



Investigation on lemon juice gel as food material for 3D printing and optimization of printing parameters



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ABSTRACT

The aim of this paper is to develop a new 3D printing food constructs based on lemon juice gel system. We investigated the effect of potato starch (10, 12.5, 15, 17.5 and 20 g/100 g) on the rheological properties and mechanical properties of lemon juice gels. Besides, the influence of printing parameters (nozzle height, nozzle diameter, extrusion rate and nozzle movement speed) on the quality of printed products were also studied. The results show that it is suitable to make the size of the nozzle height the same with that of the nozzle diameter, which could not be regarded as a key factor that affects print quality. An equation is proposed to explain the relationship between extrusion rate, nozzle diameter and nozzle movement speed. In this printing system, the 1 mm nozzle diameter, 24 mm³/s extrusion rate and 30 mm/s nozzle movement speed were found to be the optimal parameters to print 3D constructs matching the target geometry with fine resolution, more smooth surface texture, and fewer point defects with no compressed deformation.

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1. Introduction

3D printing, also known as additive layer manufacturing, is a type of rapid prototyping technology, which involves the integration of computer, direct writ systems, precision transmission, numerical control technologies and material science. Although it was invented in 1980s, only in recent years does it develop rapidly. In the printing process, the material is generally extruded through a nozzle whose position is computer-controlled in accordance with a shape design model. For the production of a product of a particular shape, the traditional process usually requires a model to be produced, while 3D printing eliminates the process. Therefore, compared with the traditional technology, 3D printing has the advantages of time-saving, simple operation and customizable in the whole manufacturing process. In addition, 3D printing provides the possibility to print complex objects with an internal structure, and users have a substantial liberty of design or download their

favorite models. In recent years, 3D printing has been widely applied in many fields, such as machinery, biomedicine, polymer, food technology and so on (Chia & Wu, 2015; Lipson & Kurman, 2013). Food 3D printing is expected to be a breakthrough in the popularization of 3D printing industry. Food 3D printing may be an important direction of future food processing. Since the food is closely related to people's lives, consumers will intuitively understand the 3D printing through food (Godoi, Prakash, & Bhandari, 2016; Sun, Zhou, Huang, Fuh, & Hong, 2015).

3D printing provides a new frontier in food processing, helping us to realize and produce new foods with complicated shapes using particular material formulation mixtures and potentially enhancing its nutritional value (Pallottino et al., 2016). Some food materials such as chocolate, dough and meat paste has been utilized to print 3D object (Hao et al., 2010; Wegrzyn, Golding, & Archer, 2012). Sometimes, the printed objects need to be processed or cooked post-printing. Lipton, Cutler, Nigl, Dan, and Lipson (2015) used transglutaminase and bacon fat as additives to make printable scallop and turkey meat-puree. These final meat products well kept their shape after cooking (Lipton et al., 2010). The researchers also investigated the effect of variations in the amount of butter, yolk and sugar relative to the nominal recipe on the shape stability for

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sugar cookies after cooking. They reported that yolk concentration contributed to the stability in the X direction (Lipton et al., 2015). Severini, Carla, Derossi, and Antonio (2016) analyzed the first examples of 3D food printing available in literature and reported the production of 3D printed wheat-based snacks enriched with insect powder (*Tenebrio molitor*) with the aim to improve the quality and the content of proteins (Severini et al., 2016).

The properties and composition of materials have been considered to be the most important factors in 3D printing process. These materials should be homogenous and have appropriate flow properties for extrusion as well as can support its structure during and after printing process (Godoi et al., 2016; Shao, Chaussy, Grosseau, & Beneventi, 2015). Zhang et al. (2015) used dual-responsive hydrogels to fabricate three-dimensional objects via extrusion from a nozzle in 3D printing. They found gels with a rapid and reversible modulus response to shear stress and to the temperature were suitable for direct-write 3D printing as they were easily extruded out from nozzle tip during printing and could maintain sufficient mechanical integrity to support the next printed layer without deformation (Zhang et al., 2015). Other researchers have come to a similar conclusion (Shao et al., 2015; Wang, Zhang, Bhandari, & Yang, 2017).

Some researchers have also investigated the effects of printing parameters on the geometrical accuracy and dimension of food pattern. Hao et al. (2010) demonstrated a linear relationship between the extrusion rate used in the ChocALM software and the bead diameters obtained. However, these studies do not explicitly account for the relationship between print parameters.

The main aim of this work is exploring the opportunity of 3D printing of a lemon juice gel. Lemon gel can be regarded as a kind of gel-type fudge which is translucent, flexible and chewy. Potato starch has good water retention capacity, transparency and aging resistance properties (Zhang, Zhang, Yang, & Chen, 2001). Thus, in this research starch was chosen as a gelling agent. Printing size parameters such as nozzle diameter size, nozzle height, nozzle movement speed, extrusion rate and material properties (Rheological properties, moisture orientation and texture properties) on the final qualities of the 3D constructs were investigated.

2. Materials and methods

2.1. Raw materials

Lemon juice was provided by Jiahao Co. Ltd. Guangdong, China and stored at 4 °C. The moisture content of the lemon juice was 59.82 g/100 g as determined by the vacuum-drying method. The pH of lemon juice was 2.28. Potato starch were purchased from Shanghai Tianyu Food Co. Ltd. The moisture content was 13.47 ± 0.4 g/100 g, 97.5 ± 0.5 g/100 g purity with amylose and amylopectin ratios of 25.7 ± 0.2 g/100 g and 74.3 ± 0.2 g/100 g respectively. Starches were stored at normal atmospheric temperature.

Lemon juice was firstly mixed with different starch content (10, 12.5, 15, 17.5, 20 g/100 g). Afterwards, the mixture was completely homogenized by a mixer (ULTRA-TURRAX® IKA® T18 basic, Model: T18BS25, Germany). Then the samples were moved to glass containers and steam cooked for 20 min (Center temperature 86 ± 2 °C). During the cooking process, the container was wrapped with food grade plastic protective film to prevent the water loss. Finally, the sample was cooled to room temperature to form a weak gel-like structure and stored at 4 °C. Then the nuclear magnetic resonance (NMR) analysis, rheological properties, texture profile analysis (TPA), to determine the status of water and printing behavior of stored sample was investigated.

2.2. Testing of material properties

2.2.1. Low-field nuclear magnetic resonance (NMR) analysis

As water distribution and condition have a close relationship with the material structure and rheological properties, thus a low field pulsed NMI 20 analyzer (Shanghai Niumag Corporation, China) at 22.6 MHz was used in this experiment. About 5 g sample was chosen and packed with a thin layer of plastic film. Then they were put into a 10-mm glass tube, and the NMR probe was inserted into the analyzer. Carr–Purcell–Meiboom–Gill (CPMG) sequences were used to measure spin-spin relaxation time T₂. Each measurement was performed for three times.

2.2.2. Rheological properties measurements

Rheological measurements of samples were tested by a hybrid rheometer (Discovery HR-3, DHR, TA Instruments, USA) with a parallel plate (diameter = 20 mm). The samples were allowed to rest for 2 min after loading and the temperature was maintained at 25 °C. Viscosity and shear stress measurements were carried out for all samples in the shear rate ranging from 0.1 to 100 s⁻¹. Shear stress, shear rate, and steady shear (apparent) viscosity (η) were recorded by a RheoWin 4 Data Manager (Rheology Software, Thermo Fisher Scientific, Waltham, MA). The relevant responses of the samples were recorded as functions of shear rate.

Dynamic viscoelastic properties were characterized using small-amplitude oscillatory frequency sweep mode. The frequency was oscillated from 0.1 to 100 rad/s, and all measurements were performed within the identified linear viscoelastic region and made at 0.4% strain. The elastic modulus (G'), loss modulus (G''), and loss tangent ($\tan \delta = G''/G'$) were recorded. Experiments were conducted in triplicate for each type of sample.

2.2.3. Texture profile analysis

A texture analyzer (model TA-XT2, Stable Micro System Ltd., Leicestershire, UK) with was used to assess the instrumental texture properties of different samples (2 cm cube printed using 1.0 mm diameter nozzle). The instrument was calibrated with a 1 kg load cell, then fitted with a flat-ended aluminum probe of diameter 25 mm (Code P/25, Stable Micro System Ltd.). The parameters were presented as follow: pretest speed 5 mm/s, test speed 1 mm/s, post-test speed 5 mm/s, trigger force 5N, at room temperature (25 ± 1 °C). All tests were repeated three times.

2.3. Printing process

The 3D printing system composed of the following three major parts: (i) feed hopper with auger mixer and conveyor, (ii) an extrusion system, (iii) and a X-Y-Z positioning system using stepper motors. The nozzle height from the printing bed was achieved by adjusting the whole feeding device. The pressure exerted on the sample was applied via the extruder conveying screw. The samples were extruded onto a polished transparent plastic polymer plate using nozzles of circular shape with diameters of 0.5, 1.0, 1.5 and 2.0 mm. The printing process was conducted at room temperature (25 ± 1 °C). The motion and positioning control were provided by a computer with a specifically designed Java program and micro controller.

To assess the effects on the extruded geometry, line tests and cylinder tests were used. Lines of sample were extruded at varying extrusion rates, for same movement rates to determine the appropriate extrusion speed. The cylindrical is a suitable model to evaluate the printing effect, since the printing process has been related to the simultaneous motion of the X axis and the Y axis. Cylinders of sample ($\Phi 20$ mm × 15 mm) were extruded at varying extrusion rates, for different movement rates and nozzle diameters,

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