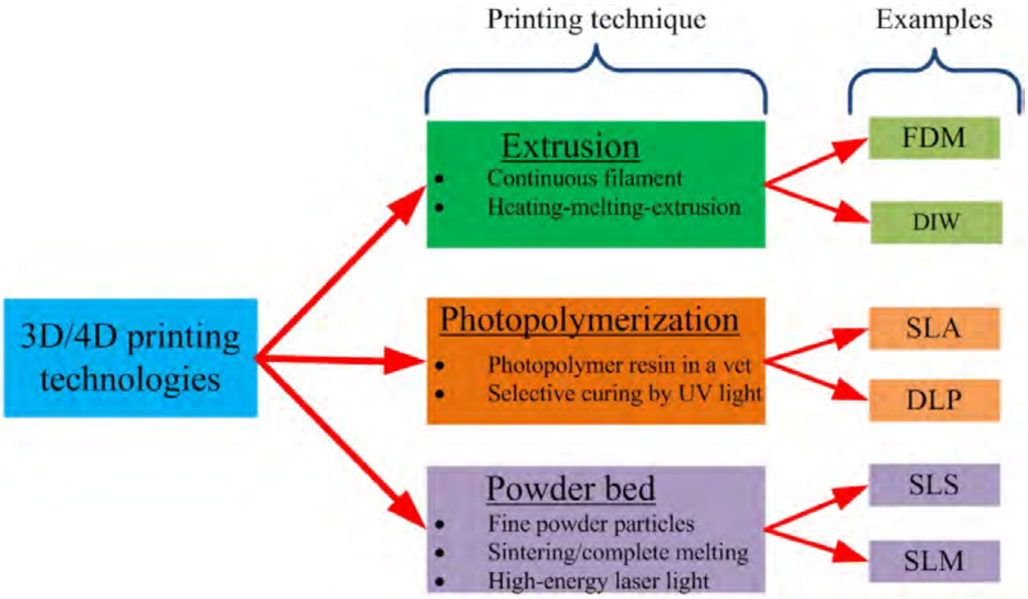

Figure 3. Categorization of 3D/4D printing methods basing on the printing technique.

the traditional manufacturing processes. Since the part begins with a digital format, AM makes it easy to modify, share, and customize the parts. This is highly advantageous as 3D CAD model data can be sent over thousands of miles and reproduced thus saving on time and transportation costs as recently done by NASA to Astronauts at the international space station [37].

As already mentioned, 4D printing achieves 3D printed parts with capabilities to change their characteristics such as shape or functionality under the action of a stimulus. This has principal advantages in various engineering and medical applications where dynamic rather than static parts are required [38]. Importantly, parts that cannot be printed because of their relatively huge size compared to the printer can be achieved via 4D printing since the action of a stimulus can change to the required shape. 4D printing is being used to invent new products and devices with wide range of applicability due to their novel properties. This is made possible by extensive research in the materials community leading to new materials such as SMPs with improved resin curability with nanosilica fillers [39]. As the materials used for 4D printing have to be smart materials, this technology advances the smart printed structures. It also enhances the technology of AM hardware and software to incorporate more advanced materials thus forming the basis for research and innovation.

2. Methods of 4D printing

It should be emphasized that 4D printing utilizes commercial 3D printing technologies to achieve the desired parts. To the best of our knowledge, there are currently no specially designed AM techniques for 4D printing. The only requirement, as stated in section 1.2, is that the 3D printer should be compatible with the selected smart material or multi-material printing capabilities for those cases where shape change is defined by the deformation mismatch in the structure. Discussion of the 4D printing methods, therefore, is basically the discussion of AM techniques.

Since the first invention of a 3D fabrication method in the mid-1980 s by Chuck Hull [40], there have been dozens of other methods introduced in the subsequent years. Although these methods form the part using the layer by layer deposition of material based on 3D CAD model data, they vary in the techniques employed. The variations have led the American Society for Testing and Materials (ASTM) International to classify these methods into various categories depending on the technique employed as shown in figure 3. A number of factors such as the starting materials, resolution, processing speed, costs, and the performance requirements of the final product influence the choice of the fabrication method to be employed [41]. A summary of the 4D printing methods is given in table 1.

2.1. *Extrusion*

As the name suggests, methods employing this technique fabricate a three-dimensional part by selectively extruding material through a nozzle over a movable platform layer by layer. The material is supplied into the printer in a filament form where it is heated at the nozzle to a semi-liquid state before being extruded [42]. The raw materials for this method should be able to be fused at adequate temperatures without degrading it [44]. Moreover, the materials should rapidly solidify at room temperature upon extrusion. In order to achieve a uniform cross-sectional area of the extruded material, both the applied pressure and the nozzle speed should be kept constant [44]. Two of the most commonly used extrusion-based fabrication methods are presented in the following sub-sections.

Table 1. A summary of 3D/4D printing methods showing the materials, applications, advantages, and limitations [42, 43].

Fabrication process	Methods	Materials	Applications	Surface finish	Merits	Limitations
Extrusion	FDM/FFF	Thermoplastics filaments e.g. PLA, ABS, Nylon	Rapid prototyping Concept parts Advanced composite parts	Standard	Low cost Versatile Simplicity High speed	Weak mechanical properties Limited materials
	DIW (Robocasting)	Plastics, Ceramics, Composites, Living cells	Packaging Scaffolds for bone regeneration	Standard	Flexible	Requires post-processing
Powder-bed fusion	SLS	Fine powders of polymers, alloys, composites, and ceramics	Aerospace components Light-weight structures Electronics	Standard	Fine resolution High quality Best mechanical properties	Low resolution High cost High porosity
	SLM	Fine powders of metals, alloys, and ceramics	Aerospace components Light-weight structures Electronics	Good	Good mechanical properties Wide range of materials	Slow process
Photopolymerization	SLA	Photopolymers UV curable resins	Biomedical Prototyping	Excellent	High precision Smooth surface finish Low cost	Limited materials Weak mechanical properties
	DLP	Elastomers, Photopolymers UV curable resins	Biomedical Prototyping	Good	High resolution High printing speeds	Expensive Requires post-processing

2.1.1. Fused deposition modeling (FDM). It was invented by S. Scott Crump in the late 1980 s and first commercialized in the year 1991 [45]. It is currently the most commonly used AM technique thanks to its numerous advantages including ease of use, low cost of the equipment and raw materials, non-requirement of chemical post-processing, faster printing speeds, and wide availability [44]. The working principle of this method is based on the hot-melt extrusion process where the raw material is heated until it melts then extruded through a nozzle. In this case, the nozzle is heated to the desired temperature and the raw material is supplied from a spool via a drive gear mechanism into the sprinter. Thin strands of the melted material (100–300 μm) are deposited onto a build platform according to the 3D CAD data to form the part. Most configurations have the extruder system connected to a three-axis system thereby enabling it to move in the three directions accomplishing a 3D part whereas other configurations have printing head moving in the x and z directions while the platform moves in the y-direction [46]. FDM supports a number of thermoplastic materials such as acrylonitrile butadiene styrene (ABS), polylactic acid (PLA), and nylon [41]. A schematic illustration of the FDM printing technique is shown in figure 4(a). The FDM is the most utilized additive manufacturing method to fabricate 4D printed structures especially those that are made of thermal responsive shape memory polymers [47]. An example of FDM printed 4D structure is shown in figure 5(a).

2.1.2. Direct ink writing (DIW). The high temperature needed to melt filament raw materials in the FDM process is disadvantageous in that it can degrade the material [53]. This has led to the development of other extrusion-based methods that do not necessarily melt the material. DIW, also known as Robocasting or Direct-Write Assembly (DWA), is one of such methods. It was invented in the late 1990 s at Sandia National Laboratories in the United States [54] as a method of fabricating geometrically complex ceramic parts. The viscoelastic material in its paste form (also known as ink) is extruded through a nozzle or syringe needle either pneumatically or mechanically [55]. Like in the FDM process, the nozzle is connected to a three-axis system and is situated above the build platform enabling it to form the 3D part in a layerwise manner. The ink should possess a relatively low viscosity under stress so as to enable it to flow through the nozzle without necessitating high pneumatic or mechanical pressure. Moreover, it should exploit the shear-thinning rheological property thus enabling it to retain shape upon extrusion. It should be noted that because of the shear-thinning property, the extruded ink behaves as semi-solid and, therefore, its shape retention does not rely on the solidification or drying as in the FDM technique. The schematic illustration of the DIW process is shown in figure 4(b). A 4D printed nanocomposite cylinder that was fabricated via DIW is shown in figure 5(b)

2.2. Photopolymerization

This refers to a number of 3D fabrication methods whose basic techniques involve the selective and spatial curing of a photopolymer liquid in a vet (tank) by a light source such as laser beam or ultraviolet light. The curing process ensures solidification of the material layer by layer according to the CAD data until completion of the 3D object. Examples of fabrication methods basing on this technique include stereolithography (SLA), digital light processing (DLP), continuous light interface production (CLIP), daylight polymer printing (DPP), and two-photon polymerization (2PP). A brief discussion of the two most common processes (SLA and DLP) will be presented in the next sub-sections.

2.2.1. Stereolithography (SLA). It is the first-ever additive manufacturing method invented in 1984 by Chuck Hull [40]. Utilizes high energy light (Ultraviolet or laser) to induce polymerization and subsequent solidification of a photopolymer resin or monomer solution [42]. Basing on the part CAD data, the resin is cured to a depth equal to the thickness of a single slice then the platform is translated along the z-direction enabling the built layer to be recoated with resin again and the process repeats until the part is complete. The excess resin not polymerized in the process is then removed and the post-processing of the part follows.

Depending on the orientation of the light source and the built platform, SLA can be classified into two categories namely ‘right side up’ and ‘Inverted’ as illustrated in figure 4(c) [55]. The former has the light source situated above the built surface thus forming the subsequent layers by lowering the platform whereas the latter has the light source beneath the build surface and has a transparent window in its base. The build platform moves up a distance equal to the thickness of an individual layer to allow for uncured resin flow under the cured part. Compared to other methods, SLA can produce parts of superior quality and operates at a relatively lower temperature [56]. A 4D printed shape memory polymer based bucky-ball fabricated by SLA method is shown in figure 5(c)

2.2.2. Digital light processing (DLP). Unlike SLA where the high energy light source is focused on a point of a single layer, DLP utilizes a digital light projector screen to flash a single image of the whole layer across the built platform at once [44]. As a result, DLP is a relatively faster (shorter built time) process compared to SLA. A schematic illustration of the DLP technique is given in figure 4(d). The application of DLP method for 4D printing was demonstrated by Zhang *et al* [51] through fabrication of a high-resolution self-healing structure based on shape memory polymer solution (see figure 5(d)).

2.3. Powder-based

Powder-based processes essentially employ the same procedure as in polymerization-based methods except that instead of the raw material as a photopolymer resin we have fine powder particles in a ‘powder bed or built platform’. Depending on the

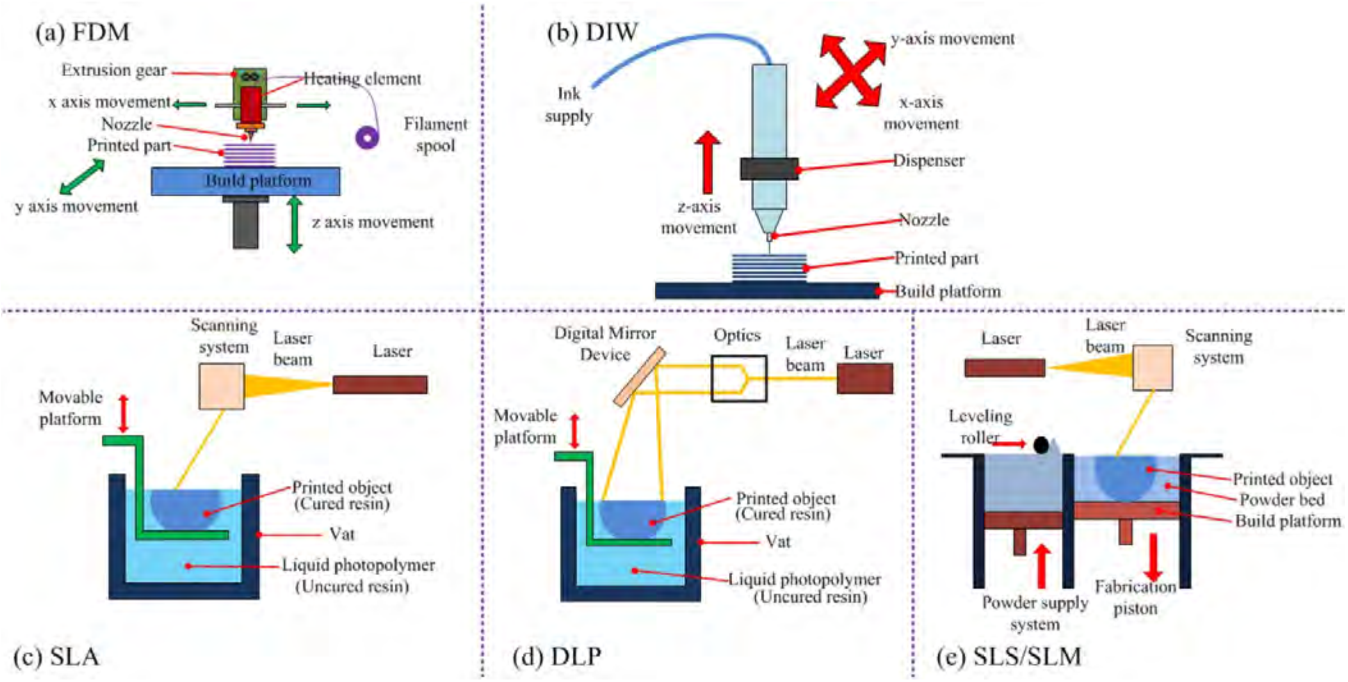

Figure 4. Schematic illustrations of some of the 4D printing methods. (a) FDM, (b) DIW, (c) SLA, (d) DLP, and (e) SLS/SLM.

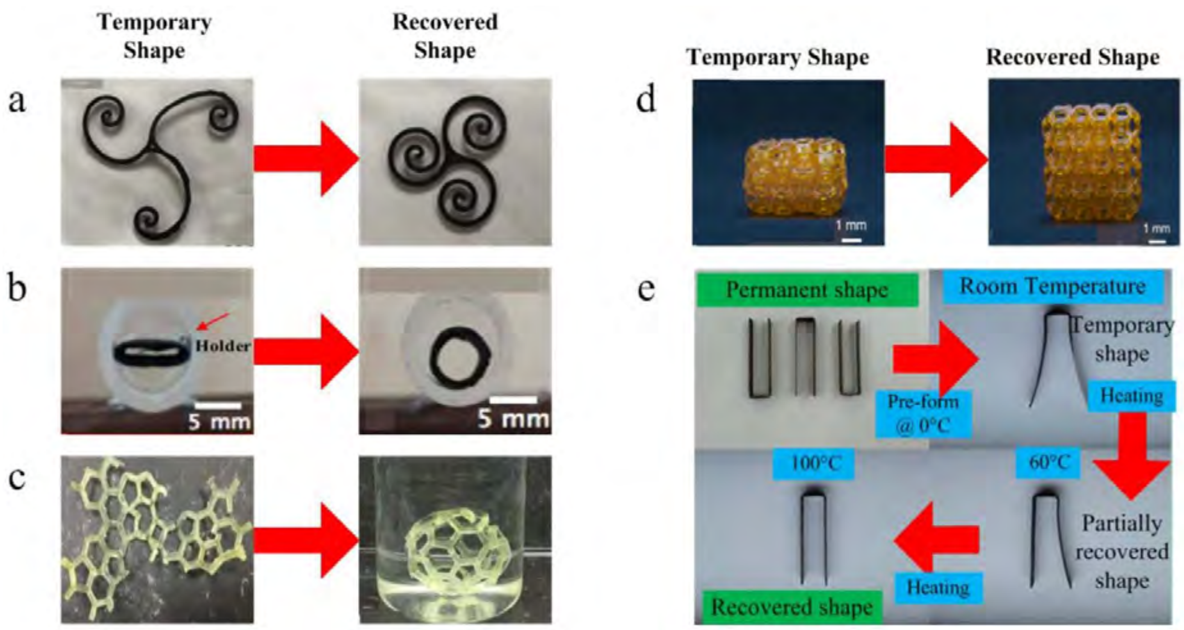

Figure 5. Examples showing the temporary and the recovered shapes of 4D printed structures realized via various printing methods namely (a) FDM (adapted with permission from [48]. Copyright (2019) Elsevier), (b) DIW (adapted with permission from [49]. Copyright (2019) American Chemical Society), (c) SLA (adapted with permission from [50]. Copyright (2017) Elsevier), (d) DLP (adapted with permission from [51]. Copyright (2019) American Chemical Society), and (e) SLM [52].

energy intensity of the laser beam, thin layers of the powder material is either fused or melted together layer by layer until the whole 3D part is formed [57]. Two main powder-based technologies (based on the intensity of energy source) include selective laser sintering (SLS) and selective laser melting (SLM).

2.3.1. Selective laser sintering (SLS). It was developed in the 1980 s at the University of Texas -Austin by Carl Deckard [58]. The method uses laser light to sinter or partially sinter powdered materials layer by layer into a 3D part [59]. Mirrors and lenses are utilized to control the motion of the laser beam and focus it onto the powder bed. The beam raises the temperature on the focused powder material sufficient enough for the neighboring particles to experience necking [57]. To avoid undesirable oxidation effects due to high heat, the fabrication chamber is filled with an inert gas such as argon. Moreover, the powder bed is pre-heated to limit the intensity of laser power required to sintering thus avoiding degradation of the material. The pre-heating also helps to void the warping of the printed part [44]. A wide variety of materials can be used in this method mostly thermoplastic polymer types since moderate energy is required to sinter the particles together. Complex 3D parts can be fabricated, unlike other methods, without additional supports as the un-sintered powder itself acts as supports [60]. The schematic illustration of the SLS set-up is shown in figure 4(e).

2.3.2. Selective laser melting (SLM). The difference between SLS and SLM is that the latter employs high-energy intensity laser beams such that instead of sintering the powder particles it completely melts them [57]. The process is identical to the SLS but because of the high heat involved, SLM is carried out in a more controlled environment. It is a method of choice for fabricating metallic parts as it supports a number of materials including stainless steel, aluminum alloys, titanium, and nickel-base alloys, etc. The main disadvantage of this method is the high residual stresses induced into the printed part because of the high heat involved in the process. However, this may be advantageous for 4D printing as under special conditions the residual stresses may be released leading to a controlled deformation [61]. This was demonstrated by Ma *et al* [52] by fabricating a nickel-titanium shape memory alloy part via the SLM process with spatially controlled deformation courtesy of the differences in thermal history during fabrication (See figure 5(e)).

3. Applications of 4D printing

Although 4D printing emanated from the already established 3D printing technology, it is still in its formative stages of development as it is barely half a decade old. Nevertheless, it has attracted massive interests from researchers all over the world leading to a myriad of both demonstrated and potential applications. The technology is expected to have a significant impact in numerous areas such as biomedical, aerospace, soft-electronics, and consumer appliances.

3.1. Medical applications

Because of the complexity of artificial medical parts in both geometry and expected mechanical behavior in response to the living system's environment, biomedical applications takes a lion's share of 4D printing. The potential areas include smart stents, tissue regeneration, and drug delivery [5].

3.1.1. Smart stents. Stents are diminutive tubular structures that are inserted into the hollow parts of a living system such as blood vessels to serve different purposes. 4D printing technology has enabled fabrication of adaptive stents that can change shape or functionality in response to the body temperature thus alleviating the challenges with the conventional static stents.

Zarek *et al* [62] employed 4D printing to fabricate a thermo-responsive tracheal stent that could be deformed into a temporary shape, inserted into the body, and finally deployed back into its original shape with the increase in the body temperature figure 6(a). It is more advantageous and safe to use compared to conventional stents as complications like stent migration and injuries during insertion could be avoided. In order to mitigate the tracheal collapse (known in medical terms as tracheobronchomalacia), Morrison *et al* [63] fabricated stents via 4D printing that could be implanted around the trachea to both support breathing and the growth of the air passage figure 6(b, i–iii).

Cardiovascular diseases such as high blood pressure due to blockage of the vessels as a result of plaque build-up are some of the most common health problems in the world. Treatment of such conditions has been achieved by using thermo-responsive stents that expand with increasing temperature thus enabling normal blood flow [65]. Wei *et al* [49] incorporated magnetic iron oxide into polylactic acid (PLA)-based ink and employed direct wire fabrication method to 4D print remotely actuated stents for intra-vascular applications as shown in figure 6(c). Bodaghi *et al* [64] employed 4D printing to fabricate self-expanding/shrinking thermo-responsive stents with potential applications in treating blocked blood vessels figure 6(d, i–iii).

3.1.2. 4D scaffolds for tissue engineering. With the advance in biomedical technology, damaged tissues due to injury or trauma can be regenerated with the help of 4D printed scaffolds [66]. As illustrated in figure 7, the main difference between 4D printing and 4D bioprinting is that the latter involves the incorporation of cells intended for growth to repair the damaged tissues into the 4D printed scaffold [67]. Readers interested in more detailed information on the applications of 4D printing for tissue regeneration are directed to the recent comprehensive review by Miao *et al* [68].

Hendrikson *et al* [69] demonstrated the application of4D printed scaffolds for regeneration of tissues in areas such as bones and muscles where the dynamism of the scaffold is required due to the varying activity of the body. The working process of the scaffold is schematically illustrated in figure 8(a). Generally, the materials for fabricating the scaffolds should be both biocompatible and biodegradable to ensure non-rejection by the body and also resorption. Miao *et al* [70] utilized a novel soybean oil epoxidized acrylate to fabricate biocompatible scaffolds via 4D printing figure 8(b, i–ii). The scaffolds exhibited excellent shape memory property and were capable of supporting the growth of human bone marrow stem cells.

3.1.3. Vascular repair devices. Damage to blood vessels may happen during major surgery or injury. The conventional method of sewing up the fracture using biocompatible suture may be time-consuming and the healing process is long. The most desirable properties for 4D printed vascular repair

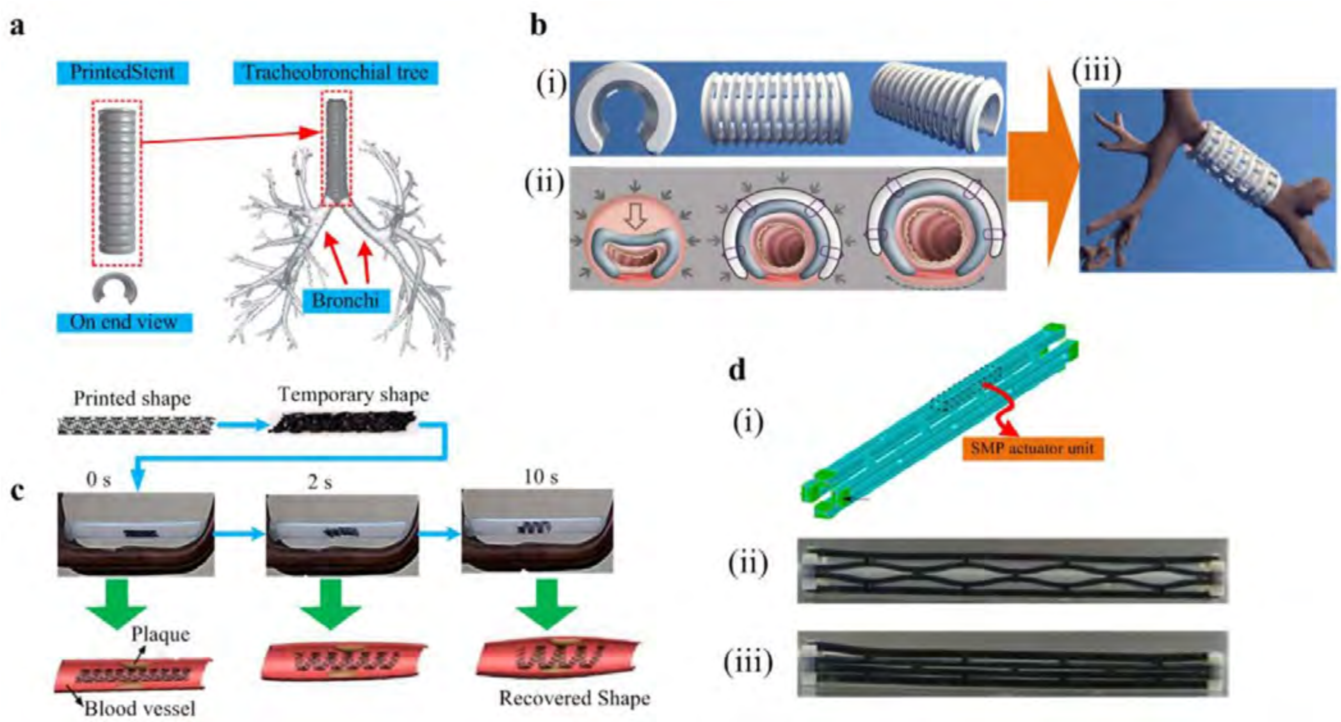

Figure 6. Applications of 4D printed smart stents. (a) Tracheal stent for treating airway diseases (adapted with permission from [62]. Copyright (2016) John Wiley and Sons), (b, i–iii) 3D models of tracheobronchial splints, mechanisms of action in supporting airway growth, and the virtual assessment of the splint over an airway model [63], (c) remotely actuated stents for treating plaque blocked blood vessels (reprinted with permission from [49]. Copyright (2017) American Chemical Society), and (d, i–iii) schematic illustration of the self-expanding tubular stent, its deployed state, and the initial configuration (reprinted with permission from [64]. Copyright (2016) IOP Publishing).

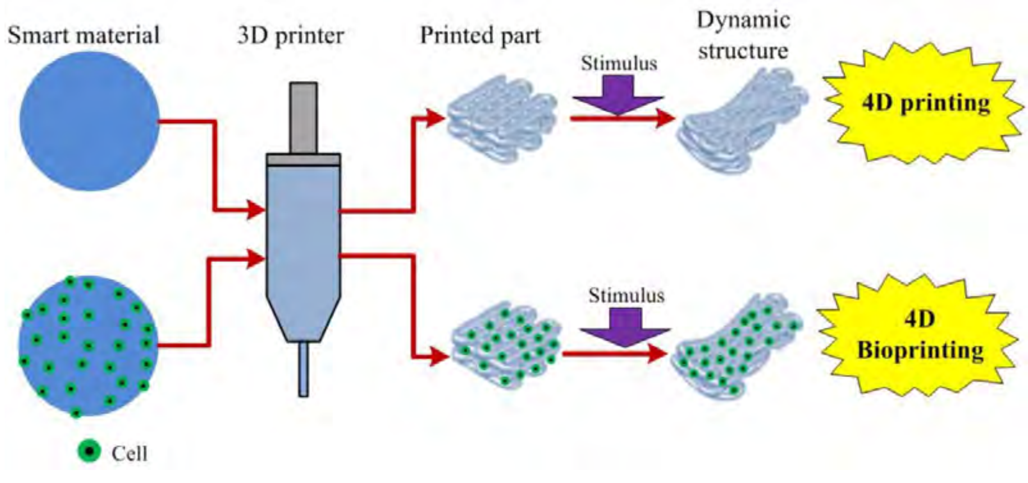

Figure 7. Schematic presentation of the difference between 4D printing and 4D bioprinting.

devices include the self-healing and the shape memory capabilities [51]. Kuang *et al* [72] fabricated a highly stretchable elastomer with shape memory and self-healing capabilities via 4D printing that has the potential to be used for repairing the damaged vessels. As demonstrated in figures 9(a) and (b), the device is programmed to a temporary shape having an outer diameter smaller than the vessel's inner diameter then implanted to the vessel via the damaged area. Upon recovery due to increase in temperature, the device outer diameter closely attaches to the inner surface of the vessel thus reconnecting it. The authors mentioned that the process takes about 3–5 min which is way faster than the conventional methods.

3.1.4. Drug delivery. The expedited healing process can be achieved if drugs were to be delivered in a controlled manner to the intended location in the body and at the required rates. This can be achieved through 4D printed devices as they

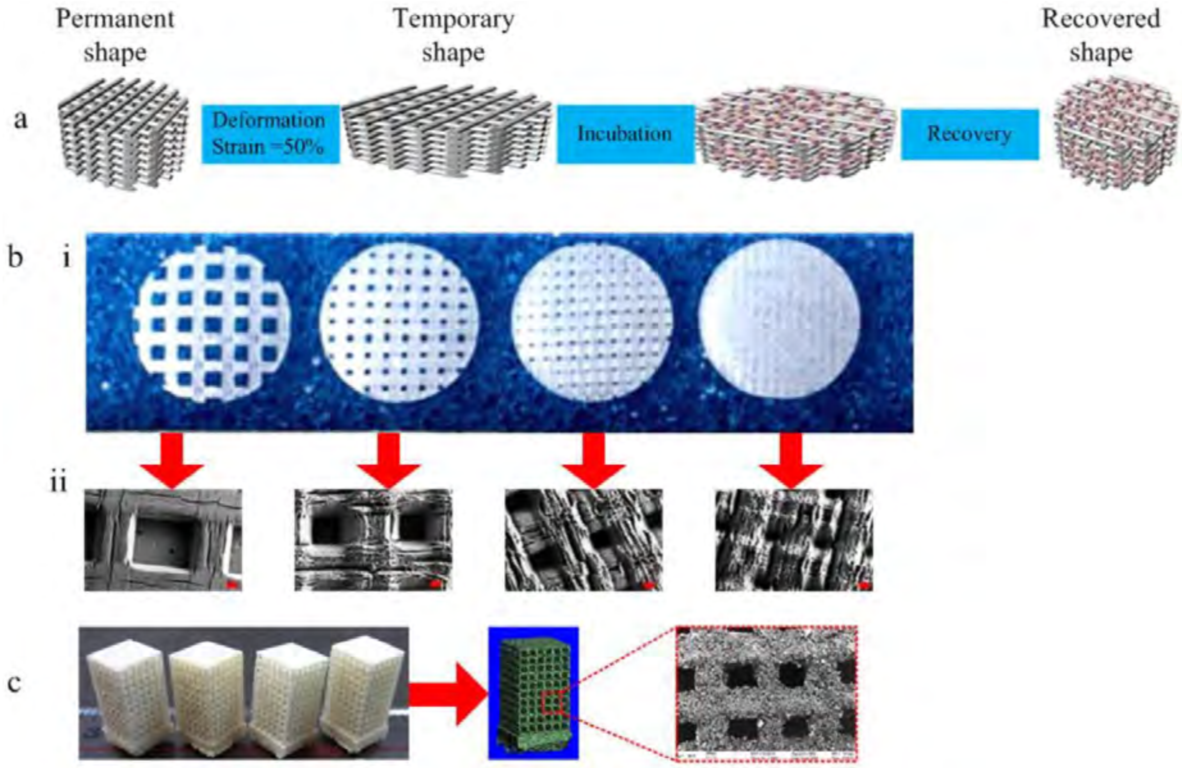

Figure 8. 4D-printed scaffolds for tissue engineering. (a) Schematic illustration of the working principles of 4D scaffolds (reprinted with permission from [69]. Copyright (2017) IOP Publishing), (b, i–ii) digital images of 4D printed scaffolds with variable infill densities and their corresponding scanning electron microscope (SEM) images [70], and (c) scaffolds fabricated via selective laser sintering process showing its micro-computed tomography and scanning electron microscope (SEM) images (reprinted with permission from [71]. Copyright (2010) Elsevier).

can respond to a specific stimulus such as infrared light and open up thus releasing the drugs. Mirani *et al* [73] fabricated a drug-eluting scaffold that can be used for detecting and treating infections in a wound as shown in figure 9(c). It could sense the pH change due to bacterial infection and release the antibiotic agents to the wound.

3.1.5. 4D printed organs for transplant. The demand for organ replacements is ever increasing due to diseases, injury, and even genetic problems. Organ donation is very challenging as getting the donor involves long waits and sometimes gets rejection. 4D printing has the potential to fabricate fully functional human organs for transplant purposes [68, 75].

Kang *et al* [74] designed an integrated tissue-organ printer (ITOP) that can be used to fabricate stable human-scale tissue constructs of any shape. As shown in figure 9(d), they demonstrated its capability by 4D printing human ear cartilage.

3.2. Soft electronics

In contrast to the conventional electronics which are rigid in nature, the current technological trend is shifting towards more human-friendly electronics that are flexible and portable. Indeed, the demand for stretchable and flexible devices that can be flawlessly integrated with flexible surfaces such as human skin is skyrocketing [76]. Such devices not only have the potential of allowing electronics to be intertwined with our everyday lives but could also be indispensable in areas such as health monitoring, artificial skins, and implantable bioelectronics [77]. A recent comprehensive review pertaining to soft electronics has been done by Wang *et al* [78].

Su *et al* [79] demonstrated the application of a 4D printed self-morphing polymer as a soft electronic actuator device. As shown in figure 10(a, i–ii), the two ends of the 4D printed polymer were connected by alumina foil strips to act as conductive electrodes. The bending of the device was triggered when immersed into acetone thus connecting the two ends of the aluminum strips thereby connecting the circuit. Such a 4D printed soft electronic device not only reduces the design complexity but also on the cost. In a similar fashion, Zarek *et al* [80] achieved a flexible electronic temperature sensor by inkjet printing a conductive layer of silver nanoparticles into a 3D printed thermo-responsive SMP. As shown in figure 10(b, i–iii), the temporary shape of the device is set to be an open circuit. When the temperature increases beyond the melting temperature of the SMP, the device returns to its original shape thus closing the circuit and lighting the LED.

Sundaram *et al* [81] developed a method to fabricate self-folding 3D printed composites with embedded electronics. Electrochromic elements were printed within the composite to allow the electrical control of the device through its legs. As shown in figure 10(c, i–iii), voltage application causes the structure to change shape (folding angle). The device can be

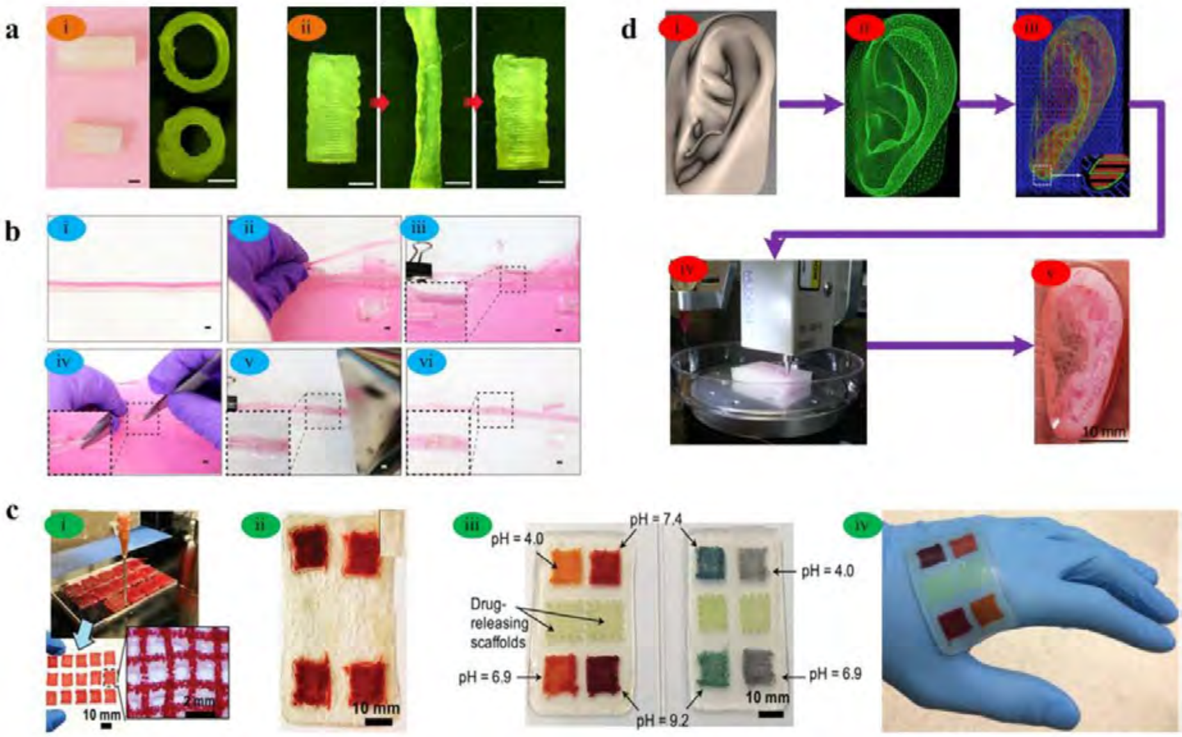

Figure 9. (a, i–ii) digital and optical microscopic images of 3D printed shape memory tubing, (b, i–iv) demonstration of its application for rapid vascular repair and reconnection (a and b adapted with permission from [72]. Copyright (2018) American Chemical Society), (c, i–iv) the fabrication of an advanced multifunctional dressing and the demonstration of its application for drug delivery and wound monitoring (reprinted with permission from [73]. Copyright (2017) John Wiley and Sons), and (d, i–iv) 4D printing of a human ear cartilage (reprinted with permission from [74]. Copyright (2016) Springer Nature).

applied in sensing applications. Muth *et al* [82] reported on embedded three-dimensional printing (e-3DP) for fabricating highly stretchable elastomeric matrices. In this method, conductive ink is printed unto an uncured elastomeric reservoir which will then form a resistive sensing element resulting to a flexible strain sensor. The electrical resistance of the device will change as a function of time with changing strains in response to different positions of the hand.

3.3. Active origami structures

Origami is a decades-old tradition associated with Japanese culture that has spread all over the world. Basically, it involves the folding of a flat piece of material mostly a paper into a complicated three-dimensional structure. In recent years, origami has attracted increasing interests from engineers in that the process can be parameterized and applied to develop new structures and devices that can provide technological solutions to engineering problems. Possibilities include saving on the storage volume by packing large objects into small volumes that can be deployed for use such as solar arrays for space applications [15].

Ge *et al* [83] fabricated active origami structures via 4D printing using printed active composites (PAC) as the hinge. As shown in figure 11(a), the printed flat shape assembles into the desired 3D box upon the application of a stimulus (heating). Almost similar kind of work was presented by Mao *et al* [84]. They fabricated a sequentially self-folding structure that turns into a box upon the application of thermal energy. The flat structure composed of SMPs with disparate shape memory behaviors which consequently facilitated the time-dependent behavior of each polymer with an increase in temperature leading to the sequential activation. The idea of sequential folding of a flat structure into a box was further demonstrated by Yuan *et al* [85]. Unlike in the previously mentioned work, they achieved the sequential folding by applying a current through the conductive wires embedded into the flat structure. The activation process is shown in figure 11(b). It is worth noting that the folded box was reported to maintain the shape as long as the electrical circuit remained closed and would return to its initial flat shape upon disconnection of the circuit.

Liu *et al* [86] fabricated a Miura-origami structure via 4D printing then investigated its recovery force when subjected to compressive loading. They reported shape recovery ratios of more than 94% and volume changes of up to 289%. The folding and the unfolding process of the 4D printed structure is shown in figure 11(c).

3.4. Morphing structures

Morphing structures have the ability to change their geometric parameters in response to environmental conditions such as temperature or humidity [87]. As such, they have a wide variety of potential applications especially in the

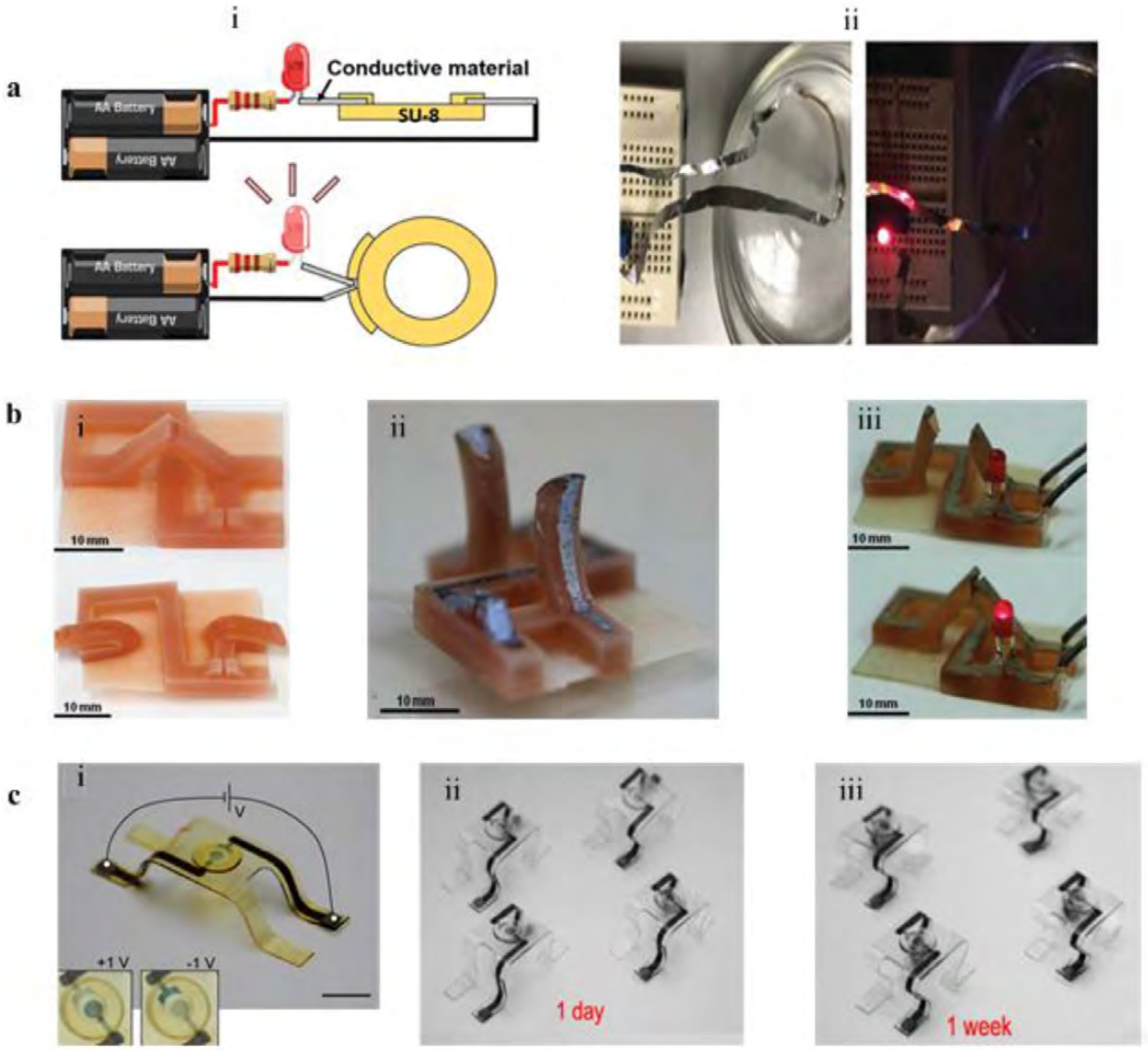

Figure 10. Applications of 4D printing in soft electronics. (a, i–ii) schematic of a soft-actuator that acts as a relay for a LED light and its experimental implementation (reprinted with permission from [79]. Copyright (2017) Royal Society of Chemistry), (b, i–iii) 4D printed temperature sensor (reprinted with permission from [80]. Copyright (2015) John Wiley and Sons), and (c, i–iii) photographs of 3D printed self-folding electronics and their change in folding angles over the course of one week [81].

aerospace industry [88, 89]. A growing number of researchers are directing focus on utilizing 4D printing to achieve morphing structures.

Wang *et al* [90] reported on morphing composites embedded with continuous fibers accomplished via 4D printing technology figure 12(a). The difference in the coefficient of thermal expansion (CTE) between the fibers and the matrix facilitated the deformation of the composite. In its shape morphing, the size and the direction of the principal curvature were determined by the angle between the intersecting fibers and the bisector of the angle respectively. Ding *et al* [20] employed direct 4D printing to fabricate composite structures that could transform into different shapes upon the application of a stimulus (heat). As shown in figure 12(b), flat and ring-shaped printed composites transform into wavy-shaped structures when the heat is applied. The trick is to alternate both the elastomer and SMP segments along the lengths of the printed composite strips with a prescribed period that can result in the wavy structure when activated by a stimulus. Both the pitch and the angle of the pattern including the thickness ratio of the elastomer and the SMP determine the geometry of the resulting activated structure. As a concept that can be applied to aircraft wings, Yuan *et al* [85] demonstrated the synergistic action of multiple laminated hinges in a wing-like structure to

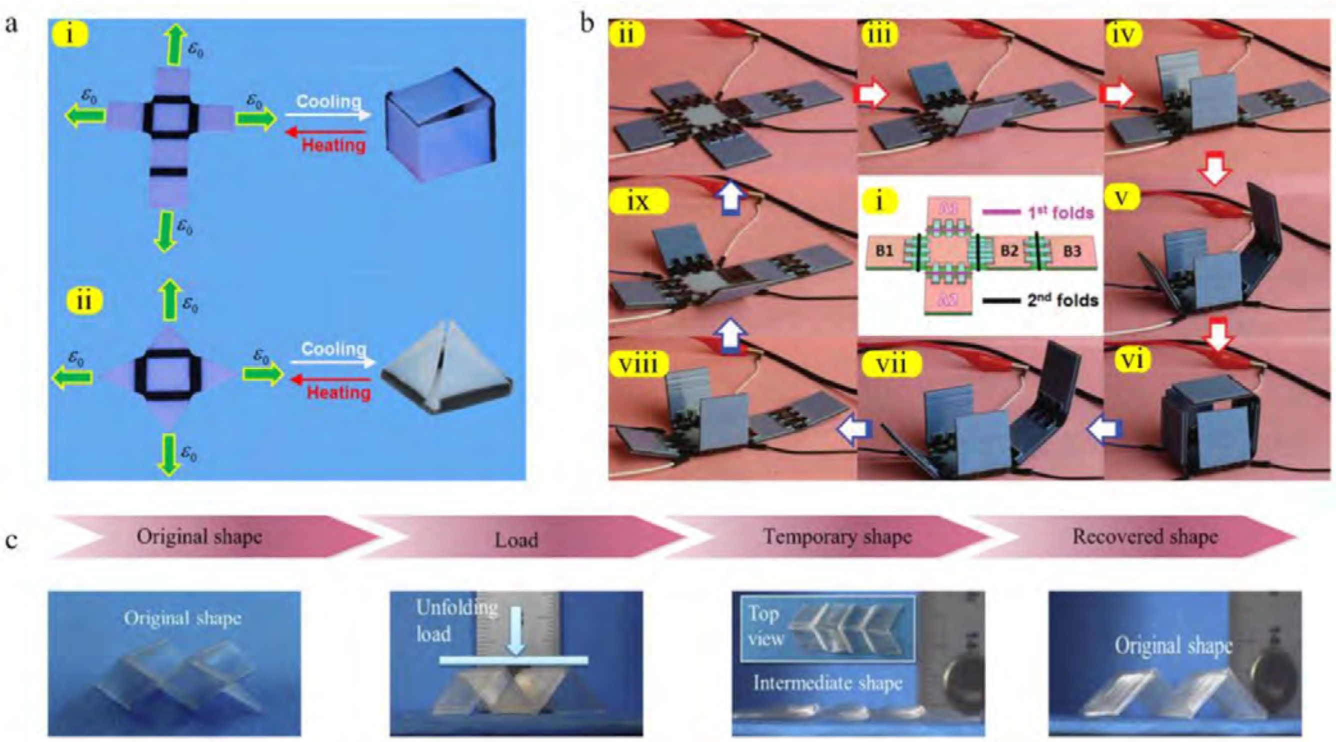

Figure 11. Some of the 4D-printed active Origami structures. (a, i–ii) two-dimensional flat cross and star shapes and assembles into a three-dimensional box and a pyramid respectively upon heating (reprinted with permission from [83]. Copyright (2014) IOP Publishing), (b, i–ix) schematic of the design of sequential folding box and the snapshots of its folding and unfolding process (reprinted with permission from [85]. Copyright (2017) Royal Society of Chemistry), and (c) photographs of the recovery process of a 4D-printed Miura-origami structure after a compressive loading (adapted with permission from [86]. Copyright (2018) Elsevier).

facilitate morphing. Figure 12(c) shows the snapshots of the action and the subsequent de-activation of the soft morphing airplane.

3.5. Smart grippers

Devices that have the ability to grab or release objects can be applied in a number of applications such as drug delivery [91]. By utilizing multi-material SMPs and a high-resolution projection microstereolithography, Ge *et al* [65] achieved smart grippers and demonstrated their ability to grab and drop an object as shown in figures 13(a) and (b). Akbari *et al* [92] exploited the same technique of multi-material 3D printing to fabricate a soft gripper that could grasp a 15 g cylindrical object as shown in figure 13(c). 4D printing provides a simple and straightforward approach to fabricating object gripping devices compared to the conventional methods.

3.6. Dynamic fashionwear products

Potential applications of 4D printing extend beyond engineering and technological needs to fashion wear and consumer products. There has been growing interest from leading fashionwear firms to utilize the technology in developed revolutionary products. As Helmore [93] reports, 4D printed dresses are expected to have a huge impact in the near future. Such dresses are self-adaptive to environmental changes such as temperature or humidity adjusting to the desired shape without tensile loading thus giving the wearer more comfort [94]. The top shoe company, Adidas is already employing 4D printing to fabricate futuristic sports shoes which are light and perfectly tailored for the person wearing them [95].

With thermally-responsive SMPs, Zarek *et al* [96] utilized 4D printing to fabricate dynamic objects. As shown in figures 14(a), (b), they demonstrated their application as jewelry (smart ring) and shoe accessory (smart heel). Yang *et al* [97] fabricated sunflower-like structures based on shape memory composites that could change from closed to opened state upon heating as shown in figure 14(c). Almost a similar kind of structure was demonstrated by Ding *et al* [20] except that their flower-like structure consisted of multiple petals at different layers. The layers could assume different curvatures upon heating hence a different configuration of the structure as shown in figure 14(d).

4. Current challenges and future perspective

Whereas 4D printing technology is considered to be relatively new (just half a decade old), it has its foundations on the very well established 3D printing technology (over three decades old). Like any other new technology with some known foundation, 4D printing brings in new challenges of its own on top of the already explored inherent challenges of 3D printing.

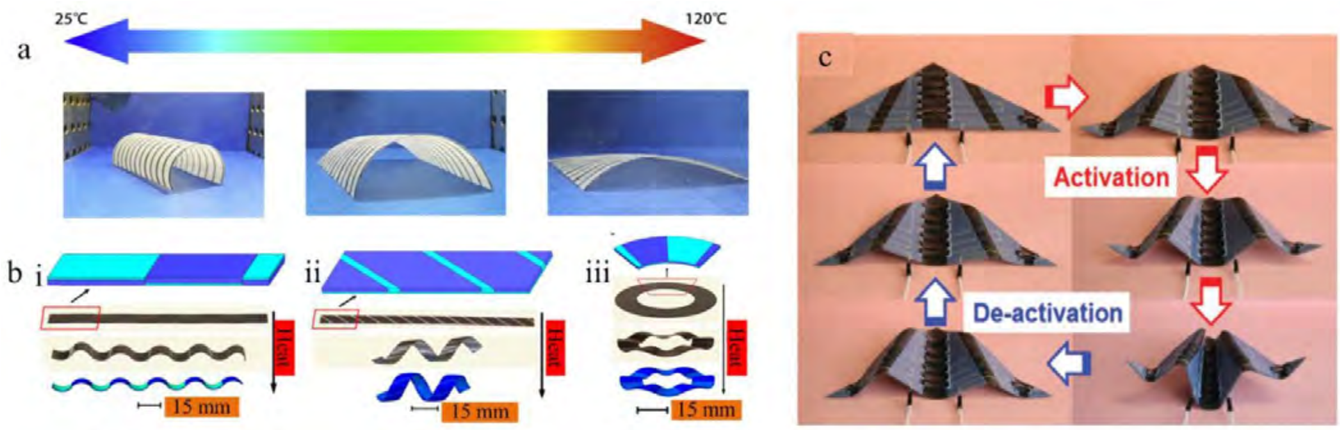

Figure 12. Applications of 4D printing on morphing structures. (a) Morphing structures with embedded continuous fibers under the influence of temperature (reprinted with permission from [90]. Copyright (2018) Elsevier), (b, i–iii) structural elements with various deformation modes such as the transformation of a flat plate into a wavy and helix structure [20], and (c) snapshots of activation and de-activation of a morphing airplane model (reprinted with permission from [85]. Copyright (2017) Royal Society of Chemistry).

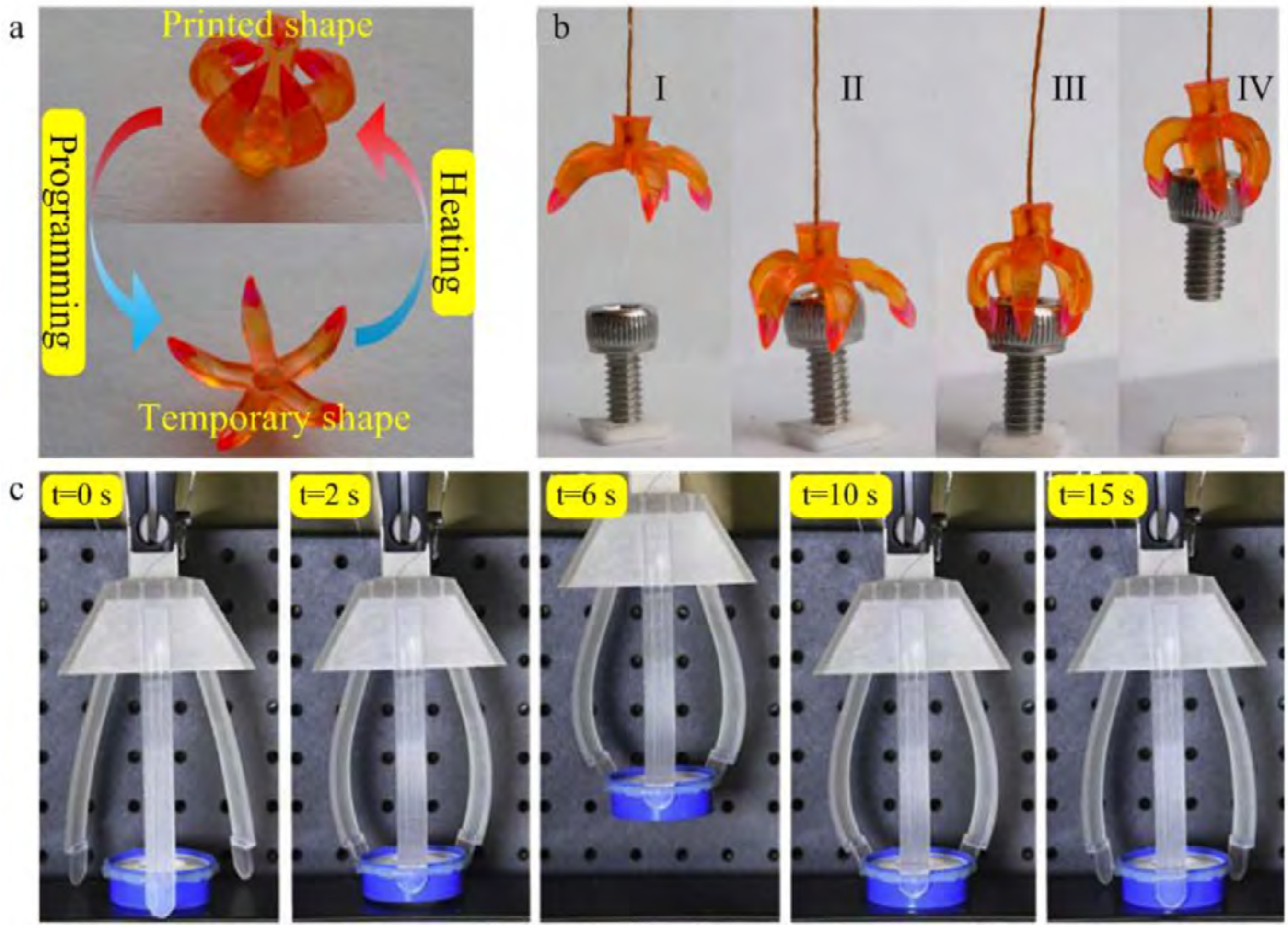

Figure 13. 4D printed multi-material smart grippers. (a) Photographs of the fabricated smart gripper showing the transition from the temporary to the permanent shape and vice versa, (b, i–iv) snapshots showing the process of gripping an object [65], and (c) snapshots showing the process of grasping and releasing of a 15 g cylindrical object by the smart gripper in a span of 15 s (reprinted with permission from [92]. Copyright (2019) Elsevier).

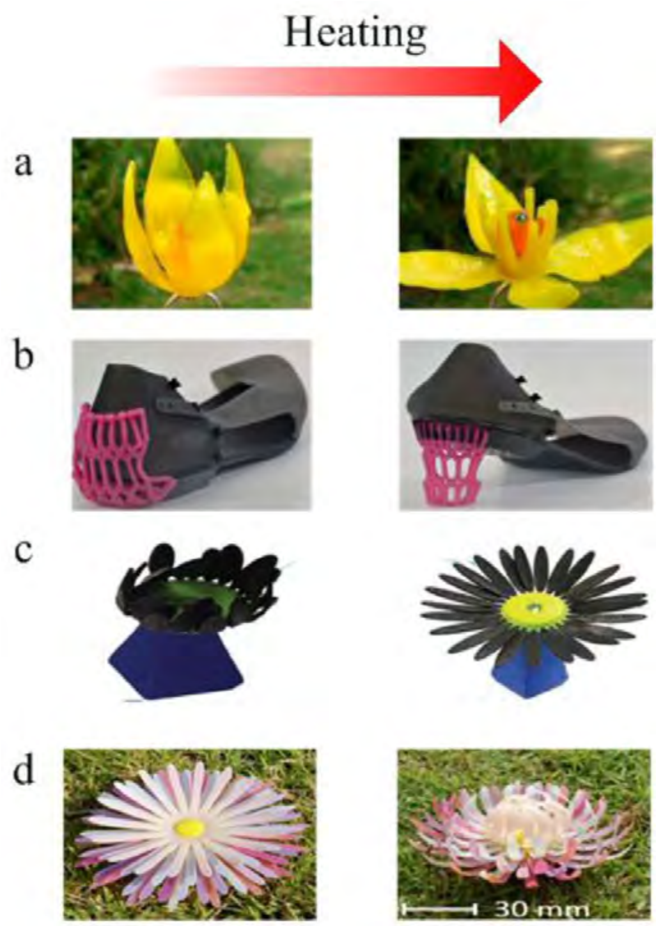

Figure 14. Examples of fashionwear products realized via 4D printing. (a) Photographs of the flower-themed ring that changes shape with temperature, (b) dynamic heel attachment showing its temporary and permanent shapes (adapted with permission from [96]. Copyright (2016) Taylor and Francis), (c) sunflower-like structure responding to stimulus by changing from closed to opened state [97], and (d) printed flower with multiple petals assuming final configurations with different curvatures upon heating (adapted with permission from [20] Copyright (2017) John Wiley and Sons).

Most 3D printing technologies have low resolution and thus affect the overall quality and precision of the printed components [41]. For instance, poly-jet printers which are typically used for multi-material 4D printing have a resolution of few tens of micrometers which might not be good for some applications such as medical devices. High resolution is desired since it leads to good surface finish. Resolution and printing speed go hand in hand. An attempt to improve the resolution by employing a small nozzle size leads to slower printing speed.

There is also a challenge to the stringent requirements of the raw materials for 3D printing. For instance; 3D printing technologies suitable for 4D printing such as FDM, SLM, or SLA require the raw material to be in filament, powder, and liquid respectively. This greatly limits the variety of smart materials to be explored for 4D printing [2]. It also means that the introduction of new material types may require designing of new printing hardware. On top of that, the use of laser light to cure the photopolymer or melt the powder may lead to degrading the material due to high heat thus affecting the durability of the part [98].

All the additive manufacturing techniques form the 3D part in a layer by layer manner. This poses a challenge to the overall mechanical properties of the printed part as it depends on the adhesion between layers [99]. More research, therefore, should focus on ways of improving the interlayer adhesion. Enhanced adhesion of the layers translates to better mechanical properties which are good for the development of 4D printing technology.

Stimulus makes it possible to achieve the 4th dimension of a 3D printed part. Most currently available smart materials respond to one type of stimulus [100]. This limits the applicability of the 4D printed parts. For instance, 4D printed devices destined for biomedical applications are expected to interact with a complicated network of tissues experiencing multiple stimuli [67]. Therefore, it is desirable for these materials to be able to respond to such stimuli present in human bodies. Moreover, the glass transition temperature for thermally activated 4D printed parts has been reported to be too high for the human body temperature [101]. This affects the recovery of the part in the body and, therefore, more research ought to be done concerning materials with lower activation temperatures.

Research in the modern era is seldom accomplished without numerical simulation using commercial finite element codes such as ABAQUS. Simulation tools that can accurately predict the behavior of the 4D printed parts under various stimuli need further development.

In summary, the current challenges associated with 4D printing technology can be categorized based on four areas namely; technological limitations of the printing hardware, smart material, simulation tools, and design of the 4D structure. Despite all these, 4D printing has been hailed as one of the revolutionary inventions in the recent past. It is envisaged to have a significant impact on the manufacturing of dynamic structures for various applications. As the research advances, we expect that most of the challenges mentioned will be addressed. Particularly, the varieties of smart materials that are 4D printable are expected to increase. It is worth noting that one of the most active researches being undertaken is in the search for smart materials with tunable properties that can be tailored for specific purposes. A direct result of such research will be the surge of 4D printing applications. These will range from complicated engineering and technological applications such as self-deployable space structures to simple household applications such as smart clothing or wearable devices.

5. Conclusions

Emerging from the already established additive manufacturing technologies, 4D printing inherits the highly attractive flexibility and versatility in fabricating parts with super complicated geometries. On top of that, it adds yet another fascinating and admirable trait of fabricated components; the ability to change functionality or shape when a change in a specific stimulus such as temperature is detected. While it is still an upcoming technology, the mentioned traits project it to be the future indispensable manufacturing technology for dynamic and geometric-complicated structures.

Recognizing the impact of 4D printing in academia and industry, this review work has sought to present a brief research summary with more focus on the current 4D printing methods. After a brief introduction on the main research directions in AM and the difference between 3D and 4D printing, we elucidated on the 4D printing process from the design, manufacturing to post-processing. Furthermore, we expounded the AM methods that have been demonstrated to accomplish 4D printing whilst giving examples from the literature. Moreover, we have enumerated the emerging applications in areas including medical, soft electronics, Origami structures, morphing structures, and fashionwear products.

The advance and eventual ubiquity of 4D printing technology rely on the progress in other areas such as 3D printing hardware, smart materials, mathematical modeling, simulation software, and stimulus methods. Specifically, printing hardware must possess multi-material printing capabilities. For high-quality surface finish and overall mechanical properties of a complex 4D printed part, printer resolution plays a significant role. The advance in the science and engineering of smart materials is a recipe for extending the horizons of 4D printing. That means materials that are more versatile with enhanced properties that are tunable for specific functions can be realized. Like most engineering problems, 4D printing utilizes mathematical models to predict the behavior of the designed component thus ensuring optimization of the part to the desired behavior before production. Together with the simulations software advancement, mathematical models will accelerate the developments in 4D printing.

Various research groups and funding institutions are spending a fortune to expedite the advancement of 4D printing technology. With these accelerated researches, we expect the various challenges mentioned to be overcome in the near future leading to more intriguing applications. It is our hope that the brief review presented herein inspires more interest and research towards the success of 4D printing technology.

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Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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