Journal of Food Engineering xxx (2017) 1–8

Contents lists available at ScienceDirect

Journal of Food Engineering

journal homepage: www.elsevier.com/locate/jfoodeng

Design and characterisation of food grade powders and inks for microstructure control using 3D printing☆

Sonia Holland [a, b, *], Tim Foster [a], William MacNaughtan [a], Chris Tuck [b]

[a] Division of Food Science, Sutton Bonington Campus, University of Nottingham, Loughborough LE12 5RD, United Kingdom
[b] 3D Printing and Additive Manufacturing Research Group, University Park, University of Nottingham, Nottingham NG7 2RD, United Kingdom

A R T I C L E I N F O

Article history:
Received 16 September 2016
Received in revised form
5 June 2017
Accepted 7 June 2017
Available online xxx

Keywords:
Binder jetting
Amorphous cellulose
Polysaccharides

A B S T R A C T

Additive Manufacturing techniques have been previously applied to food materials with direct consumption in mind, as opposed to creating structural ingredients as shown in this study. First, semi-crystalline cellulose was mechanically treated by ball milling to render an amorphous powder, which has been characterised. Requirements for the subsequent recrystallization of this powder with a view to structuring have been determined through the control of moisture and thermal energy. Food inks based on xanthan gum have been formulated to enable successful jetting with a FujiFilm Dimatix ink jet printer. The polymer inks were subsequently jetted onto the amorphous cellulose powder to observe powder-binder interactions. Material combinations and parameters were optimised to produce cohesive geometric structures. The results of this study are promising when looking towards using these materials in a binder jetting additive manufacturing technique using designer particles and inks to create structures for use in food products.

1. Introduction

Mass customisation, geometric design freedom, low volume economy and less processing waste are key benefits that Additive Manufacturing (AM, often referred to as 3D printing) technology provides to other platforms which could be advantageous in the food industry. The application of AM to food materials is unusual due to the open source nature and hobbyist culture stemming from a relative ease to source hardware, as well as experimental interest from academic institutions, large companies, small start-ups and laymen – thus many developments are not formally documented. Key reviews by Godoi et al. (2016), Lipton et al. (2015), Sun et al. (2015) and Wegrzyn et al. (2012) provide insight into current developments by a number of parties into AM of food materials. Though material feedstocks and processes differ among the various applications they have a common thread; all end use products are designed on the macro scale for direct consumption or post process cooking by the consumer. In these instances the freedom of design complexity is being utilised but with end consumer wants in mind. More recent publications are indicative of the many facets of food printing that are beginning to emerge such as its use as an educational tool (Hamilton et al., 2017), to enable molecular gastronomic creations (D'Angelo et al., 2016), to influence food textural properties (Le Tohic et al., 2017) and even provide a cheap, sacrificial material feedstock for 'lab-on-a-chip' capabilities when combined with PDMS (He et al., 2015). Similarly, 3D Printing technology options for food materials are beginning to be discussed to suit specific processing needs (Sun et al., 2017). Food systems are complex; microstructures determining textural attributes, stability and flavour release typically arise through interaction and self-assembly of components during processing. If AM technology were to be truly exploited by the food industry it would be advantageous to achieve this intricacy on more relevant, smaller length scales within food products.

There are seven categories of AM recognised as per the ASTM standard terminology for Additive Manufacturing Technologies (ASTM International, 2015). The AM technology commonly used for food products involves material extrusion from a nozzle, therefore a limiting factor for feature size is the nozzle diameter used. Taking into account the multi-component nature of the ingredient feedstock as a potential for blockages and probable negative effects caused by pressure and shear on extrusion of the material, nozzle

☆ This work was funded as a gift from The University of Nottingham and by the Engineering and Physical Sciences Research Council [EP/I033335/2] [EP/K030957/1].

* Corresponding author.
E-mail address: Sonia.Holland@nottingham.ac.uk (S. Holland).

http://dx.doi.org/10.1016/j.jfoodeng.2017.06.008
0260-8774/© 2017 Published by Elsevier Ltd.

diameters are typically greater than 1 mm and rarely below 0.5 mm, unless feedstock homogeneity and print viscosity can be assured. Thus this is the minimum feature size for products produced; often layering adhesion sites are visible to the naked eye in the z direction.

Binder jetting is a category of AM in which a liquid binder is ink-jetted onto a powder base in cross sectional layers based on the digital design file. Once one layer is printed, a roller or blade deposits a fresh layer of powder for the next cross sectional design to be jetted on to, as depicted in Fig. 1. This sequential layering and jetting process is continued until, encased in surrounding unbound powder, the adhered structure, which is handleable, may be removed from the powder bed. Ink jet nozzles can deposit as little as 1 pL of ink per droplet, but are typically 10 pL when a 21 μm diameter nozzle is used.

The powder component is ideally bimodal or multimodal to allow for ease of spreading as well as to enhance adhesion. Smaller particles will fill the gaps created between larger particles creating more potential junction zones, ultimately reducing unwanted porosity in the finished product. Depending on the application or desired minimum feature size the d_{50} (particle diameter at 50% in the cumulative distribution) of the powder can range from 100 μm to below 20 μm. It has been suggested that use of particle blends with d_{50} between 30 and 100 μm gives higher mechanical strength than finer particles (Shirazi et al., 2015). In addition, using particles with a size range 80–150 μm, which is close to the intended layer thickness, or those below 5 μm, inducing Van der Waals mediated agglomeration, is also likely to have detrimental effects on print quality (Vorndran et al., 2015).

Binding materials must be designed in such a way that droplets ejected achieve the correct form (spherical, without a trailing ligament or non-merging satellite droplets) and at sufficient speed to keep a straight flight path, landing at the intended position on the substrate. Reis and Derby (2000) defined a parameter to assess the jettability (Z) of a given material. Z, derived from equation (1), is based on the Reynolds (Re), Weber (We) and Ohnesorge (Oh) numbers which take into account the viscous, surface tension and inertial forces on the material under jetting conditions, respectively.

$$Z = \frac{1}{Oh} = \frac{\sqrt{We}}{Re} = \frac{\sqrt{\gamma \rho L}}{\eta} \quad (1)$$

In this equation, η, γ, ρ and L represent viscosity, surface tension, density and a nominal dimension (e.g. nozzle/drop diameter), respectively. A Z-value in the 1–10 range is indicative of stable droplet production, though some researchers suggest this range may be extended. There are also other factors which affect jetting success, such as the molecular weight and concentration of particles or polymers contained in the ink (McIlroy et al., 2013; Soleimani-Gorgani, 2015). The application of binder jetting to food has typically utilised sugar and starch powder mixtures with water or alcohol based binders to produce macro scale, decorative 3D structures (3D Chef, 3D Systems and Walters et al., 2011). Although a useful scoping exercise in applying binder jetting to food materials, a diet in which consumption of sugary foods is high has been linked to numerous health risks such as obesity and type 2 diabetes and its reduction or substitution in the diet is being lobbied for by activist groups and governments globally (Edwards et al., 2016).

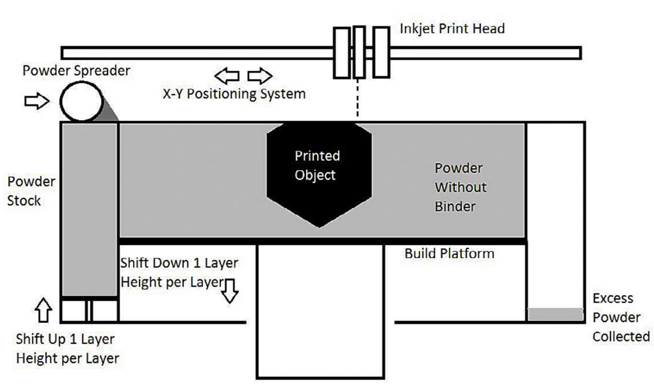

Fig. 1. Representation of the Binder Jetting Process, adapted from Gibson et al., 2010.

Cellulose is a major structural component within plant cell walls. It is comprised of β-(1–4) linked D-Glucose units arranged in a hierarchical series of chains to render a rigid, semi-crystalline, fibrous structure. Cellulose is naturally present in the human diet and consumed daily as part of the cell wall of any plant products however, humans do not possess the necessary enzymes to digest it so cellulose passes through the GI tract chemically unchanged, thus it is a 0kCal dietary fibre. Microcrystalline cellulose (MCC) is used as a bulk filling agent in food formulations and crystalline cellulose may be derivitized to create water soluble viscosity modifying ingredients such as carboxymethylcellulose (CMC) or methylcellulose (MC), these applications are not able to utilise cellulose as a structural or 'load-bearing' component as found in its primary role in the plant cell wall (Wuestenberg, 2014). Crystalline cellulose is used in the textiles industry; it is dissolved in ionic liquids and subsequently regenerated and recrystallized when exposed to an anti-solvent (such as water) to produce fibres which may then be woven together to create garments able to bear stresses (Rosenau et al., 2001). However, such cellulose solvents would not be suitable for food applications, but amorphisation and restructuring of cellulose into different architectures could be of interest to the food industry particularly as cellulose is a low cost, readily available ingredient. It is well documented that mechanical breakdown of crystalline cellulose via ball milling can render an amorphous cellulose powder which, under the correct conditions, may be recrystallized (Abbaszadeh et al., 2014b; Avolio et al., 2012; Hermans and Weidinger, 1946; Paes et al., 2010). Utilising this process could be a way in which cellulose is restructured in a food grade manner, and provides potential for use as a particle in ink-jet printing.

There is also interest in the interactive effects that structurally similar biopolymers (those with β-(1–4) backbone linkages) have with cellulose, particularly on controlling the formation of crystal allomorphs (Chanliaud et al., 2002; Gidley et al., 2002); xanthan gum is one such polysaccharide of interest and is also frequently used in food applications. Xanthan gum is a high molecular weight polysaccharide produced by the bacterium *Xanthomonas campestris* which exhibits very high solution viscosity at low concentrations, its solutions are also shear-thinning (Wuestenberg, 2014). Though in traditional printing applications an element of shear-thinning can aid the jetting process, non-Newtonian, high viscosity fluids due to the inclusion of a high molecular weight polymer are very difficult to ink jet print yet most food biopolymers fall into this category. Polymer chains unravel and stretch out under flow conditions, above a critical concentration or chain length this can result in the failure of an ink droplet to detach from the nozzle (McIlroy et al., 2013).

In this paper we demonstrate that an amorphous cellulose powder and a xanthan-based binder may be utilised in a 2D jetting process to create designed particles and structures for food use through controlled application of the binding agent and thermal energy. In a further 3D application, the moisture provided by the ink in combination with heat recrystallizes the amorphous powder,