Research Status of Material Design for Intelligent Structures in 4D Printing

Zhe Li

{lzsnbdr@outlook.com}
College of Physics and Materials Science, Nanchang University, NanChang, 330000, China

Abstract. Four-dimensional printing extends additive manufacturing by producing structures that transform over time in response to external stimuli. This review summarizes recent progress in smart materials for 4D printing, focusing on shape memory polymers, hydrogels, liquid crystal elastomers, and magneto-/electro-responsive composites. We compare their response mechanisms, reversibility, actuation speed, and printability, and discuss advances in artificial intelligence -assisted design and multiphysics simulation for structural optimization. Challenges such as energy coupling, interfacial durability, and scalability are identified. We conclude that sustainable material development, cross-scale modeling, and integrated closed-loop control are key to advancing 4D printing toward engineering applications.

Keywords: 4D printing, Smart structures, Smart materials, Artificial Intelligence.

1 Introduction

4D printing can be seen as an upgraded form of 3D printing technology that does not merely focus on shape formation, but enables the material used to deform after printing due to external factors such as time, temperature, and light, that is, to make the printed object "move". Using shape memory material to execute the printed structure, it can automatically return to the shape set by the designer when heated [1]. Compared with traditional 3D printing which focuses more on static construction, the more prominent feature of 4D printing is that it can achieve dynamic functional transformation of structures. In the design of intelligent structures, material properties, geometric configurations, and external field stimuli work together to affect the overall performance of the system. In recent years, new material systems such as shape memory polymers, hydrogels, liquid crystal elastomers, and multi-responsive composites have gradually begun to be applied. These materials give the structure special properties such as adaptive and self-healing [2], but they have shortcomings such as slow response speed, limited deformation accuracy, and performance may decline after long-term use, so there is still room for improvement. This paper examines the relationship between materials and structures in 4D printing, the response mechanisms and print fit of major materials, as well as the development

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trends in recent years such as AI-assisted design, multi-field coupling control and green materials.

2 Basic Concepts of Intelligent Structures and 4D Printing

2.1 Physical Mechanisms of Intelligent Structures and Key Points of Their Classification

The operation of smart structures is supported by clear physical principles. The core consists of two major parts, namely the material response mechanism and the structure amplification mechanism. The material response mechanism is related to the way the structure senses external stimuli and converts this perception into physical changes, classified according to the physical and chemical properties of the material. The more common types of sensing include heat-responsive (such as shape memory polymers), light-responsive (such as liquid crystal elastomers), humidity or ion-responsive (such as hydrogels), and electromagnetic-responsive composites, which may experience volume changes, phase transitions, or internal structural reconfigurations when exposed to external factors. And the speed at which they respond and whether they can be reused often serve as key references for differentiating artificial intelligence structures.

Mechanical amplification mechanisms, by means of specific geometric strategies, transform the fine deformations at the material level into macroscopically visible functional outputs, which is an essential part of intelligent structures. Common deformation amplification designs include folding, pleated structures, beam and plate configurations, pore array Settings, and gradient functional structures, depending on the geometric framework. Smart structures can be further classified into types such as origami structures, buckling excitation structures, flexible deformable structures, and field-responsive adaptive structures.

As mentioned above, the classification of smart structures can be carried out in terms of material response and structural amplification. When the temperature changes and is stimulated by many external factors such as light, electromagnetic fields or humidity, the structure can achieve controllable deformation, displacement or functional transformation through the mechanisms mentioned earlier. This physical principle is also an important theoretical basis for the realization of 4D printing, a smart manufacturing technology.

2.2 Ways to Integrate Smart Structures with 4D Printing

After getting familiar with the basic meaning of 4D printing and the physical principles of smart structures, we can delve deeper into how to combine the two. The main ways of integration are reflected in materials and design. 4D printing focuses on achieving dynamic changes in structure through programmable reactions of materials, and thus combines material synthesis, geometric distribution, and external stimuli. This is crucial for creating smart structures.

Tibbits and his colleagues were the pioneers who first suggested bringing in time as an extra dimension to the whole 3D printing process. Their big emphasis was on tapping into the natural behaviors and properties of the materials themselves, so that the printed structures could evolve or transform on their own as time passes. This fresh concept really paved the way and established the core ideas behind the rise of 4D printing technology [1]. This idea also implies that material constitutive behavior and structural topology should be treated as design variables in a systematic way. Based on this, Yan et al. further demonstrated that by using multi-material printing or functional gradient design, different regions of the structure could respond differently to the same external field stimulus, thereby achieving delayed or staged deformation control [3], which provides a feasible design approach for the development of complex forms.

The adaptation of the external field excitation to the material response mechanism is crucial for the performance of intelligent structures, not just depending on the partition design of the material. Zang et al. have shown that liquid crystal elastomers can achieve rapid and reversible deformation responses when in the light-thermal coupling excitation state [4], indicating that the combination of different excitation forms is beneficial for optimizing the response speed and controllability of the structure. Relevant studies show that the appropriate selection of the type of external field and its association with material properties. It is the main approach to achieving high-performance 4D printing of intelligent structures.

In terms of co-design of material and structure, Chu's review show that multi-material 4D printing give a possible way to control time-dependent deformation of smart structure. That means use polymer with different glass transition temperature or shape memory effect in one same structure. In this way, different part will response at different temperature and have different recovery process under same outside stimulus [5]. Based on this, by implementing differentiated design for the material formula or printed area, the structure can gradually undergo morphological changes such as local bending, unfolding or folding when heated, thereby achieving step-by-step deformation development. This approach links the shape memory characteristics of the material itself to the structural design without adding additional excitation sources, The sequence and process of deformation can be adjusted, which reflects the main idea of the co-design of material properties and structural functions in 4D printing.

In summary, the realization of smart structures through 4D printing cannot be achieved by a single factor. It not only needs many things work together, like material response, design of structure, and outside force, but also need to think about material behavior and how structure change shape when design. To some extent, the functional integration and application potential of 4D printed structures can be optimized.

3 The Material System and Design Concept of Intelligent Structures

3.1 Material Classification and Basic Response Mechanisms

To achieve controllable and repeatable deformation of 4D printed structures, the key lies in the response characteristics of the selected material system and fit with the process parameters. Unlike traditional additive manufacturing which only focuses on geometric accuracy, 4D printing places more emphasis on the "functional programmability" of the material, that is, the ability to exhibit pre-set performance under external force excitation, Reversible physical or chemical changes.

The ideal 4D printing material should meet two conditions. First, it should produce stable and reversible structural or phase changes when stimulated by factors such as heat, light, electric field, magnetic field, or humidity. Second, it should have rheological properties similar to common printing processes such as fused deposition modeling (FDM), stereolithography (SLA), direct ink writing (DIW), and inkjet printing. and inkjet printing. In terms of curing speed and interlayer bonding strength, if the reaction characteristics of the material do not match the process requirements, even if it is highly stimulation-sensitive, it is difficult to ensure the consistency of the printed structure in terms of geometry and function. According to the differences in response mechanism and design programmability, the more common smart materials in current 4D printing can be roughly classified into three categories.

3.1.1 Shape Memory Polymers (SMPs)

Shape memory polymers, a top smart material in 4D printing, allow reversible shape changes from outside triggers. This relies on the “freeze-recovery” trait of their molecular chains: when heat tops the glass transition temp, chains get moving and restore the original form. On cooling, they lock back in place, thus preserving the transient structure. Tibbits et al. were the first to use SMP material in the 4D printing concept, which exhibits adaptive properties over time under thermal excitation, laying the theoretical foundation for subsequent studies [1].

3.1.2 Hydrogels

Hydrogels are typical environmentally responsive materials, and their volume changes are reversibly regulated by swelling equilibrium. Once the external temperature, humidity, pH value, or ion concentration changes, hydrogels will absorb water and expand or lose water and contract, thereby achieving morphological changes. For instance, Gladman et al. utilized hydrogel composite materials to achieve programmed shape in water through swelling for bionic 4D printing [6]. Wan et al. claimed that by adjusting the crosslinking density and light curing parameters, the mechanical strength and interlayer bonding accuracy of the hydrogel structure could be significantly improved [7], thereby providing better deformation controllability and printing adaptability in 4D printing. Hydrogels, with their good biocompatibility and environmental sensitivity, have been widely applied in biomimetic structures, drug controlled-release, wearable devices, and soft robots, but their long-term stability and dehydration shrinkage remain key points of study, and improvements need to be made in both material system design and printing environment regulation.

3.1.3 Liquid Crystal Elastomers (LCEs)

Liquid crystal elastomers, which have the orientation order of liquid crystal molecules and the elasticity of elastomers, are one of the key representatives of photothermal responsive materials. Their deformation principle is reflected in the reversible conversion between the liquid crystal phase and the isotropic phase: once the orientation of the molecules is disrupted by heating or light exposure, the material undergoes macroscopic deformation; When the stimulation disappears, the molecules rearrange themselves, thus restoring their own form.

3.2 Printability and Processing Constraints of the Material

In the actual operation of 4D printing, the printability of smart materials is closely related to process parameters. The degree of fit between the two will affect the forming quality and structural performance. If the material is prone to delamination, warping or interface instability during processing, it may cause inaccurate final deformation or even loss of response function. Therefore, material properties need to be taken into account in the design stage. The synergistic relationship between process conditions and structural design.

In extrusion printing techniques such as DIW or FDM, the viscosity of the material and the proportion of particle mixture directly affect the stability of inkjet or stacking, Mocerino et al. found [7] that even for the same material, as long as there are differences in the geometric structure design, the final print fit will be different, it highlights the significance of "fit" between the structure and the material.

When using light-curing technologies such as DLP or SLA, the situation is different. In a light-curing system like DLP, the polymerization rate of the photosensitive resin and the depth of light can affect print resolution and crosslinking. SMPs and hydrogel systems often require adjusting the ratio of photoinitiators to monomers to coordinate print accuracy and stimulation response sensitivity. Wan et al. also proposed the strategy of "closed-loop photocuring detection" [8], which can promptly correct exposure energy and interlayer bonding conditions to further improve print accuracy and response consistency.

There is also a multi-material printing form synthesized from multiple materials, where interfacial effects can cause interfacial stress concentration due to the significant differences in thermal expansion coefficients of different materials. However, through chemical modification or the formation of functional transition layers, the toughness and thermal conductivity of the interface can be greatly optimized [9], the printing path can be improved and the heat treatment process can be enhanced. It helps to reduce interfacial fatigue, which is crucial for extending the service life of multifunctional composite structures.

In the LCEs system, liquid crystal elastomers (LCEs) are subject to scale effects and orientation control. Their molecular orientation can influence macroscopic deformation performance. Printing speed, nozzle shear rate, and light polarization conditions all affect molecular alignment [4]. In practical applications, by adjusting these process parameters, It can effectively enhance the consistency of LCES-like structural deformations, thereby making them more reproducible. Jiang et al. reported that continuous fiber-reinforced LCE composites can significantly improve deformation by controlling the direction of the fibers [10].

In recent years, researchers have increasingly focused on integrating composites with multifunctionality for design, and Lalegani Dezaki and Bodaghi have developed a sustainable magnetoelectrically active shape memory polymer composite that uses magnetic particles to achieve controllable deformation [11]. Moreover, researchers have developed multi-stimuluse-responsive materials, such as the protein-based hydrogel system reported by Narupai et al., which can deform on its own under various stimuli such as heat and light [12], using design methods such as layered or gradient distribution, where each region shows a lag or multi-stage response when subjected to the same excitation More complex functional purposes can be achieved [5].

Generally speaking, whether 4D printing materials can be printed is not determined by a single parameter. It is influenced by the combined effect of three aspects: material formula, process path, and structural design. In the future, a digital feedback system of "material - process - performance" can be created by relying on artificial intelligence and multi-physics simulation technology This leads to precise control of the printing process and performance prediction. Wan et al. summarized the latest trends in advanced materials and structures for 4D printing in their review [8], providing an important reference for related research.

4 Challenges and Trends

4D printing faces multiple bottlenecks as it moves from the lab to engineering. First, energy transfer and timing coordination in multi-physics coupling are difficult. If the stimulus sources are not synchronized or the energy coupling efficiency is low, it can lead to slow, excessive or uncontrollable responses; The durability and interfacial fatigue problems of materials under repeated external field effects also seriously affect long-term stability. Secondly, processing compatibility issues such as poor interfacial bonding between materials and mismatched coefficients of thermal expansion in multi-material printing can weaken the overall mechanical properties and cycle life of the structure. Furthermore, sustainable smart materials for engineering needs are inadequate in terms of mechanical strength, thermal stability and process compatibility, which limits the promotion of green and recyclable systems. Finally, the current modeling and simulation are mostly at a single scale (from molecular dynamics to macroscopic finite element as arrays), lacking a cross-scale framework that integrates molecular structure, interface effects, microscopic configuration and macroscopic response; At the same time, there is a lack of unified parametric representation and standardized evaluation metrics (such as response speed, cycle fatigue life, stimulus conversion efficiency, etc.), resulting in poor repeatability, difficulty in comparison with each other and hindering industrial transformation.

Future research and engineering are expected to evolve around an integrated four-part framework comprising materials, control, simulation, and standards. In the materials dimension, development is anticipated to shift toward sustainability, with increasing emphasis on recyclable systems, bio-based materials, reprocessing smart polymers capable of bond reformation, and thermoplastic liquid crystal elastomers (LCEs). Such materials enable the preservation of shape-morphing functionality while simultaneously addressing lifecycle management and environmental impact.

At the same time, multi-stimulus actuation and control are likely to become more prevalent. Materials will be engineered to respond to multiple external stimuli, while embedded sensing elements and closed-loop feedback mechanisms—such as real-time monitoring or digital twin frameworks—will allow precise regulation of deformation timing, mode, and energy coupling across different stimuli.

In parallel, there is a growing need for material-centered modeling and simulation platforms. Supported by high-throughput simulations and machine learning techniques, these platforms can accelerate material discovery, optimize design parameters, and predict long-term performance and durability. To ensure consistency and facilitate translation toward industrial applications, standardized testing methodologies for 4D printing must be established. This includes clearly defined key performance indicators, unified evaluation procedures, and reproducible test protocols, enabling meaningful comparison across studies and alignment with engineering requirements.

In general, if three main parts grow together — sustainable smart materials, multi-field cooperation control, and AI-based modeling 4D printing can move from "material first" stage to "smart manufacturing first" stage, and be used more in soft robot, flexible electronic, medical device, and space tech..

5 Conclusion

Programmable smart materials are very important for 4D printing to change shape. This passage talks about how shape memory polymers, hydrogels, liquid crystal elastomers and multi-response composites work and can be printed. Shape memory polymers can remember original shape and go back when heated. They need heat to make deformation happen. Hydrogels can swell or shrink with water, so they use moisture as trigger. Liquid crystal elastomers can change shape under light or heat because of their inner structure. Multi-response composites can react to many stimuli like heat, light and magnetic field at same time.

Their performance depend on material property, how the shape is built (like geometry), and what outside force is used. These three things together decide how accurate the shape change will be. If one of them not good, the final shape may not right. So we must control all factor carefully during printing and activation. Whether reversible and stable or not, using a variety of different types of materials to perform printing operations. Link multiple triggering factors together and regulate them in terms of direction. It is necessary to develop sustainable sources of raw materials and establish closed-loop multi-physics regulation mechanisms to adapt to practical engineering applications. It is also indispensable to build a cross-level model creation framework and a comprehensive evaluation standard system, and to optimize the design speed and improve its reproducibility by means of data-driven methods and by referring to standardized indicators, so as to move towards the goal of large-scale production.

References

[1] Tibbits, S.: 4D Printing: Multi-Material Shape Change. Archit. Des. 84(1), pp. 116–121 (2014)

[2] Momeni, F., Liu, X., Ni, J.: A Review of 4D Printing. Mater. Des. 122, pp. 42–79 (2017)

[3] Yan, S., et al.: Shape Memory Polymer Composites: 4D Printing, Smart Structures, and Applications. Res. 6, p. 012345 (2023)

[4] Zang, T., Fu, S., Cheng, J., Zhang, C., Lu, X., Hu, J., Xia, H., Zhao, Y.: 4D Printing of Shape-Morphing Liquid Crystal Elastomers. Chem. Bio Eng. 1(6), pp. 488–515 (2024)

[5] Chu, H.: 4D Printing: A Review on Recent Progresses. Micromachines. 11(9), p. 796 (2020)

[6] Gladman, A. S., Matsumoto, E. A., Nuzzo, R. G., Mahadevan, L., Lewis, J. A.: Biomimetic 4D Printing. Nat. Mater. 15(4), pp. 413–418 (2016)

[7] Mocerino, D., Ricciardi, M. R., Antonucci, V., Papa, I.: Fused Deposition Modelling of Polymeric Auxetic Structures: A Review. Polymers. 15(4), p. 1008 (2023)

[8] Wan, X., et al.: Recent Advances in 4D Printing of Advanced Materials and Structures for Functional Applications. Adv. Mater. 36(34), p. e2312263 (2024)

[9] McLellan, K., Sun, Y. C., Naguib, H. E.: A Review of 4D Printing: Materials, Structures, and Designs. Bioprinting. (2022)

[10] Jiang, H., Chung, C., Dunn, M. L., Yu, K.: 4D Printing of Liquid Crystal Elastomer Composites with Continuous Fiber Reinforcement. Nat. Commun. 15, art. 8491 (2024)

[11] Dezaki, M. L. L., Bodaghi, M.: Sustainable 4D Printing of Magneto-Electroactive Shape Memory Polymer Composites. Int. J. Adv. Manuf. Technol. 126, pp. 35–48 (2023)

[12] Narupai, B., Smith, P. T., Nelson, A.: 4D Printing of Multi-Stimuli Responsive Protein-Based Hydrogels for Autonomous Shape Transformations. Adv. Funct. Mater. 31, p. 21011012 (2021).