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# A Review of 4D Printing Technology and Future Trends

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## *Abstract*

**3D printing has gained immerse popularity since its introduction and finds application in areas such as prototyping, engineering and medical field largely due to its advantage of being able to quickly and inexpensively transform computer 3D files into physical objects. 3D printing has the capability of printing geometrically fixed structures which are static and not suited for multifunctional use. 4D printing was developed when researchers combined smart materials and 3D printing. 4D printing uses the fourth dimension of time to create shape morphing 3D printed objects when exposed to stimuli after using conversional 3D printing technology such as fused deposition modelling (FDM) and selective laser sintering (SLS). 4D printed materials respond to stimuli such as pH, humidity and temperature to activate the 3D printed components without the use of electronics or motors. There has been a lot of research done on smart materials capable of sensing external stimuli and responding to it. In this paper 4D printing is reviewed according to activation stimuli and the uses of this technology are explored. 4D printing has the prospective to simplify the design and manufacturing of different products and has the vast potential to create parts that self-actuate to react to their environment. Applications of 4D printing are in areas such as biomedical devices, security, fabrication of patterned surfaces for optics and structures with multi directional properties.**

*Keywords*: 4D Printing; activation; smart mterials; stimuli

# 1. Introduction

3D printing is a well-known additive manufacturing technology that allows researchers, manufacturers, and private users to fabricate custom 3D objects using computer software such as computer aided design (CAD) [1]. Due to the highly customizable nature of 3D printing it has found use in a number of fields such as fabrication of fashion jewellery [2], polymer printed textiles [3], supercapacitors, mechanical metamaterials and sensors, bio-hybrid robotics [4] and tissue scaffolds. Several additive manufacturing technologies have been developed for processing pure polymers and polymer nanocomposites [5] such as stereolithographic (SL), digital light projection (DLP), direct inkjet and extrusion-based printing as well as liquid deposition modelling (LDM). They enable less expensive free form fabrication of complex, customized and multi-scale 3D geometries for application in a vast range of fields, from tissue engineering scaffolds [6]to strain and skin like sensors.

The introduction of smart materials which are responsive to external stimuli has found use in shape recovery, sensors and actuators [7]. 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, dependent on environmental stimuli. Shape morphing systems can be found in many areas including smart textiles [8], autonomous robotics, biomedical devices, drug delivery and tissue engineering. The natural analogues of such systems are exemplified by nastic plant motions, where a variety of organs such as tendrils, bracts, leaves and flowers respond to environmental stimuli such as humidity, light or touch by carrying internal turgor, which leads to dynamic conformations governed by tissue composition and microstructural anisotropy of cell walls. 4D printing is inspired by these botanical systems. 4D printing has the economic, environmental, geopolitical, and strategic implications of additive manufacturing while providing new and unprecedented capabilities in transforming digital information of the virtual world into physical objects of the material world. The fourth dimension in 4D printing refers to the ability for material objects to change form and function after they are produced, thereby intelligent materials become a key issue in this technology. This paper reviews the development and capabilities of the 4D printing technology and investigates its applications and suggests its future impact.

# 2.0 Fundamentals of 4D printing

3D printing technology has been used to make static structures from digital data in 3D coordinates, 4 D printing adds the concept of change in the printed configuration over time, dependent on environmental stimuli. The key difference between 3D and 4D printing are the smart design and smart materials as 4D printed structures may transform in shape or function. This implies that the 4D printed structures should be fully programmed in detail by accounting for any anticipated time-dependent deformation of the object [9]. 4D printing was firstly introduced by a research group of Massachusetts Institute of Technology (MIT) and defined as the fabrication of 3D printed structures with adaptable and programmable shapes, properties or functionality as a function of time. [10]

Intelligent materials are able to sense stimulus from the external environment and create a useful response. Thus, intelligent materials can be seen as those which provide a means of achieving an active intelligent response in a product that would otherwise be lacking and have the potential to yield a multitude of enhanced capabilities and functionalities [11].

Three key aspects must be fulfilled for 4D printing to take place. The first is the use of stimuli responsive composite materials that are blended or incorporate multi-materials with varying properties being sandwiched layer upon layer. The second is the stimuli that will act on the object causing it to animate. Examples of these stimuli include heating, cooling, gravity, ultraviolet (UV) light, magnetic energy, wind, water or even humidity. The last aspect is time for the simulation to occur, and the final result is the change of state of the object [12].

**2.1 Smart Materials**

Smart materials are essential to the development of 4D printing research. Smart materials don't necessarily have to possess the ability to change shape. Materials that have the ability to change colour, hardness or transparency are also important in camouflage technology, signalling, detecting foreign substances and biomedical applications [13]. The time dependent change in shape, property or functionality can be triggered by different type of stimuli. Gladman et al used water to activate 4D printed water-sensitive biomimetic structures inspired by nature [14]. Other stimuli can also be utilized in 4D printing such as heat [15], pH [16], a combination of heat and water as well as of heat and light. The complex structures may evolve several orders in magnitude of length scale through controlled buckling, pneumatically transformation, controlled creasing, photo-induced folding, stress induced curing, thermal controlled swelling, and use of shape memory composites [17].

Table 1.0 Typical smart materials used in 4D printing [18]

| Material | Input/Stimulus | Output/Response | Application |
|---|---|---|---|
| Polymeric gal | pH change | Swelling or contracting | Artificial muscle |
| Electro-rheological fluid | Electrical signal | Viscosity change | Torsional steering system damper |
| Pyroelectric material | Temperature | Electric signal | Personnel sensor (open supermarket door) |
| Polymer (e.g thin film cellulose, ceramic) | Humidity change | Capacity/resistance change | Humidity sensor |
| Self-healing materials | Force | Force | Smartphone chassis |
| Smart metal alloys | Temperature | Shape | Motor actuators |
| Dielectric elastomers | Voltage | Strain | Robotics |
| Ceramics (e.g Ladoped $BaTiO_3$) | Current (or template) | Resistance | Thermistor/overcurrent protector |
| Varistor (e.g Bidoped ZnO) | Voltage | Resistance | Surge protector |
| Piezoelectric material | Deformation/strain electric signal | Electric signal | Active noise control devices, pressure and vibration sensitizing |

The materials shown in table 1.0 can be exploited and used in 3d printing as smart materials to yield objects that can morph over time giving 4D printed objects.

## 3.0 Motivations

4D printing opens new fields for application in which a structure can be activated for self-assembly, reconfiguration, and replication through environmental free energies. There are several advantages brought about by this technology such as significant volume reduction for storage, and transformations that can be achieved with a flat pack 4D printed structures. Another example is instead of directly creating complex structures using 3D printing, simple components from smart materials can be 3D printed first and then self-assembled to reach the final complex shape [19]. The potential applications of 4D printing can be broadly classified into three main categories which include self-assembly, multi-functionality and self-repair. The ability of 4D printed structures to self-assemble and self-repair opens new opportunities of application, such as the fabrication of minimally invasive surgery devices that can be placed in human body through a little surgical incision and then assembled at the required position for surgical operations [20].

### 3.1 Self assembly

A future application can be on a large scale and in a harsh environment. Individual parts can be printed with small 3D printers and then self-assembled into larger structures, such as space antennae and satellites [21]. This capability can be exploited for the creation of transportation systems for complex parts to the International space station. Further applications include self-assembling buildings, this is especially useful in war zones or in outer space where the elements can come together to give a fully formed building with minimum work force [22]. There is also the added advantage that some limitations in construction can be eliminated by the use of 4D printing. Rigid materials can be can be 3D printed along with smart materials to create specific areas of a part that act as joints and hinges for bending. Raviv et al argue that construction must be made smarter and solve problems of wasting large amounts of energy, materials, money and time for building. These issues can be solved using design programs and software to embed information into the materials that makes the material and construction more accurate [23]. Self-assembly may not be efficient for every purpose, which implies different sectors and applications must be identified that benefit most from self-assembly [24].

**3.2 Self adaptability**

4D printing allows the integration of sensing and actuation directly into a material rendering external electromechanically systems unnecessary. This decreases the number of parts in a structure, assembly time, material and energy costs as well as the number of failure prone devices, which is associated with electromechanical systems [10]. This technology is finding use in self-adaptive 4D printed tissues and 4D printed personalized medical devices such as tracheal stems.

**3.3 Self Repair**

The error correct and self-repairing capability of 4D manufactured products show tremendous advantages with regard to reusability and recycling. Self-healing pipes and self-healing hydrogels are some of the potential applications of 4D printing [22]. Self-healing of polymers can be achieved by a few categories of reactions, which include covalent bonding, supramolecular chemistry, H-bonding, ionic interactions, and π-π stacking [25]. Self-healing materials have also been shown to have great potential for producing soft actuators with enhanced durability, due to their ability to self-repair damage ranging from bulk cracks to surface scratches [26]. The use of self-healing hydrogels as inks for additive manufacturing has been successfully demonstrated [27].

**3.4 Piezoelectric materials**

Those materials capable of generating electric charge in response to applied mechanical stress are piezoelectric materials. Not all smart materials exhibit shape change but they may have other significant changes in their properties such as electro-magneto rheological fluids. Piezoelectric materials are smart materials which accumulate electric charge across the material surface upon mechanical stress and this charge can change the shape of the material.

## 4.0 Shape Memory Effect

After being quasi-plastically deformed in the presence of the right stimulus a Shape memory material (SMM) is able to recover its original shape [28]. This phenomenon is termed the Shape memory effect (SME). Shape memory polymers (SMP) are able to mechanically transit between the printed shape and a predetermined shape in response to external stimuli. Xie et al printed a shape memory polymer through the light curing process, which evolves in structure when immersed in water. Hydrogels are an interesting area for 4D printing. Hydrogels are generally 3D networks of polymer chains formed by crosslinking. The crosslinks may either be temporary or permanent by forming physical contacts or covalent bonds. These networks tend to swell when immersed in water due to the network gaps. The hydrogels are biocompatible and useful for biological applications and their ability to swell makes them a candidate for 4D printing. The swelling can be controlled by crosslink site density, surface modification, and size of the hydrogel particles. Raviv et al synthesised a hydrogel by mixing vinyl caprolactam, polyethylene and diacrylae oligomer which can expand by approximately 200% under water [29].

There are a few notable drawbacks that may need to be overcome for practical application of shape changing hydrogels. Their response speed is low to stimuli especially so for large objects because the swelling mechanism is intrinsically limited by mass diffusion. The actuated shape may not be stable due to the volatility of water and most hydrogels are relatively brittle and may undergo mass loss during the hydration/dehydration cycle. These challenges may be overcome by introducing porosity into the hydrogels significantly improving the response speed and innovative molecular designs such as double network can lead to robust mechanical properties.

SMP are an alternative to hydrogels for fabricating morphing structure which are more robust and provide a wider range of mechanical properties. The essential programmability of the shape shifting pathways distinguishes SMPs from hydrogels. During the programming process, a temporary shape as defined by an external deformation force can be fixed and the shape shifting pathway goes from this temporary shape to the synthesized or fabricated original shape. As the original shape can be fixed into unlimited number of temporary shapes by variation of the external deformation force, the corresponding shape shifting pathway is also unlimited. This is different from hydrogels whose shape shifting is typically non-programmable, implying they can only morph between two geometric shapes defined in their fabrication no matter how sophisticated they are [30].

A research group at Massachusetts Institute of Technology (MIT) has 3D printed multi-materials that change shape under water. Two different materials with different porosities and water absorption capacities to print biomaterial structures. These structures have a porous water absorbing material on one side and a rigid waterproof material on the opposite side. When the printed object is immersed in water there is water absorption on both sides. The side which absorbed water increases in volume whereas the opposite side remains unchanged resulting in a bending motion towards the rigid side as shown in Fig 1.



Fig 1: Transformation of a structure from 1D to 3D with water absorption materials printed by Massachusetts Institute of Technology [31]

Ge et al developed a model that took into account different design options to fabricate composite hinge structures. Their research focused on characterizing hinge behaviour with respect to hinge bending angle as a function of geometrical parameters, thermomechanical loading parameters and programming parameters [32]. Ge et al reported a paradigm of 4D printing to create printed active composites directly printing shape memory polymer fibres in an elastomeric matrix to enable shape change of the composite. SMPs could recover their original shape and size when heated above their glass transition temperatures. They experimentally proved this by producing the folding box shown in Fig 2 and also 4D printed a pyramid and an origami airplane [33]. Tibbits and his colleagues experimentally demonstrated how 4D printed objects could perform self-assemblies. A strip of hydrophilic polymer that expands by 150% when it encounters water was printed over a corresponding strip of rigid material, which causes the primitive to fold.

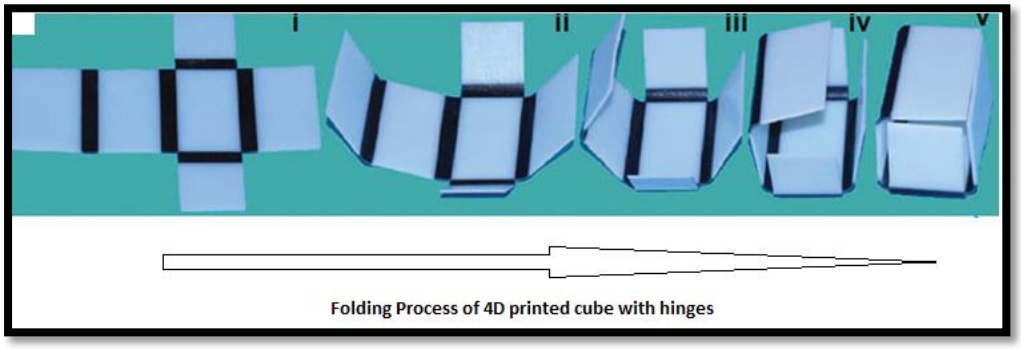



Fig 2: Folding process of 4D printed cube with hinges [32]

Yang et al exploited thermoplastic shape memory polyurethane and a Fused Deposition Model (FDM) printer to fabricate 3D structures. Carbon black was introduced into the resin as a filler, yielding 3D morphing structures that can be triggered by the photo-thermal mechanisms as shown in Fig 3 [34]. Thermoplastics generally have poor performance over their thermoset counterparts, which limit their application potential. Traditional thermoset SMPs are chemically cross-linked and not melt process able once the crosslinks are established in the polymer network.

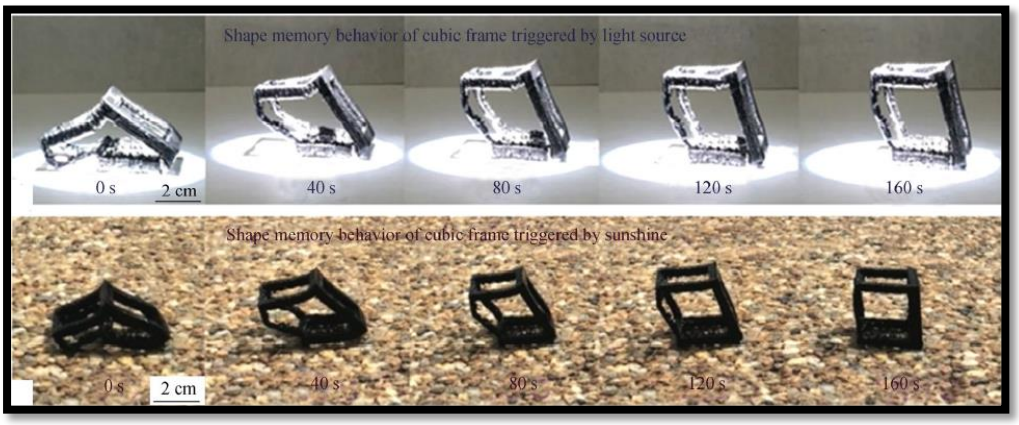



Fig 3: Photo responsive materials based on carbon black and polyurethane extruded from a FDM printer to form a 3D printed object with photo responsive shape memory effect. The shape recovery of cubic frame under 87mW/cm$^2$ of light source [34]

Figure 4 shows the printing of a 3D printed structure with multiple SMPs. Multi-material grippers that have the potential to function as micro grippers [35] that can grab objects or drug delivery devices [36], [37] and the release the objects. Fig 4 shows different sized multi-material grippers with different designs. Fig 4b shows the 3D printed gripper as printed in its open state and functionality if grabbing objects is triggered upon heating. Fig 4c shows a time lapse image of the gripper grabbing an object. By controlling the dynamic properties of the different SMPs it is possible to design the time dependent sequential shape recovery [38].

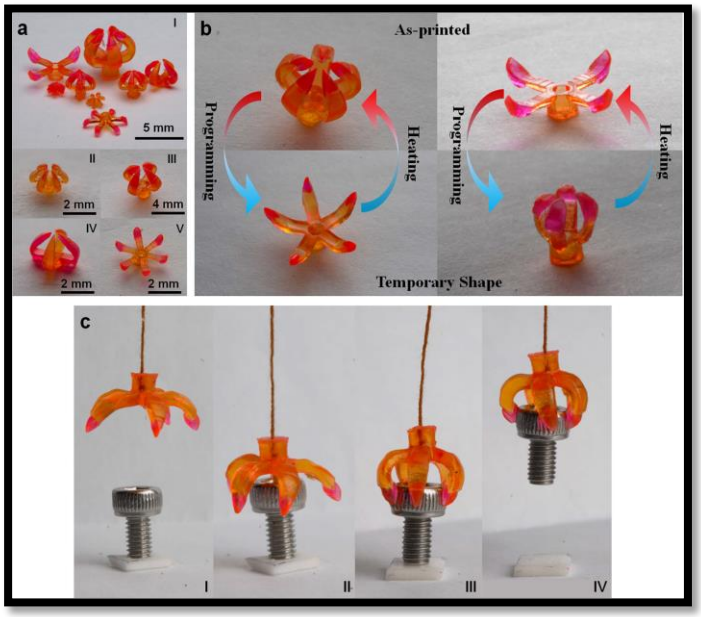



Fig 4: 4D printed griper [38]

Recently methacrylated macronomers for 3D printing of shape memory objects were synthesized by Zarek et al. These materials were used in flexible electrical circuits as shown in Fig 5 which was an open electric circuit [39]. When heated above the $T_m$ the circuit was closed and lit an LED.

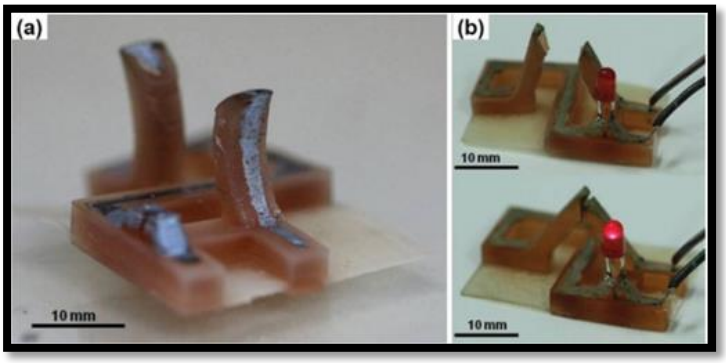



Fig 5: Shape memory based electrical device a) conductive ink printed on the shape memory construct b) fabricated temperature sensor in its off state (top) and on state (bottom) [40]

They used a heated bath for the photopolymer where a projection Sintering Laser (SL) process was used to create the structures and inkjet printing was used to print the conductive inks. Basing on this proof of concept, 4D printing technology can be used in the fabrication of soft robotics, medical devices, sensors, and wearable electronics [41].

**4.1 Light activated SMPS**

Light is an effective activation technique for SMPs due to its abundant nature, it being wireless and controllable. Light activated SMPs have been used in areas of self-assembly structures, complex folding methods, and transformative surface deformations.

## 5.0 Composites in 4D printing

Ge et al used a multi-material 3D printer to print an active composite material [15]. The printed active composite (PAC) consisted of a glassy polymer fibre embedded in an elastomeric resin. The glass fibres exhibited a shape memory effect with a shape fixity ratio of approximately 80% whereas the elastomeric resin was not capable of shape shifting and had a shape fixing ratio of 0. This bilayer laminate comprising a pure elastomer lamina and a PAC lamina with a prescribed fibre structure which includes the shape, size and orientation was printed, heated, stretched, cooled and realised. Upon release of the deformation stress, the laminate turned into a complex temporary shape due to the mismatch in the shape fixity ratio between the elastomer lamina and the shape memorizing PAC lamina. Depending on the fibre properties complex 3D configurations can be produced including bent, coiled, twisted, and folded shapes. This PAC laminate can be integrated with other structures or functional components to create active devices. For an example the PAC laminate could be used to enable active origami as a means to creating 3D structures.

## 6.0 Future of 3D printing and trends

4D printing technologies may require multi-material and multiple nozzles which is currently a limitation with 3D printing technology. Exploring different types of printing smart materials will give 4D printed parts that are lighter, stronger, react to different types of stimuli and give different property changes. 4D printing can find use in textiles and camouflage technology by altering not just the shape but also the colour and texture of the surface of the text. Textiles could react to different stimuli from the weather elements and morph allowing better ventilation or insulation to the wearer increasing the comfort [42].

Self-healing polymers have the possibility of increasing the life span of the 3D printed parts as any damage that occurs on the material can be self-repaired [43].

## 7.0 Conclusion

Additive manufacturing is still a growing industry still in its infancy. New materials, printing methods, software's and machines are constantly being developed and improved. Recently 4D printing has been gaining attention because 4D printed structures have the capability to change in form or function over time in response to stimuli such as pressure, temperature, wind, water, and light. 4D printing technology uses smart materials, designs to forecast change processes and smart printing can be applied to various fields from simple shape changes to bio printing for organisms. Using multi-material 3D printing and smart materials, 4D printing has been developed. This new technology provides a feasible method to fabricate a compact deployable structure. Smart materials are the cornerstone for 4D printing.

- Smart materials can allow the self-assembly of structures which are 3D printed and when exposed to external stimuli morph into the desired shape saving on time and costs of fabrication
- Use of smart materials also allows self-healing polymers. The use of self-healing hydrogels can allow pipes to self-repair if they develop a leak.
- Smart materials can serve the function of sensing and actuation directly into a material rendering external electromechanically systems unnecessary. This decreases the number of parts in a structure which require electronics and electro mechanic actuators.

4D printing has various potential end uses not only utilising the shape change property but other properties such as the colour and texture of the polymer. This could be useful in smart textiles which would react to different stimuli from the weather elements and morph allowing better ventilation or insulation to the wearer by change in colour and texture of the fabric increasing comfort and functionality.

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