Advances in Food Science and Human Nutrition (2024)
Clausius Scientific Press, Canada

DOI: 10.23977/afshn.2024.060112
ISSN 2616-258X Vol. 6 Num. 1

Research Progress of Sustained-Release Hydrogels Based on 3D Printing Technology in the Food Industry

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Keywords: 3D printing, hydrogel, slow-release function, food industry, fermentation

Abstract: 3D printing technology is a new manufacturing technology based on digital model files, which constructs objects by printing layer by layer. As an advanced manufacturing method, it can realize the rapid prototyping of complex structures. Hydrogel prepared by 3D printing technology has a slow-release function, which can control the release rate of active ingredients (such as nutrients and probiotics) wrapped in it, and realize more accurate nutrition delivery or function play. Hydrogels come from a wide range of materials, including natural polysaccharides (such as sodium alginate and chitosan), protein materials (such as gelatin) and synthetic polymers. At present, 3D printed hydrogels have many important applications in functional food, fermented food, food structure customization and food packaging. In this paper, the material types of 3D printed hydrogels and their current applications in food industry are deeply summarized, aiming at providing valuable reference for the further application of 3D printed hydrogels in food industry and promoting the development of food industry in the direction of intelligence, personalization and high efficiency.

1. Introduction

3D printing technology is a method to build objects by stacking materials layer by layer through digital models [1]. It can transmit the designed model data to the printer, and the printer will stack the materials layer by layer, and finally form the required objects. This technology has been widely used in aerospace, biomedicine, automobile manufacturing and other fields [2-3]. In recent years, 3D printing technology has gradually penetrated into the food industry, including the process of producing chocolate, dough, cheese, fruit slices, hydrogel and other products, customized food, special medical food, cell cultured meat and so on [4-5].

With the continuous improvement of people's requirements for nutrition and health, foods containing bioactive components are sought after more and more [6]. In the food industry, the precise control and slow release of active substances and processing AIDS is a key research field. This directly affects the functionality, nutritional value, preservation effect and safety of food. However, this process faces many challenges, including the solubility, stability and bioavailability of active substances, as well as the residual control and slow release of processing AIDS.

In view of these problems in food industry, molecular embedding can protect the stability of food functional factors and improve the accessibility [7]. For example, embedding with edible coating materials can improve the resistance of functional factors to environmental adverse conditions [8]. However, the absorption efficiency of embedding with conventional sustained-release carriers is still low, which leads to the unsatisfactory function of food active substances. With the advantage of 3D printing technology, the fine design of sustained-release hydrogel structure can be realized through its layer-by-layer construction characteristics. By adjusting the pore structure, cross-linking density and distribution of active substances in hydrogel, the release rate can be accurately controlled, and then various biological or edible materials can be prepared.

To sum up, the application of 3D printing technology to hydrogels not only improves the manufacturing accuracy and flexibility of sustained-release hydrogels in food industry, but also provides strong support for the development of new food products with higher nutritional value and health benefits. In this paper, the research progress and application of sustained-release hydrogel of 3D printing technology in food industry are reviewed, aiming at promoting its application in industrial development.

2. Slow-release hydrogel material and preparation method thereof

Sustained-release hydrogel is a kind of polymer material with three-dimensional network structure, which can absorb a lot of water and swell in water, but is insoluble in water, and is often used in sustained-release systems [9]. Different synthesis conditions and material composition of hydrogel will directly affect the release rate of embedded substances in hydrogel. For example, hydrogels with high crosslinking degree usually show a slower release rate, while hydrogels with low crosslinking degree may show a faster release kinetics [10].

2.1. Chitosan based hydrogel

Chitosan is a natural cationic polysaccharide extracted from chitin, the second most extensive natural polysaccharide. As a hydrogel material, Chitosan has a good three-dimensional structure. It can swell into gel under acidic conditions, which can delay the release of drugs [11], and has biocompatibility, non-toxicity and good slow-release ability, and is widely used in medicine, food, water treatment and biomedical engineering [12].

In Henriksen et al.' s research, Chitosan and Chitosan malate were used as excipients, and three model drugs were used to study the release of Chitosan granules with different qualities [13], which successfully delayed the drug release rate. Chitosan base has the opposite effect by enhancing the dissolution of active compounds [14], that is, it is stable in alkaline medium, and the solubility of chitosan is affected by PH, so its application in vivo is limited. At the same time, the instability of mechanical properties has become a key factor restricting its wide application [15].

2.2. Alginate hydrogel

Alginate is mainly extracted and transformed from seaweed, and calcium alginate hydrogel can be obtained by Ca2+ crosslinking, which has the advantages of easy gelation, biocompatibility and non-toxicity, and has been applied in many fields [16]. Such as cell fixation, tissue engineering, drug delivery, controlled release and food applications [17-19]. It is used as a stabilizer and thickener in food [20], and it is one of the common raw materials in "popping beads" milk tea. Studies have shown that the drug capsules prepared by embedding insulin in calcium alginate have a good effect on the treatment of diabetic patients [21], but the gel formed by calcium alginate, under acidic conditions, the metal ions in the gel ball are replaced with H+, which leads to the weakening of the stability of the gel ball [22].

Because of the existence of hydroxyl, carboxyl and other functional groups, alginate can also be compounded with other substances to form alginate composite gel. The preparation methods of composite gel include grafting method, sol-gel method, embedding method and one-step synthesis method. Alginate can improve its adsorption site, stability, mechanical properties and elasticity after being compounded with other substances [23]. However, in practical application, the blending of alginate with other polymers may affect its degradation rate and pathway [24].

2.3. Gelatin hydrogel system

Gelatin is a kind of protein biomaterial, which is easy to obtain and decompose. Gelatin hydrogel is a mixture of peptide and protein, which can be obtained by denaturation and partial hydrolysis of collagen extracted from connective tissue, skin and bones of animals. By adjusting the PH value of the solution to neutral or slightly acidic, the best solubility of gelatin and the stability of gel formation can be ensured. After heating and cooling, the final crosslinking reaction forms a three-dimensional network structure with solid-like rheology. Water is entangled in its porous mesh and gradually protrudes to assemble a layered cross-linking arrangement [25], thus forming a gelatin hydrogel system.

Gelatin hydrogel has good elasticity and toughness, and is suitable for manufacturing artificial tissues and drug delivery systems [26]. At the same time, gelatin hydrogel can be used as drug carrier, and the drug release rate can be controlled by adjusting its crosslinking density and molecular weight [27]. However, gelatin biomaterials have poor mechanical properties and slow degradation rate.

2.4. Starch-based hydrogels

Starch-based hydrogels is a network structure formed by dissolving starch in water and cross-linking reaction. The preparation methods of starch hydrogel are divided into two methods: chemical crosslinking and physical gelation. Chemical crosslinking method is suitable for pursuing high crosslinking degree and drug loading capacity, while physical gelation method is suitable for scenes with higher requirements for biocompatibility [28]. In Maniglia et al.'s research [29], ozone oxidation starch technology was applied to the preparation technology of starch-based hydrogels, which improved the printability of hydrogels. Gels made of native and oxidized cassava starch can be printed in 3D at a certain temperature [30]. Drying and heating treatment can improve the viscosity, structural strength and compressive strength of wheat starch, and reduce dehydration shrinkage, making hydrogel harder and more stable, which is beneficial to improve the quality of 3D printed products [31].

In addition, frost-resistant starch materials are also suitable for 3D printing because of their excellent mechanical properties and water retention capacity. Using potato and corn starch, combining different proportions of amylose and amylopectin, and adding calcium chloride as gelling agent, an antifreeze hydrogel can be formed at room temperature. This hydrogel can still maintain its strength and flexibility at low temperature, which is convenient for 3D printing of complex structures. However, specific temperature and time requirements may affect production efficiency, and the use of calcium chloride increases the cost [32].

2.5. Nanocellulose (CNC) composite hydrogel

Nano-cellulose is an ultra-fine fiber, less than 100 nm. It has a wide range of sources and is the smallest physical structural unit of cellulose. At present, the preparation methods of nanocellulose include chemical method, mechanical method and biological method, and the most commonly used method is chemical method [33]. Nano-cellulose has good dispersibility in water and can form a relatively stable suspension [34]. CNC was dispersed in water by high shear mixer at different concentrations, and hydrogels with different viscoelastic properties were formed, which showed obvious shear thinning behavior at high shear rate, which was beneficial to extrusion through fine nozzles and suitable for 3D printing [35]. The use of nano-cellulose in the construction of hydrogel can improve the mechanical strength and has broad prospects in the field of sustained release. For example, when used as a drug controlled release material, nano-cellulose can controllably release the drug embedded inside after being implanted or injected into a living body, thus improving the therapeutic effect of the drug. In nanocomposite hydrogels, nanocellulose can cross-link with polymer molecular chains, thus enhancing the properties of composites in all aspects [36].

2.6. Other composite hydrogels

Because of the limitation of single material, many researchers began to prepare hydrogels with composite materials, such as loading drugs into physical-chemical cross-linked double-network hydrogels based on hyaluronic acid by physical or chemical means, and adjusting the drug release rate by controlling the swelling and diffusion characteristics of hydrogels [37]. Based on sturgeon surimi (URSS), composite hydrogels were prepared by adding polysaccharide colloids (including κ -carrageenan, konjac gum, xanthan gum, guar gum and sodium alginate) from different plants, which showed good 3D printing performance, kept the designed geometric shape and improved the printing quality and accuracy [38].

3. Application of sustained-release hydrogel in food industry

3.1. Functional food field

Functional food is designed to meet specific health needs, including all kinds of foods rich in nutrients or with specific physiological activities. The nutrients in it generally have the characteristics of strong pertinence, high activity and special release conditions. How to maintain the activity and stability of nutrients during processing, storage and digestion has become a noticeable factor limiting its application. 3D printing hydrogel technology can focus on the delivery of nutrients through personalized nutrition, and package and release nutrients slowly, which provides a new way to solve this kind of problem. The combination of microcapsule technology and 3D food printing technology has also brought many new possibilities and opportunities for the food industry [39].

Lu Zhang, Yimin et al. printed the dough containing probiotics into two structures with different surface to volume ratios. The experimental results show that 3D printing is helpful to change the microstructure of food, improve the survival rate of probiotics, and may be beneficial to the development of innovative food containing functional ingredients [40]. Li et al. used polysaccharide-based high internal phase emulsion for nutritional encapsulation and 3D printing to enhance the stability and accessibility of curcumin. Rosas-Val et al. developed a formula of microencapsulated probiotics based on 3D printing technology, which was used for oral delivery to ensure the delivery of a specific dose of live bacteria in the large intestine to achieve the expected biological effect. These studies have jointly promoted the new progress of functional food in nutrition maintenance, stability and personalized demand satisfaction of consumers.

3.2. Fermentation engineering field

The application of sustained-release hydrogel in the field of food fermentation engineering is mainly reflected in its ability as a carrier to control the growth and metabolic activities of microorganisms during fermentation, thus improving the quality and production efficiency of fermented food.

Through 3D printing technology, such as photo-curing 3D printing technology, precise control of hydrogel structure can be achieved, and specific functional requirements can be provided for fermentation process by adding functional materials. The research team of the University of Washington has made a breakthrough in this field. They developed a bioreactor prepared by 3D printing technology, which combined hydrogel lattice with yeast and could continuously ferment in glucose solution, simulating the fermentation process in brewery environment. This innovation not only realized the long-term activity maintenance of yeast in several months, but also realized the rapid manufacture of the reactor through the combination of low-cost chemicals and 3D printers. This continuous fermentation method provides a new idea for industrial production, which is expected to replace the traditional batch processing mode and bring about a significant improvement in production efficiency and flexibility.

Improving the rheological and textural properties of yogurt by adding hydrocolloids such as gelatin can not only improve the gel strength of yogurt, improve its elasticity and resilience, but also help to maintain the printing form in the 3D printing process and reduce the seepage of water, thus improving the stability and quality of printed products. In addition, the slow-release hydrogel can also be used for packaging and delivery of fermented food. By adjusting the pore size and cross-linking density of the hydrogel, the controlled release of fermented products such as organic acids and alcohols can be achieved, the shelf life of fermented food can be extended, and its nutritional value and flavor characteristics can be improved.

3.3. Food structure customization field

Customized food structure can improve the freshness and shelf life of food, and can meet the specific nutritional needs of individuals, help people better manage their health and provide more personalized nutritional support, especially for people with special health needs. In 3D food printing technology, sustained-release hydrogel can control the nutrient release rate by adjusting its composition and crosslinking density.

Double cross-linked systems such as carboxymethyl cellulose/sodium alginate double cross-linked hydrogel provide additional mechanical strength and stability, which are suitable for biomedical fields and can also be used for customized nutritional supplements. In addition, in the research of Zhang et al, various applications of natural polymers such as hydrogels in food industry were discussed, such as customization of food structure and slow release of functional components . In the research of 20Park et al, 3D printing technology based on callus was used. Especially using carrot tissue as an example, it shows the potential of 3D printing in food manufacturing.

3.4. Food packaging field

Food packaging can limit the loss of nutrition and flavor and extend the shelf life of food. Specific functions include reducing the interaction between water and food (preventing moisture), avoiding bacterial invasion and food spoilage, and maintaining the structural integrity and biodegradability of food. Using hydrogel as an intelligent food packaging system is a solution that has attracted much attention.

Nancy Jun and her team prepared Chitosan-based hydrogels by reacting Chitosan with cross-linking agents, and found that they have antioxidant and antibacterial properties and are suitable for the preservation of meat, fruits and vegetables. Wang Hao and his team found that adding konjac gum can significantly improve the 3D printing characteristics of blueberry gel system and improve its printing accuracy and structural stability, so they developed PVP-CMCH hydrogel films modified by bacterial cellulose and guar gum, which were used for packaging fresh fruits such as blueberries. To sum up, 3D printed hydrogels have shown great potential in the field of food packaging, especially in extending the shelf life of food and improving food safety. However, the hydrogel used in smart food packaging is still in the experimental stage, and it needs appropriate raw materials, reuse rate and sensitivity to realize industrialization.

3.5. Food nutrition field

Through 3D printing technology, researchers can develop more innovative and diversified food products to meet people's needs for various nutrients. The addition of sustained-release hydrogel can further study the taste and flavor of food and improve the consumer experience of food.

Zhang Lu and his team made sustained-release hydrogel microcapsules by 3D printing technology for sustained-release vitamins and minerals in beverages. This method improves the bioavailability of nutrients while maintaining the taste and flavor of the beverage. Li et al. used 3D printing technology to make a biscuit containing sustained-release hydrogel for sustained-release protein and vitamins. This kind of biscuit can not only provide better nutrition, but also improve the taste and texture of food. These studies show that the combination of 3D printing technology and sustained-release hydrogel materials in the field of food nutrition can provide consumers with more abundant and diversified nutrition choices and improve the overall experience of food.

4. Application and limitation of food industry

The application of 3D printing technology in the food field, especially the printing of sustained-release hydrogels, has brought the possibility of innovation to the food industry and has many advantages in the food industry, but there is little research on developing technologies that can realize large-scale production systems.

Printing of sustained-release hydrogels requires specific materials. These materials usually require good biocompatibility, edibility, and stable structure and performance during printing. However, the types of food-grade hydrogel materials available for 3D printing are limited, and the preparation process is relatively complicated. In addition, not all food ingredients are perfectly compatible with hydrogels, and some food ingredients may interact with hydrogels, resulting in changes in food taste and nutritional value. The printed food with sustained-release hydrogel may be different from traditional food in taste and flavor. In the application of 3D printing, 3D printing of sustained-release hydrogel requires high-precision printing equipment and technology to ensure that the printed food has the required shape, structure and performance. However, due to the viscosity and fluidity of hydrogel, the printing process is prone to problems such as material breakage, excessive discharge or poor molding effect. At the same time, the printing speed of 3D printing technology is relatively slow, which may increase the production time cost. At present, the relevant regulations and standards on 3D printed food are not perfect. This may lead to legal risks and compliance problems in the production and sales process.

5. Prospect

At present, the application of 3D printing technology in food industry is more and more. In this paper, the research progress of sustained-release hydrogel of 3D printing technology in food industry is reviewed, in order to provide reference for the application of 3D printing hydrogel in food industry. The slow release hydrogel based on 3D printing combines the advantages of 3D printing technology and hydrogel materials to prepare hydrogel products with complex structures and functions, which can be used in all aspects of the food industry. 3D printing technology makes the preparation of hydrogels more accurate and flexible, and can meet the needs of different applications. It is believed that with the development of technology research, universality and industrialization will be realized quickly.

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