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Review

4D Food Printing: Transforming Culinary Creation with Time-Responsive Technology

Sindhu P. M.[1]*, Ranjani M.[1], Kiran M.G.[2], Sachin M. S.[3]

[1]Division of Food Science and Postharvest Technology, ICAR-IARI, New Delhi
[2]Department of Fruit Science, UHS, Bagalkot, Karnataka
[3]Department of Entomology, UAHS, Shivamogga Karnataka
*Corresponding Author: sindhupm1997@gmail.com

Abstract

Four-dimensional (4D) printing represents an innovative extension of 3D printing technology, introducing the concept of temporal transformations in fabricated structures. This review explores the key aspects of 4D printing, including materials, techniques, and applications, with a special focus on 4D printing in the realm of food. Smart materials, such as shape memory polymers, liquid crystal elastomers, and composite hydrogels, are fundamental to achieving dynamic responses in 4D printing. The review discusses the potential of 4D bioprinting and its applications in fields like tissue engineering and drug delivery. In the context of 4D food printing, the customization of food products and the use of stimulus-responsive materials are examined, along with the role of printing software in shaping the quality of printed objects. The paper also underscores the current research gap in 4D food printing and its nascent stage. Ultimately, 4D printing offers a promising avenue for creating dynamic structures that adapt to various stimuli, with far-reaching implications across multiple domains.

Keywords: 4D printing, additive manufacturing, 4D bioprinting, food printing, printing software.

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Introduction

Four-dimensional (4D) printing, a new area of additive manufacturing, is a development of 3D printing. This innovative technology introduces the concept of altering the printed configuration over time, initially conceptualized by a team at the Massachusetts Institute of Technology (MIT) in 2013. The process of 4D printing shares similarities with 3D printing, encompassing stages such as 3D design development and the use of a 3D printer to fabricate structures. The key differentiators between 4D and 3D printing lie in the incorporation of intelligent design and smart materials, enabling 4D-printed structures to undergo shape or functional changes [1].

Materials used in 4D printing offer unique attributes such as self-assembly, versatility and self-healing properties [2]. The primary constituents of 4D printing materials predominantly consist of single or multilateral polymers, which encompass shape memory polymers, liquid crystal elastomers and composite hydrogels. The capacity for stimuli-responsive shape alterations in 4D printing contributes to space-saving benefits for transportation and storage [3]. Consequently, 4D printing technology finds applications across various fields, including robotics, biomedical applications, tissue engineering, electronic devices, among others [4]. Nevertheless, in the domain of food, there exists a dearth of literature and it is still in its infancy stage. The adaptability of materials in 4D printing offers the advantage of tailoring material responsiveness configurations, which, in turn, aids in reducing material consumption [5]. Moreover, 4D printing has the potential to cater to consumer preferences for unique food products. It also presents opportunities for chefs to precisely control the final cooking stages, enhancing the visual appeal and taste of food [6]. In the realm of 4D printing, it becomes possible to attain desired product attributes within an appropriate timeframe, although these qualities may diminish during the storage of printed items.

Additive manufacturing has received a lot of interest over the last three decades because of its capacity to create intricate 3D structures [1]. A more recent innovation, known as 4D printing, extends the capabilities of 3D printing by introducing a temporal dimension. This allows for the creation of dynamic structures that may alter shape, function, and attributes in response to diverse stimuli such as temperature, pH levels, ionic concentration, and electric and magnetic fields. Nature offers numerous examples of 4D transformations, seen in phenomena like the hygroscopic unfolding of pinecones and the sun-tracking behaviour of plants like sunflowers [7]. The essential elements of the 4D printing process include the choice of additive manufacturing techniques, the selection of stimuli, the use of responsive materials, the interaction mechanisms at play and mechanical modelling [8]. Various technologies are employed in 4D printing, including micro extrusion-based printing, fused deposition modelling, stereolithography and digital light projection [9]. Because of its simplicity and versatility in dealing with a range of inks, micro-extrusion-based printing stands out as a commonly utilized approach across diverse printing technologies.

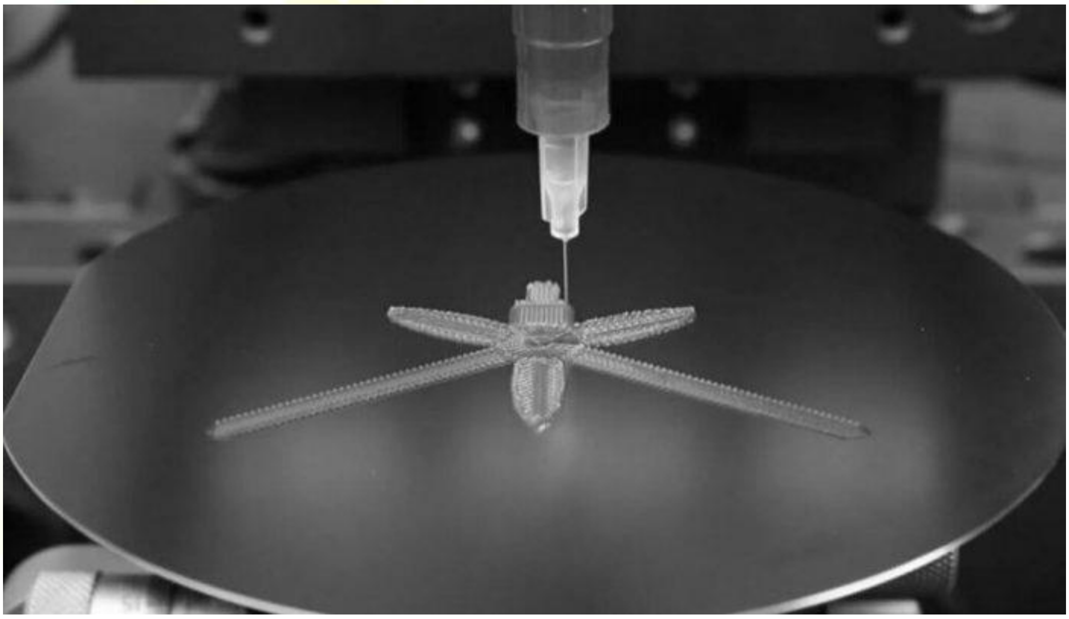


Fig. 1. 4D printing model

4D printing materials

4D printing relies on the use of specialized materials referred to as “smart materials.” These materials exhibit the remarkable ability to modify their properties in response to various external stimuli, such as heat, pH levels or light exposure. Smart materials exhibit a range of unique behaviours, including self-assembly, self-healing, shape memory and the capability to adapt autonomously. Furthermore, they can undergo changes in characteristics like colour and flavour [10]. The smart materials employed in 4D printing typically fall into two categories: single materials, such as shape memory polymers (SMP) or liquid crystal elastomers (LCE) and metamaterial composites like hydrogels.

Shape memory polymer (SMP)

When exposed to appropriate stimuli, polymeric materials used in 4D printing can experience transitory changes in shape and structure before returning to their original form. Shape memory polymers (SMPs), in particular, offer several advantages compared to other printing materials, including enhanced recovery capabilities, lightweight properties, ease of processing, superior shape deformability and cost-effectiveness. SMPs exhibit time-dependent dynamic changes, returning to their pre-programmed shape or their initial design when exposed to certain stimuli, a phenomenon known as 4D printing [11]. Among the SMPs widely used in 4D printing, polylactic acid is the most regularly used, although other materials such as bisphenol and poly cyclo-octene are also used [12]. Thermo-responsive SMPs, characterized by their high adjustability in transition temperature, optical and mechanical properties and ease of triggering shape memory, are the subject of extensive research. SMPs are often printed in a distorted condition during the 4D printing process, which is done by exposing them to temperatures greater than their transition temperature. They are then cooled to a temperature below the transition temperature before being unloaded. The recovery step involves the SMPs regaining their pre-programmed shape due to entropic elasticity when exposed to temperatures above the transition temperature [10]. Polymeric materials utilized in 4D printing can undergo temporary changes in their shape and structure, reverting to their original form when subjected to specific stimuli. Shape memory polymers (SMPs), in particular, offer several advantages compared to other printing materials, including enhanced recovery capabilities, lightweight properties, ease of processing, superior shape deformability and cost-effectiveness. SMPs exhibit time-dependent dynamic changes, returning to their pre-programmed shape or their initial design when exposed to certain stimuli, a phenomenon known as 4D printing [11]. Among the SMPs commonly employed in 4D printing, polylactic acid stands out as the most frequently used, while other materials like bisphenol and poly cyclo-octene are also utilized [12]. Thermo-responsive SMPs, characterized by their high adjustability in transition temperature, optical and mechanical properties and ease of triggering shape memory, are the subject of extensive research. In the 4D printing process, SMPs are typically printed while in a deformed state, achieved by exposing them to temperatures higher than their transition temperature. Subsequently, they are cooled to a temperature below the transition temperature before unloading. The recovery step involves the SMPs regaining their pre-programmed shape due to entropic elasticity when exposed to temperatures above the transition temperature [10].

Liquid crystal elastomers

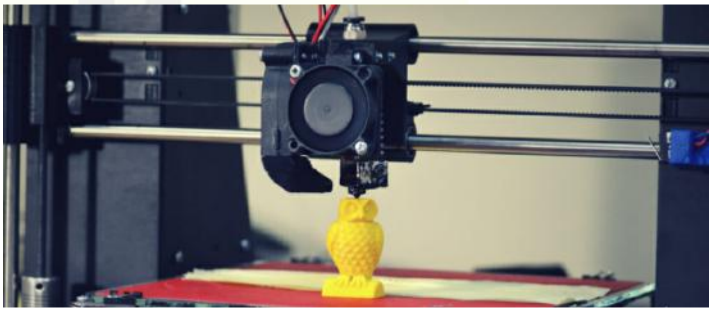
Liquid crystal elastomers (LCEs) represent another class of intelligent materials employed in 4D printing, displaying properties that combine the ordered structure of solid crystals with the fluidity characteristic of liquids. In general, LCEs achieve their anisotropic characteristics by incorporating mesogenic moieties within the polymer network, either in the chain's backbone or as side groups. The most frequently used precursor materials for synthesizing LCEs include poly (hydrosiloxane) polymers, acrylates and methacrylates [13]. LCEs are recognized for their responsiveness to various environmental stimuli, making them reversible, anisotropic shape-changing materials. These materials can undergo shape transformations in response to external factors such as changes in temperature, exposure to light or the influence of electric and magnetic fields. When LCEs containing liquid crystal molecules, also known as mesogens, experience an external stimulus, they can undergo substantial contraction along the direction of the mesogens or nematic director. This results in microscopic contraction at temperatures above the transition point and elongation below the transition temperature [14].

Composite hydrogel

Hydrogels, like shape memory polymers and liquid crystal elastomers, are widely employed in 4D printing. These materials consist of a 3D network of hydrophilic substances capable of absorbing moisture without dissolving. Hydrogels exhibit remarkable printing capabilities, enabling the creation of diverse structures that can fold, bend, stretch and geometrically expand. However, they have a slow response rate when reverting to their original state. To address this, hydrogels need to be programmed to introduce anisotropy, enhancing their swelling properties [10]. Researchers have harnessed the properties of hydrogels to develop shape-changing materials. 4D printing with hydrogels is typically carried out using light-based or extrusion-based printing techniques. Light-based methods leverage photopolymerization to convert monomers in the printing ink into hydrogels. Stereolithography and digital light processing are commonly used variants employing laser beams and projectors to initiate gel formation. In extrusion printing, conditioned gel is pushed through a nozzle, with deposition assisted by forced air or mechanical means. Multiple extruders enable the simultaneous deposition of various compounds and the creation of multi-coloured products, making it a faster process. However, one challenge with extrusion printing is the low polymerization kinetics, which may affect the dimensions of the printed object.

Hydrogels can be categorized into active and passive responsive types based on their stimuli-responsive mechanism. Active responsive hydrogels change in volume or phase when exposed to a stimulus, primarily due to molecular changes and rearrangements of polymer chains and networks [15]. In a specific case, a 4D printed alginate and methylcellulose shape morphing hydrogel was developed by controlling network density gradients perpendicular to the patterned strips. This 4D structure comprises patterned 2D structures with anisotropic stiffness and swelling characteristics. The shape morphing results from non-uniform internal stress, leading to localized uneven swelling or shrinkage of the hydrogels. This technology allows for variations in compounds across the hydrogel's thickness, plane or both thickness and plane, enabling the creation of complex shape-morphing hydrogels [9].

Fig. 2. 4D printer



4D bioprinting

4D bioprinting is the process of creating 3D biocompatible materials or live biological structures that alter dynamically over time. This advancement entails the purposeful alteration of printed items by including stimulus-responsive biomaterials or cells. 4D bioprinting holds promise for applications in in-vivo experiments, enabling the development of distinct structures after printing, responsive to environmental changes. Additionally, it is being explored in the fields of tissue engineering, drug delivery and wound repair [16].

4D printing in food

MIT researchers were the first to use 4D printing in the food sector, creating a 2D film using starch, cellulose, and protein as the triggering agent, when exposed to water, this 2D film turned into a 3D shape [17]. The advent of 4D food printing holds the potential to customize food products and craft distinctive, unique flavours. The stimulus-induced alterations in 4D printed food can be achieved by utilizing various combinations of food materials in the printing ink, guided by the structure and specific food formulation [18]. These variations in 4D printed food might include changes in colour, texture, fragrance, and form in response to stimuli such as temperature and pH. When microwave heating was used as the stimulus, for example, a 4D change was seen in 3D printed buckwheat bread and lotus root powder gel. Similarly, pH-induced colour changes in 3D printed mixes of soy protein isolate, pumpkin, and beetroot have been described.

Inks in 4D food printing

In the realm of 4D food printing, the components utilized for printing are commonly referred to as “printing ink” [19]. The printability of food printing ink is a combination of rheological characteristics necessary for smooth extrusion and mechanical strength to preserve mass and texture throughout the printing process [20]. The ink’s printability also influences aspects like extrudability, flowability and post-printing stability of the resulting structure. Importantly, the food ink used for printing should have both liquid-like and solid-like properties before and after printing. Researchers investigated the effect of particle size on the printing performance of food inks. They discovered that bigger particles, measuring 307 m and 259 m, are more permeable than smaller particles, which generally measure up to 172 m. The bigger particles form a skeletal, cellular structure. Furthermore, the particle size of the printing ink can impact the density, oil content, water absorption capacity, and other physical and functional features of the printed item [20]. Using printing technology, a diverse range of food products has been produced in recent years. Food components utilised in the manufacturing of food ink include a wide range of foods such as chocolate, meat, starch, fruits and vegetables, and food hydrocolloids [6]. The moisture content in the food material used as food ink plays a significant role in determining its printing performance. Hence, utilizing dried and powdered forms of food materials is beneficial for preserving their nutritional value and functional properties [20].

In 4D food printing, the ink mixture comprises specific materials known as stimulus-response substances. These compounds undergo changes in colour, flavour, texture and more under particular conditions or stimuli. Consequently, the sensitivity of these stimulus-response materials within food printing ink is pivotal in bringing about 4D transformations in printed objects. Pigments present in the printing ink have the ability to induce colour changes in food in response to shifts in pH levels. Curcumin, for example, is a famous stimulus-responsive substance utilised in 4D printing, with a red hue in alkaline pH and a yellow hue in acidic or neutral pH. Another chemical with stimulus-response capabilities is anthocyanin, which changes colour in response to pH variations [2]. Furthermore, a complex composed of gelatin, gum Arabic, and flavour oil has shown thermal responsiveness and is used as a stimulus-response material in the printing of buckwheat dough with yellow flesh peach. Researchers noted simultaneous changes in both colour and flavour in the product under microwave stimuli. To ensure the desired 4D changes in printed objects are controlled, careful design and distribution are crucial. Controlled changes in the properties of 4D printed items under various stimuli can be achieved through strategic coding and the appropriate distribution of their components, be it uniform, gradient or patterned [21].

Printing software

The concept behind 4D food printing research involves the creation of a 3D model that integrates various aspects such as material, structure, stimulation signals, and more. This model is then used to simulate the printing process, accounting for potential deformations, colour changes, and other characteristics. The goal of this simulation is to optimise the 3D model. When the multi-information optimised 3D model is complete, it is cut into slices and slicing software is used to direct the printer during the printing process. Following the production of the printed item, researchers study acceptable methodologies and tools for measuring the controlled adjustments obtained. In essence, the performance of the printing software, simulation software, modelling software, and slicing software all have a significant impact on the quality of 4D food printing.

Currently, 4D intelligent software largely includes forward and reverse design methodologies. The purpose of forward design is to determine the final modifications based on material structures, material qualities, and stimulation variables. This is foundational study, with a focus on discovering fundamental concepts, hypotheses, and linkages. In contrast, reverse design is centered on biomimicry, which draws inspiration from the traits of plants and animals. It, for example, regulates tissue composition and cell wall mechanics to mimic natural phenomena such as how flowers respond to external stimuli such as heat, water, and light. One example is the creation of responsive hydrogels from pectin, which is present in plant vascular tissues. These hydrogels change volume in response to variations in ion concentration.

Conclusion

To summarize, 4D printing is an intriguing prospect in a variety of sectors, including materials research, bioprinting, and the food business. This technique enables dynamic alterations in printed things, opening up a variety of applications and possibilities for creativity. In 4D printing, the choice of materials is crucial, with shape memory polymers, liquid crystal elastomers and hydrogels being key players in creating responsive structures. These materials undergo changes under different stimuli, such as temperature, pH and light, enabling the production of dynamic and adaptive objects. The food industry, in particular, has leveraged 4D printing to create unique and customizable food products. Stimulus-responsive materials within food ink, like curcumin and anthocyanin, can lead to changes in colour, flavour and texture, allowing for a more interactive and personalized dining experience. However, it's essential to carefully control and design these 4D changes to ensure the desired outcomes. Proper distribution and coding of components are crucial in achieving predictable and controlled transformations. As 4D printing continues to evolve and expand its applications, it holds the potential to revolutionize various industries, offering new avenues for creativity, customization and problem-solving. Whether in materials science, bioprinting or the culinary world, 4D printing is set to shape the future of design and manufacturing.

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