A REVIEW ON 4D ADDITIVE MANUFACTURING - THE APPLICATIONS, SMART MATERIALS & EFFECT OF VARIOUS STIMULI ON 4D PRINTED OBJECTS

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Graphical abstract

Abstract

Additive Manufacturing, often known as 3D printing, is a process that uses a variety of materials to manufacture highly accurate items layer by layer. However, this technology has its limitations. One such limitation is the inability to print components larger than the printer’s size due to the limited size of the build chamber. 4D additive manufacturing or 4D printing is an innovative development based on 3D printing that allows objects to be reshaped after printing, using stimuli-responsive materials that require external stimuli. This article presents a review on 4D printing. Four databases, Google Scholar, ScienceDirect, IEEE Xplore, and Scopus database, were systematically searched for relevant review articles to identify and classify research studies. Specific keywords (4D printing, additive manufacturing, smart materials, stimuli, and application) were identified and used to guide the discovery of relevant studies. A total of 28 full articles were reviewed to study the applications of 4D printing, smart materials for 4D printing, as well as the effect of various stimuli on 4D printed objects. In conclusion, this review gives a summary of 4D printing and highlights its potential for revolutionizing manufacturing and further research is required to solve the limitations posed by this technology.

Keywords: 4d printing, additive manufacturing, smart materials, stimuli, application
1.0 INTRODUCTION

Additive manufacturing (AM), also known as fast prototyping or 3D printing, is a technique that allows simple or complex objects to be manufactured layer by layer under computer control using computer-aided design (CAD) [1], [2], [3], [4]. Since its creation in the 1980s, the popularity of three-dimensional (3D) printing has steadily increased. Chuck Hull developed the 3D technologies used for Stereolithography (SLA) technology in 1986, which helped to the development of 3D printing [5]. Several sectors, including healthcare, industrial production, automotive, and military, have used 3D printing [6]. Meanwhile, new 3D printing applications based on sophisticated materials are developing rapidly [7]. Common 3D printing processes include Digital Light Process (DLP), Fused Deposition Modeling (FDM), Selective Laser Sintering (SLS), Multi Jet Fusion (MJF), Electron Beam Melting (EBM), etc. [8], [9]. Hence, four-dimensional (4D) printing is a novel technical development in which materials may be distorted when subjected to external stimuli [10], [11], [12].

4D printing, also known as precisely controlled 3D printing, is the use of additive manufacturing to construct stimulus-responsive components that may alter form, structure, or functionality in response to particular stimuli without the need of electromechanical or electrical equipment [13]. Prof. Skylar Tibbets introduced the 4D printing technique for the first time in 2013. At his 2013 TED lecture at the Massachusetts Institute of Technology (MIT), he revealed a revolutionary design based on the deformation or change of a complex spontaneous structure as a consequence of environmental interaction over time [14]. This gave rise to the notion of 4D printing. It is a 3D printing invention intended to maximise the utilisation of deformation, self-repair, and self-assembly [15] in terms of form, structure, and function. The advent of 4D Printing technology has attracted a great deal of interest since it has created an unlimited number of opportunities.

For 3D printing, many material filaments, including metal, polymer, and ceramic, may be utilised to fabricate the pieces [16]. Polymer is the most often used material for 3D printing due to its large array of components and features. In fact, polymers are often employed for 4D printing because they are intelligent materials that can respond to physical stimuli [17]. Smart materials, also known as programmable materials or stimulus-responsive materials (SRM), are functionally and morphologically flexible [18], [19]. The kind of smart materials is a critical aspect in the capacity to modify itself. It defines the sort of stimulants necessary to induce changes in the functionalities, shapes, and qualities of 4D-printed components or parts. Polymers such as shape memory polymers (SMPs), liquid crystal elastomers (LCEs), and hydrogel are utilised for 4D printing [20]. Polylactic acid (PLA) from the SMPs is one of the most popular polymers for 4D printing due to its inexpensive cost and extensive variety of 3D printing applications [21]. Apart from that, PLA is a bio-based polymer with exceptional shape memory behaviour and features that may be used in the design and manufacture of a vast array of contemporary items [22].

The stimuli for 4D printed components can be partitioned into chemical, physical and biological stimuli. Chemical stimuli include chemicals, pH levels, reducing agents, and oxidizing agents [23]. Physical stimuli include humidity, force, current, light, ultraviolet (UV) light, and magnetic energy, whereas an example of biological stimulants is molecules, such as the presence of enzymes and glucose [24]. The majority of experimental investigations concentrated on the utilisation of heat or water [14]. The major shape-shifting reactions or outcomes are curling, folding, bending, contraction and expansion [25]. The influence of stimuli on 4D printed objects is an important consideration for the development of these materials for real-world applications. The influence of stimuli on 4D printed objects is an important consideration for the development of these materials for real-world applications. By considering the influence of stimuli on 4D printed objects, researchers can develop more adaptable, responsive, and customized materials for specific applications. This can potentially lead to significant advancements in various fields such as medical, aerospace, and robotics. Thus, the objective of this research is to examine the stimuli, smart materials, and the application of 4D additive manufacturing. A literature review was conducted to satisfy the objectives of the research.

2.0 METHODOLOGY

This study intends to offer a summary of the present state of 4D additive manufacturing. To achieve this goal, a comprehensive search of relevant literature
was conducted in four major academic databases: ScienceDirect, IEEE Xplore, Scopus and Google Scholar. These databases were selected as these databases enable access to several academic journals, conference proceedings, and other publications in the engineering and materials science domains, which are strongly relevant to 4D printing and additive manufacturing. Moreover, these databases are well-established and respected in academic communities, ensuring the reliability and relevance of the articles obtained. The search strategy involved using relevant keywords that were identified based on the research question and the review objectives. The Boolean combination of search terms was used to identify relevant articles from ScienceDirect and Scopus, which included “4D Printing” OR “4D Additive Manufacturing” AND “Smart Materials” AND “Stimuli” AND “Application.” In order to ensure the quality and relevance of the selected articles, only research and review articles from the Engineering subject area were included in the search on these databases, covering the period from 2019 to 2023. These restrictions were chosen because they are the most pertinent to the research and would provide the most comprehensive results while eliminating irrelevant or low-quality articles. However, IEEE Xplore and Google Scholar do not provide the same options for article/document type and subject area restrictions. Therefore, to obtain a manageable number of articles for review, the Boolean combination was modified to “4D Printing” OR “4D Additive Manufacturing” AND “Smart Materials” AND “Stimuli” AND “Application of 4D Printing,” with the same year’s restriction from 2019 to 2023 applied as in ScienceDirect and Scopus. After conducting the initial search in the three databases, the search results were exported and screened for relevance. The screening process involved evaluating each article’s title, abstract, and keywords against the selection criteria. The selection criteria were articles that discussed 4D printing, smart materials, stimuli, and applications of 4D printing. Articles that did not fulfill the selection criteria were unavailable in full text were excluded. The chosen publications were then evaluated and analyzed to determine the present status, recent advancements, obstacles, and prospective future research areas of 4D additive manufacturing. The information obtained from the selected articles was used to synthesize the findings and provide a comprehensive overview of the topic. Overall, this approach ensures that a thorough and systematic review is conducted and that the results obtained are comprehensive, relevant, and accurate.

**3.0 RESULTS AND DISCUSSION**

The initial search phase for this literature review produced a total of 583 titles across four databases, including Google Scholar, ScienceDirect, IEEE Xplore, and Scopus. After the title review phase, 432 articles were excluded from further consideration based on their titles. The remaining 151 abstracts were retrieved and thoroughly reviewed. Out of the 151 abstracts, 76 were eliminated as they did not meet the selection criteria of this review. The remaining 75 full papers were then retrieved and carefully reviewed to determine if they met the selection criteria. Among these 75 publications, an additional 47 were excluded because they failed to satisfy the selection criteria, leaving 28 papers to be included in the review. Figure 1 depicts a summary of the literature search and evaluation process.

**3.1 4D Printing**

Professor Skylar Tibbits demonstrated how alterations occur in a stationary object produced by 3D printing throughout time during the TED talk, “The emergence of 4D printing,” at MIT in 2013 [26]. It has been shown that a simple 3D object or part can be converted into a more complicated form over time. This has led to a new printing paradigm that embraces a new dimension of 3D printing, namely time, known as 4D printing. In 4D printing, the materials used must have specific properties that allow them to respond to external stimuli in order to change the function and shape of the 3D print after printing [15]. This responsiveness allows printed objects to be triggered with particular functions after manufacturing. Several printing processes, including fused deposition modelling (FDM), polyjetting, Selective laser melting (SLM), and stereolithography (SLA), are now utilised for 4D printing [27], [28]. Shape-memory polymers (SMPs) and hydrophilic polymers are examples of 4D printing materials that use the FDM printing process. In PolyJet printing, SMPs and other hydrophilic polymers can be used to create materials that respond to stimuli such as changes in humidity or temperature. SLA printing can produce materials made of SMPs and piezoelectric materials, which can change their shape in response to either an...
electrical signal or temperature. On the other hand, SLM printing can create objects that are made of shape memory alloys [29]. Figure 2 shows the different between 3D Printing technology and 4D Printing technology.

Figure 2 3D Printing and 4D Printing Technology [30]

Figure 3 shows the overall flow of the 4D printing process using polymer, including the stimuli and effects [31]. The smart polymeric materials are placed into the 3D printer, along with any essential additives like the nanoparticles. The coding of the print path is digitally sent to the 3D printer after being programmed by mathematics and CAD. The 3D printed item is in a “programmed 4D printed state,” which is ready for controlled changes over time if it is programmed during the printing process. In contrast, the 3D-printed item will remain “unprogrammed,” which needs to be programmed manually after printing if programming does not occur during printing. The programmed 4D-printed object is subjected to a predetermined stimuli in order to achieve the desired result.

Figure 3 Process Flow of 4D Printing [32]

3.2 Smart Materials For 4D Printing

As a result of rapid improvements in 3D printing, materials can now be positioned more accurately and flexibly, which is of great assistance to 4D printing. 4D printing employs intelligent materials with the capacity to alter their properties over time. When stimulated, these intelligent materials can self-repair, self-assemble, and self-heal [33]. Nowadays, polymeric smart materials such as shape memory polymers (SMPs), Liquid Crystal Elastomers (LCEs), and hydrogels are the most often used materials in 4D printing [34].

3.2.1 Shape Memory Polymers (SMPs)

SMPs are intelligent materials that display shape-memory behaviour and are capable of regaining their original form after obtaining a transitory shape [35]. When subjected to stimuli such as UV radiation, humidity, heat, etc., the SMPs exhibit a high degree of durability and a quick response to stimulation, allowing them to recover substantial deformation [36]. For instance, when heated beyond the glass transition temperature (Tg), the chain atoms of polymeric smart materials inside the printed object become mobile [37], [38], [39]. As a result, heat as a stimulus will cause the item to deform into a secondary shape. PLA, polyvinyl alcohol (PVA), polyester (PE), acrylonitrile butadiene styrene (ABS), etc., are examples of SMPs.

3.2.2 Liquid Crystal Elastomers (LCEs)

LCEs are also another polymeric smart material that can react to stimuli. LCEs, unlike SMPs, exhibit a reversible change in shape due to the transition between isotropic state and liquid crystal state, also called nematic state [40], [41]. The mesogens, rigid molecules in a liquid crystal state, are arranged by a director in a standard orientation set for use in 4D printing. When the temperature reaches a critical level, the mesogens start losing some of the order and relaxing, causing shrinkage and expansion orthogonal to the director. The temperature differs from the Tg, called the nematic-to-isotropic temperature (TNI) [42]. In the printing process, mesogens are oriented along the printing path using extrusion AM techniques such as Direct-Ink-Writing (DIW). Figure 4 shows a schematic diagram of LCEs 4D printing, including the LCE’s shape programming.

Figure 4 Schematic of LCEs 4D Printing [32]
3.2.3 Hydrogels

Hydrogels are another polymeric smart material for 4D printing. They are hydrophilic materials but do not dissolve in water due to crosslinks between polymer chains [43], [44]. Thus, hydrogels can be submerged in water and absorb water until they reach saturation. When the hydrogels are dried and taken from the water, the water will exit the polymer matrix, and the hydrogels’ primary shape is regained. This reversible shape-changing effect can exploit hydrogels to create self-folding objects. For example, Baker et al. (2019) made use of the FDM 3D printer to create active flat forms composed of polyurethane hydrogels layered between hydrophilic polyurethane elastomers [45]. The exterior elastomeric layers have gaps at each hinge, allowing water to reach the hydrogel and cause it to expand. Figure 5 shows the shape transformation of hydrogel-printed object.

Figure 5 Shape Transformation of Hydrogel-Printed Object [45]

3.3 Stimulus for 4D Printing

There are different sorts of stimulus for 4D printing. These may be categorised as physical stimuli, chemical stimuli like pH levels and ionic concentrations, biological stimuli like as enzymes and glucose, as well as combinations of stimuli. In 4D printing, physical stimuli such as light, temperature, liquid or humidity, magnetic field, and force are employed more often than chemical and biological stimuli [46]. These types of stimuli can alter the form or shape of an object by transforming the chain movements or arrangement of the inner atomic packing of smart materials. These types of shape-changing behaviour have given materials with hitherto unseen characteristics that can adapt to physical changes in ambient circumstances.

3.3.1 Light Stimulus

Light is a popular stimulus used in stimulated 4D-printed objects. However, it is an indirect stimulus for smart material deformation compared to liquid and temperature stimuli. The smart materials that respond to light stimuli are called photo-responsive smart materials [47]. Light stimulation can regulate the polymeric material shape through remote induction. It is effective as an external stimulus for changing the colour of printed items as it can conduct dominant influence in both time and space. Figure 6 shows a multicolour 4D printing of SMPs [48]. They accomplished remote light control by utilizing colour-dependent selective light absorption and temperature control in multicoloured SMPs materials. The programmed structure would curve into an n-pattern under red light, whereas reverting to its initial state under blue light.

Figure 6 Printed Samples Stimulated by Light Stimulus [48]

3.3.2 Temperature Stimulus

Temperature is also often used as a stimulus in 4D printing technology. The smart materials that respond to temperature stimulation or heat are termed thermo-responsive smart materials. Thermo-responsive intelligent materials can reorganise and remodel in response to temperature stimuli [49]. For instance, a printed item may create a secondary state by submitting it to high temperatures under stress. In contrast, the item may revert to its original condition by cooling it. Two key processes contribute to the form alterations of these smart materials in response to heat stimuli: the shape memory effect (SME) and the shape change effect (SCE) [50]. Figure 7 demonstrates that an origami pyramid printed using shape-memory polymer may self-fold when exposed to solar heat.

Figure 7 Origami Pyramid Stimulated by Heat [7]

3.3.3 Liquid/Humidity Stimulus

The earliest stimuli used in 4D printing were liquid and humidity. The smart materials that respond to
humidity or liquid stimuli are called moisture or liquid-responsive smart materials [51]. Due to their extensive irri
tancy and wide variety of uses, these materials are of considerable interest. Liquid-responsive smart materials can deform or inflate when exposed to liquid or humidity stimuli and then return to their initial shape upon drying. Nevertheless, liquid-responsive smart materials’ shrinking or swelling degree should be controlled accurately throughout the transformation to ensure the printed structure’s integrity. Figure 8 shows a film produced with water-sensitive materials created by stearyl-modified cellulose. The film will bend deformation when exposed to water owing to unequal water absorption.

![Figure 8 Film Stimulated by Water][5]

3.4 Application of 4D Printing

Owing to the current applications of 4D printing across a variety of industrial areas, such as aerospace, and the food industry, 4D printing provides an excellent means of attaining the goal of advancing technology and innovation in these fields in the near future. With its ability to create dynamic structures that can respond to various stimuli, 4D printing holds the potential to revolutionize manufacturing processes and open up new avenues for research and development. The sections that follow describe the uses of 4D printing in many areas, including healthcare, robotics, and aerospace, etc. [52].

3.4.1 Application in Healthcare/ Medical

With the increasing rise of the worldwide population and the visibility of health issues, there is a greater need for healthcare. 4D printing can address these problems through 4D bio-printing, which combines time with 3D printing to produce biomaterials for applications such as shape memory biomedical scaffolds [53]. The shape transformation in a biological environment and biocompatibility are the main prerequisites for applicability in this arena [54]. Figure 9 shows the in vitro experiment of a printed vascular scaffold to demonstrate the geometric adaptability of the shape memory bio-scaffolds for possible biomedical implants. The printed scaffold expands while it is heated over the glass transition temperature, whereas it will restore to a permanent shape after excreting the stored energy. The 4.5 mm diameter tube extends 3 mm to 7.5 mm as the printed scaffold applies force on the tube wall during its shape transformation process. This procedure mimics the implantation of a vascular stent to exert pressure and prevent the blocking and narrowing of the vascular wall due to coronary artery disease. This significantly lowers the potential threat of surgery.

![Figure 9 Shape Memory Biomedical Scaffold][53]

3.4.2 Application in Aerospace

Moreover, printing self-folding peptides and proteins are the other medicinal application for 4D printing technology [55]. This technology might be used to develop configurable biological materials that can modify structure and characteristics. These natural materials could lay the groundwork for intelligent pharmacology, tailored therapy, and programmable body tissues and cells [54].

It is crucial for the aerospace sector to produce components for space projects and missions [56]. The components should be manufactured at lower costs and be durable for an extended time. The aircraft industry has potential uses for 4D printing as it can sustain intense environmental conditions and has cheap manufacturing costs. Apart from that, 4D-printed structures may self-assemble and change in response to environmental conditions, allowing them to meet one of the main criteria of space missions: the employment of materials should be self-sustainable. These materials can be used to produce the aerospace components such as satellite parts, equipment to explore space, spacecraft gear, etc. Polyether ether ketone (PEEK), a thermoplastic polymer able to function stably under enormous pressure and temperature resilience, has been printed utilizing the SLS process to produce components for satellites and spacecraft [54]. Figure 10 shows the reflectors that can be self-deployable and used in spacecraft. These reflectors are manufactured by 4D printing using intelligent materials.
3.4.3 Application in Soft Robotics

The 4D printing technology can not only offer the capability to handle complicated structural components efficiently, but it also imbues the material structure with inherent intellectual ability and accomplishes structure-intelligence integration [5]. Much research on soft robotics produced with the 4D printing technique has been published over the past few years. The advancement of engineering design, soft programmable intelligent materials, and other fields of science all contribute to the rapid advancement of soft robotics. The mobility and flexibility of soft robots are superior to that of conventional robots created by engines, hydraulic pistons, bearings, and hinges [2]. This is because conventional robots are only intended for specific situations and environments and cannot endure all environmental factors. They are unable to achieve substantial deformations or execute activities that need flexibility. In contrast, soft robots may alter form and size to meet changing demands and can be integrated into more complicated processes with better security and ecological compliance [57]. Figure 11 shows the soft robotic finger created by 4D printing technology.

3.4.4 Application in Food Industry

The application of 4D printing technology in the food production industry has facilitated the development of this sector. Teng et al. exploited 4D-printing technology to analyse recent trends and advancements in the food and agriculture industries [58]. The appropriate studies of material characteristics, stimulants, and construction designs have been carried out to conduct several experiments with printing starches, hydrogels, and proteins. 4D printing is superior to 3D printing for the food industry since 4D-printed food has significantly more outstanding properties [20]. For instance, 4D-printed food can retain flavour, nutrients, or colour during storage, whereas 3D-printed foods cannot. Furthermore, the 4D printing technology can manipulate the dryness of printed food so that the forms or contours of food may also be manipulated. Additionally, a study conducted by Guo et al. (2021) has reported the observation of a 4D transformation in 3D-printed buckwheat dough and lotus root powder gel when they were stimulated by microwave heating [59]. Figure 12 shows the various types of food printed using 4D printing technology, such as self-encase sushi, self-folding multi-tasty biscuits, self-wrapping noodles for convenience, etc.

3.5 Advantages and Limitations of 4D Printing

4D printing is a cutting-edge process for producing sophisticated and adaptable components with great accuracy. It has the vast potential to manufacture various outstanding products through the applications of innovative materials. Nevertheless, the 4D printing technology’s limitations and constraints should be addressed. Table 1 shows the advantages and limitations of the 4D printing technology.

Table 1 Advantages and Limitations of the 4D Printing Technology [60]

<table>
<thead>
<tr>
<th>Advantages</th>
<th>Limitations</th>
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<tr>
<td>Capability to manipulate time and space during the printing operation of the part</td>
<td>Inadequate control over the various stages of deformation</td>
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<tr>
<td>Able to create dynamically structured flexible components</td>
<td>Shape prediction of the printed part requires proper modeling and simulation</td>
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<tr>
<td>The object that has been printed has the ability to change its function, colour, and shape</td>
<td>The transformation time of a 4D-printed item is relatively long</td>
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<tr>
<td>The necessity of post-processing is minimal</td>
<td>Mechanical property constraints</td>
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4.0 CONCLUSIONS

In conclusion, this article provides a comprehensive overview of 4D additive manufacturing technology, which is an innovative development based on 3D printing that allows objects to be reshaped after printing using stimuli-responsive materials. The study systematically searched four databases, Google Scholar, ScienceDirect, IEEE Xplore, and Scopus, to identify and classify research studies related to 4D printing. Through the search, 28 full articles were reviewed to study smart materials for 4D printing, including the impact of various stimuli on 4D printing, advantages and limitations of 4D printing, as well as the impact of various stimuli on 4D printed objects. The findings suggest that smart materials, such as SMPs, LCEs, and hydrogels, have significant potential for 4D printing, along with stimuli such as light, temperature, and liquid/humidity. The applications of 4D printing are wide-ranging, including healthcare/medical, aerospace, soft robotics, and food industry. However, the technology is still in its infancy, and there are several challenges to overcome, including the need for more robust and scalable manufacturing processes and the development of new materials with enhanced properties. Despite these limitations, 4D printing has the potential to revolutionize the manufacturing industry and enable the creation of intelligent, adaptive, and self-assembling structures. Overall, this review highlights the potential of 4D printing in revolutionizing manufacturing, although there are still some challenges that need to be overcome.

Conflicts of Interest

The author(s) declare(s) that there is no conflict of interest regarding the publication of this paper.

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