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3D food printing: a categorised review of inks and their development

Siew Li Voon^a, Jia An^a, Gladys Wong^b, Yi Zhang^a, Chee Kai Chua^a

^aSingapore Centre for 3D Printing, School of Mechanical and Aerospace Engineering, Nanyang Technological University, Singapore; ^bNutrition and Dietetics, Khoo Teck Puat Hospital, Singapore

Contact author: Siew Li Voon, voon0007@e.ntu.edu.sg

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3D food printing, or the use of food materials in additive manufacturing, has been a topic of interest to the commercial sector for the past decade. Recently, there has also been increased attention from the academic field, with a considerable volume of research being published. Most of these studies have focused on developing food inks for extrusion-based printing beyond conventional materials such as chocolate, resulting in a much wider range of printable and edible materials. The formulations of these materials are of particular interest, as they determine the rheological and mechanical properties of food inks, thereby affecting the final quality of the printed food. This article reviews the current status and possible directions in food ink research across five categories, namely confectionery, dairy, hydrogels, plants, and meat. The roles of additives in these inks are then discussed, followed by the remaining challenges and potential opportunities in 3D food printing.

Keywords: additive manufacturing; 3D food printing; extrusion-based printing; food design; materials properties

Introduction

3D food printing or 3DFP is one of the newer applications of additive manufacturing technology, with the first academic papers in the area only having been published around 2010. The initial foods printed in these studies were chiefly easily-extrudable materials such as frosting, chocolate, cheese (Periard et al. 2007), edible hydrogels (Cohen et al. 2009), and cookie dough, with exceptions such as celery which was pureed and mixed with hydrocolloids (Lipton et al. 2010). Depending on the intended application and target audience, 3DFP has the potential to create a range of opportunities and benefits, including personalisation to individual consumer needs, automation of the food production process, possibility of novel structures and shapes, and sustainability improvements through reducing food waste or utilising alternative nutrient sources (Fan Yang, Zhang, and Bhandari 2017; Dankar, Haddarah, et al. 2018).

To achieve these goals, food materials must be printable (i.e. they can be deposited by a 3D printer and hold their structure post-deposition), applicable in terms of material properties as well as the provision of structural or nutritional benefits, and finally resistant to any required post-processing such as baking or cooking (Godoi, Prakash, and Bhandari 2016). These factors are in turn enabled by controlling rheological and other properties of the materials to match the respective printing process and parameters.

Of the seven categories in additive manufacturing identified by ASTM, four have been used successfully in 3DFP, namely material extrusion, material jetting, binder jetting, and powder bed fusion through selective laser sintering (Z. Liu et al. 2017). Among these, material extrusion has been by far the most commonly used in 3DFP research, likely because a wide range of food materials from chocolate to vegetables can be melted, blended, or otherwise processed to into extrudable forms. In comparison, other methods such as binder jetting have been used more in commercial markets or by hobbyists with little published research, due in part to restrictions on food materials that can be used, though some efforts have been made to overcome these issues (Holland, Foster, and Tuck 2019).

Though some 3DFP studies have used custom-built printers, while others have introduced modifications to enable functions such as *in situ* cooking or gelation, existing extrusion-based printers are often already suitable for food printing as is if equipped with a syringe and nozzle for paste extrusion, with some commercial printers even having been developed specifically as food printers (Tan et al. 2018). As such, the main question in 3DFP is not the printing hardware or techniques but rather the development of printing materials, which has correspondingly formed the focus of most research efforts. Despite this, key reviews on the topic of 3DFP (see for instance (Sun et

al. 2015; Godoi, Prakash, and Bhandari 2016; Fan Yang, Zhang, and Bhandari 2017)) have tended to favour a broader perspective of the field, with only a brief section on the materials used. Moreover, there has been a surge in the volume of original research within the past two years, greatly widening the scope of food materials that have been shown to be printable; therefore, there is a need to consolidate these developments so as to better understand what has been accomplished, and potentially identify areas for future study.

In this paper, an overview of current research in food inks is first presented in each of five categories identified, where “food inks” refer to the final printable form of an edible substance prepared from one or more raw materials, often in the form of a paste or gel that is ready to be extruded from a printer nozzle. This approach has been chosen because each category of food inks differs in formulation and properties, and moreover the extent of research and development varies significantly between them, from preliminary studies to well-established materials. Next, inks used in other printing methods are briefly discussed, followed by the role of additives in food inks with respect to their functions and effects, and finally some challenges and opportunities for future development in 3DFP.

Food inks for extrusion printing: classification and status

In classifying the range of food inks that have been studied, a largely ingredient-based approach has been adopted to identify five categories, namely confectionery inks such as chocolate or dough, dairy inks based on milk or its derivatives, edible hydrogel inks, as well as inks created from plants or meat. Given the overall goal of printability in 3DFP, these have been grouped into two broader categories: natively extrudable (i.e. confectionery, dairy, hydrogels)) versus non-natively extrudable (i.e. plants, meat). Natively extrudable food inks are defined as inks with favourable viscoelastic properties that allow direct extrusion without added matrix materials such as gum or binder, although these additives may still be used to improve print quality. This classification is

summarised in

Figure 1. Classification of food inks.

It should be noted that the categorisation of certain food inks may not be mutually exclusive (e.g. mashed potato was considered a plant-based ink while potato starch suspensions were categorised as hydrogels). Although this may introduce some arbitrariness at the level of individual inks, this classification aims to provide an intuitive system that is easily applicable to most food inks, rather than focus on potential overlaps between categories.

This division of natively extrudable food materials from others has also been briefly noted in earlier publications (see for example (Sun et al. 2015; Godoi, Prakash, and Bhandari 2016)). On the other hand, different ingredient-based classification systems have also been proposed, such as traditional versus non-traditional food materials (Feng, Zhang, and Bhandari 2018), or even based on components such as carbohydrate, fat, and protein (Jiang et al. 2018). While such categorisations highlight different factors of food inks including societal acceptability or nutritional content, they may not be as immediately applicable in classification of food inks, which are created in practice from ingredients rather than individual nutrient components, and furthermore

specific ingredients (e.g. insect protein) considered unconventional in some cultures might be commonplace in another.

This section first presents a discussion of these inks according to the categories outlined above, with each subsection highlighting various formulations that have been tested in that category as well as the overall focus of current studies. The former should aid in identifying the categories or particular formulations of food inks that may be of interest for further development, while the latter will provide a broad view of the progress and directions of research, which differs considerably between categories. A tabulated summary of these five categories is then presented at the end of the section.

The scope of publications reviewed has been limited to studies specifically intended to develop food inks for 3DFP; while this necessarily excludes studies targeted at more general applications, it should be recognised that such publications can provide valuable information, especially with regards to rheological or other data where similar food materials are tested.

Natively extrudable

Inks in this category are easily extruded through a nozzle, and have been the focus of most 3DFP research, especially for chocolate and hydrogels.

Confectionery: chocolate, dough

Figure 2. (a) Chocolate with plant sterol powder and different infill patterns; (b) Cookie dough (control vs. with xanthan gum) before and after baking. Reproduced from (Mantihal, Prakash, and Bhandari 2018) and (Kim et al. 2019) respectively.

Chocolate was one of the earliest materials used in 3DFP, having been first printed as early as in 2007 by Schaal (Periard et al. 2007). This was followed in 2010 by the design of the ChocALM prototype printer by a group at the University of Exeter (L. Hao et al. 2010), later developed and released commercially as the Choc Creator V1 under the Choc Edge label in 2012 (Choc Edge Ltd 2019). The subsequent years saw several other chocolate printers being released: the Choc Creator V2 and V2.0 Plus, the CocoJet from 3D Systems and Hershey's (Buhr 2015), and the Chocobot from Global 3D Labs (O'Neal 2015).

Perhaps unexpectedly, despite this commercialization, little research on chocolate printing was evidenced until the publication of three articles in 2017, respectively describing the determination of printing parameters and effect of supports and additives (Mantihal et al. 2017), the development and optimisation of a low-cost printer from open source components (Lanaro et al. 2017), and a self-monitoring system

for physical activity using printed chocolate treats with custom designs based on each user's heart rate (Khot et al. 2017).

This highlights the importance of the printing process and platform in chocolate printing, which continues to be the focus of more recent research, such as through algorithms for adaptive layer thickness and scanning speed to improve printing time and quality (Liang Hao et al. 2019). An electrostatic inkjet printer has also been developed at Waseda University, which uses a nozzle with a thin fibre of ABS resin at the tip to decrease droplet size and thus print chocolate with high precision below 50µm (Suzuki, Takagishi, and Umezu 2018). As it is capable of handling viscous materials, chocolate can be printed without any flow enhancers (Takagishi, Suzuki, and Umezu 2018) or changes to its composition, unlike sintering which would likely require a significantly lower fat content than is typical of chocolate (Lanaro, Desselle, and Woodruff 2019); however, it is debatable whether inkjet printing is truly classifiable as a 3D printing technology, as it is often used to create only simpler 2D designs (Z. Liu et al. 2017).

From a materials standpoint, while commercial milk or dark chocolate is commonly used, an area of increasing interest is the incorporation of additives. Formulations that have been tested include milk chocolate with vitamin C or cranberry powder and methylcellulose (Liang Hao et al. 2019), as well as dark chocolate with magnesium stearate or plant sterol powder as shown in Figure 2 (Mantihal, Prakash, and Bhandari 2018; Mantihal et al. 2019). These additives can not only enhance nutritional content, but also improve flow by reducing slippage in screw extrusion due to their particulate form.

Despite these benefits, Hao et al. reported a limit on the amount of additives that can be used, as excessive addition of dry powders would reduce the fat ratio and result in an excessively fluid chocolate prone to collapse. This limit was estimated to be

around 1% by weight, beyond which the addition of methylcellulose was needed to prevent collapse, although further increase in methylcellulose content again led to collapse (Liang Hao et al. 2019). However, Mantihal et al. successfully printed chocolate with as much as 5% additive content without the need for methylcellulose or other thickeners (Mantihal et al. 2019). It is not apparent whether this discrepancy is due to the difference in chocolate (milk vs. dark) and additive type (cranberry powder vs. magnesium stearate powder) or some other reason, highlighting the need for better understanding of the factors underlying chocolate printing performance, especially in interaction with other substances.

These developments provide an overview of the directions that 3DFP research and commercialisation can take for a given food ink, namely the design of specialised printers; development of new printing platforms or methods; creation of enhanced formulations; optimisation of process and parameters; and finally exploration of potential applications in various contexts.

Another confectionery product that has been the subject of several recent studies is cookie dough, usually consisting of wheat flour with other common ingredients (e.g. water, butter, sugar) and additives such as ground mealworm powder (Azzollini et al. 2018; Severini, Azzollini, et al. 2018) or probiotics (Zhang, Lou, and Schutyser 2018); the latter is of particular interest as the heat of baking can be detrimental to live micro-organisms. By designing and printing structures with increased surface area to volume ratio, 3DFP can accelerate the baking process and achieve sufficiently high counts of viable probiotics in the baked product to meet the definition of “probiotic foods”, which would conventionally take the form of yogurt or other fermented dairy products that may not be accessible to those with allergies or lactose intolerance (Zhang, Lou, and Schutyser 2018).

In addition, the requirement of post-processing via baking results in the need for a formulation that is both extrudable and resistant to deformation even when baked. This is commonly addressed by optimising the formulation through experimentally varying ingredient ratios (Fan Yang et al. 2018; Zhang, Lou, and Schutyser 2018) or farinograph tests (Severini, Derossi, and Azzollini 2016). Some studies have also introduced additional elements, such as a fast-cooling step between printing and baking (Fan Yang et al. 2019), or texture modifiers such as calcium caseinate (Zhang, Lou, and Schutyser 2018), pea protein (Chuanxing et al. 2018), or xanthan gum as shown in Figure 2 (Kim et al. 2019). Ideally, these modifications should allow the dough to be printed and baked while maintaining a similar taste and texture to conventional cookies, without the large changes in composition as tested in previous papers (see for example (Lipton et al. 2015)).

Dairy

Figure 3. Milk protein pastes with incorporation of whey protein isolate. Reproduced from (Y. Liu et al. 2018).

The printing of dairy products has been less extensively researched, possibly due in part to their wide range from cheese to milk components such as casein and whey protein,

among others. This variety is apparent in the forms of dairy-based inks that have been studied: pastes based on semi-skimmed (SSMP) or skimmed (SMP) milk powder (Lille et al. 2018) and milk protein as shown in Figure 3 (Y. Liu et al. 2018; Y. Liu et al. 2019), as well as both processed (Le Tohic et al. 2018) and semi-hard model cheese (Kern, Weiss, and Hinrichs 2018). Additionally, a range of dairy products or derivatives have been used as ingredients in other food inks, such as milk and butter in dough (Kim et al. 2019), dried non-fat milk in fruit snacks (Derossi et al. 2018), and sodium caseinate in gels (Schutyser et al. 2018).

As with other categories of food inks, preliminary research in dairy-based inks has been chiefly directed at determining the effect of composition on material properties and printability. For instance, the printing performance of milk protein paste was improved by adding whey protein isolate (Y. Liu et al. 2018) and increasing total protein content (Y. Liu et al. 2019). In contrast with the latter observation, Lille et al. found that a SSMP-based paste with 60% solids content yielded excellent printability, while a SMP-based paste was almost unprintable despite having the same solids content and similar protein percentage, presumably due to its significantly lower fat content (Lille et al. 2018).

In the case of cheese, calcium and pH levels were shown to affect both the gel-sol transition temperature and critical shear rate (above which flow instability and melt fracture is observed); this in turn determines the printing temperature and parameter limits of the stable process area for hot-melt extrusion printing (Kern, Weiss, and Hinrichs 2018). Furthermore, the melting and shearing in the printing process has been found to affect the texture of processed cheese, with the printed sample being significantly softer than its original counterpart due to the disruption of fat globules (Le Tohic et al. 2018).

These results highlight the need to better understand the effect of the properties and components of dairy products on the printing process and vice versa. This will enable the utilisation of their structure-forming capabilities in dairy-based food inks or as components in other inks, not only in 3DFP but even outside the food industry, including pharmaceuticals where purified lactose is used as a bulking agent in tablets (Ross, Kelly, and Crowley 2019).

Hydrogels

One of the first printable and edible hydrogels in the literature was demonstrated by Cohen et al., who tested combinations of two hydrocolloids (xanthan gum and gelatin) with flavour concentrates, and found them to simulate a broad variety of textures, from weak to firm and smooth to granular (Cohen et al. 2009). Although they proposed such hydrocolloid combinations as a potential way to contain the material-set bloat by maximising the flexibility of fixed ingredients rather than endlessly creating one-off combinations, in reality the range and combinations of materials used in hydrogels-based 3DFP have only increased. This is unsurprising given the relatively high volume of research in hydrogels over other food inks. Current research in 3DFP of edible hydrogels generally aims to establish one or more of the following: (a) explore new hydrogel inks, (b) determine the effect of types and/or concentrations of different components, (c) optimise printing parameters, (d) develop printing technologies, and (e) establish theoretical models or simulations of the ink or printing process. These are elaborated further below.

Explore new hydrogel inks: With regards to variety in inks, hydrogel research has generally prioritised breadth over depth; specific exceptions aside (see notably (Vancauwenberghe, Katalagarianakis, et al. 2017) and subsequent works), almost all

studies have used different inks rather than continuously developing a particular ink. Furthermore, recent studies have tended towards more complex inks containing multiple components in addition to one or more hydrocolloids, and/or conventional ingredients not previously tested in 3DFP, as shown in Figure 4. Examples include:

- Low methoxylated pectin gel with sugar syrup and bovine serum albumin (Vancauwenberghe, Katalagianakis, et al. 2017), and encapsulation of live plant cells (Vancauwenberghe, Baiye Mfortaw Mbong, et al. 2017);
- Sodium caseinate dispersion with sucrose, pectin, and potato starch (Schutyser et al. 2018);
- Fish surimi gel with sodium chloride (Wang et al. 2018);
- Vitamin D-enriched orange concentrate with wheat starch and *k*-carrageenan gum (Azam et al. 2018);
- Egg white protein mixtures with gelatin, corn starch, and sucrose (L. Liu et al. 2019).

Figure 4. (a) Vitamin-D enriched orange concentrate with wheat starch and *k*-carrageenan; (b) Sodium caseinate with red-coloured olive oil droplets (right half) and without (left half). Reproduced from (Azam et al. 2018) and (Schutyser et al. 2018) respectively.

Determine the effect of types and/or concentration of components: While some studies used a fixed composition throughout, most studies documented its optimisation through experimentation to determine the effect of varying compositions on hydrogel rheology and printability, as well as other factors such as microstructure and gelation temperature. For example, Chen et al. tested suspensions of different starches at varying concentrations to measure the corresponding changes in rheological properties. The inks were then printed in hot extrusion to assess print quality and thus relate the printability to rheological observations, with rice starch suspensions printed at 80°C and concentration 15-25% (w/w) showing good shape stability and smooth surface (Chen et al. 2019). A similar study on rheology and printability was conducted by Liu et al. for a multi-component gel of carrageenan-xanthan gum-potato starch by varying the individual concentrations of each hydrocolloid, with best printing performance observed for inks containing 1%, 0.25-0.5%, and 2% (all w/w) of *k*-carrageenan, xanthan, and starch respectively (Z. Liu et al. 2019).

Optimise printing parameters: Though less ubiquitous than the above, many studies have also included the optimisation of printing parameters, most commonly extrusion rate, nozzle movement speed, and nozzle height and/or diameter (Fanli Yang et al. 2018). Other parameters tested are usually those specific to particular printing platforms or conditions, such as extrusion at temperatures above ambient or with multiple materials. The effect of printing temperature (at or above gelation temperature) on uniformity and dimensional accuracy of printed structures was assessed for a gelatin-carrageenan gel (Warner, Norton, and Mills 2019). In dual-extrusion printing of mashed potato and strawberry juice gel, extruder offset and retraction value were optimised to reduce deviation from designed model and avoid oozing of inks during switching and pausing of individual extruders (Z. Liu, Zhang, and Yang 2018).

Develop printing technologies: While all hydrogel studies were conducted on some type of extrusion-based printers, individual studies have looked into developing or modifying printer hardware to shorten the printing process by inducing gelation during printing rather than as a pre- or post-treatment step. Vancauwenberghe et al. designed co-axial extrusion printhead to deposit a pectin-based ink and CaCl_2 cross-linking solution in the inner and outer flows respectively, thus eliminating the original incubation post-treatment step and allowing more accurate control of object gelation and thus texture (Vancauwenberghe, Verboven, et al. 2018). In comparison, Díaz et al. developed a forced convection system using ambient air to enable printing with *in situ* gelation of *k*-carrageenan inks in several minutes. The resulting rheological properties were comparable to conventionally prepared inks requiring as much as 2 hours for gelation before printing, although shape fidelity was affected as the ink continues to flow on deposition until it is sufficiently cooled to solidify (Díaz et al. 2019). Another hardware modification was tested by Schutyser et al. to modify the texture of sodium caseinate gels by dispersing an olive oil phase through a side inlet in the dispenser nozzle. Though this method presented some difficulty in controlling the pressure for simultaneous deposition, it was able to produce structures with specific spatial distributions of the oil phase (see Figure 4), as opposed to pre-mixing the oil into the gel before printing (Schutyser et al. 2018).

Models or simulations: Recent papers have also shown a movement towards predictive models rather than solely experimental work, whether through theoretical models which are then correlated to actual results, or vice versa by developing models from experimental data. Some models take the form of equations to calculate variables like printing speed given a required gel strength, through a time and temperature optimisation function based on response surface modelling from experimental data

(Díaz et al. 2019). Other models are numerical, such as the simulation of printing parameter effects on velocity, shear rate, and pressure fields in the flow channel from barrel to nozzle (Fanli Yang et al. 2019). Vancauwenberghe et al. used analytical and finite element models to predict the relative Young's moduli of honeycomb structures printed in pectin-based ink with increasing cell size (Vancauwenberghe, Delele, et al. 2018). Though some deviation was observed from experimental measurements in both cases, further refinement of such models could provide useful reference data for design of both 3DFP processes and structures, reducing the need for experimental trial-and-error.

Among the five aims highlighted above, the last two may be of most interest for further study. While the exploration of new hydrogels may be useful as a basis for developing more conventional food types or even intrinsically as a novel food simulant, improvements in printing technology and models could be generalised to benefit other areas in 3DFP, with hydrogels used as a platform for testing or standardisation. An example is the classification system for printability of food inks under extrusion as developed by Kim et al. based on hydrogels at varying concentrations. Specifically, methylcellulose at concentrations of up to 15% (w/w) was proposed as the reference material to assign two letter grades for each food ink based on their handling properties during printing and dimensional stability after gelation, measured by their extrusion force and shear modulus respectively (Kim, Bae, and Park 2017).

Non-natively extrudable

Inks in this category cannot be extruded through a nozzle in their raw form, so more steps are required to produce a printable food ink.

Figure 5. (a) Freeze-dried broccoli powder with xanthan gum; (b) Dual extrusion of mashed potato and strawberry juice gel as alternating layers, internal/external shape, and infill/perimeter layers. Reproduced from (Kim et al. 2018) and (Z. Liu, Zhang, and Yang 2018) respectively.

While plant derivatives such as fruit juices or concentrates have been used in the creation of hydrogels, this section discusses inks that are derived from solid plant materials and therefore tend to take the form of slurries or pastes. Five steps have been identified in 3DFP of plant inks: material selection, formulation of ingredients, preparation of food ink, choice of printing conditions, and finally methods to extend shelf life of the final structures (Ricci, Derossi, and Severini 2019). Given the high water and fibre content of plants, some differences in food ink preparation can be identified, especially if fresh produce is used directly (as opposed to freeze-dried powder or other processed material). Firstly, variations in properties between different batches should be minimised or otherwise accounted for, as such variance can cause considerable deviation in final printing results (An et al. 2018). Secondly, viscosity should be adjusted by adding thickeners and/or dehydrating the ink by separating the solid and liquid phases in a juicer (Severini, Derossi, et al. 2018). Finally, the post-printing storage and stability should be considered given the highly perishable nature of most fruits and vegetables, unless the printed structure is expected to be consumed promptly upon printing. For inks where post-processing by drying is needed, a higher

dry matter content may be favourable to reduce water loss and thus minimise shape deformation (Lille et al. 2018).

One plant-based ink that has been quite extensively studied is mashed potato, which may be prepared directly from raw potatoes (Z. Liu, Zhang, et al. 2018) or from gelatinised potato flakes or powder (Z. Liu, Bhandari, et al. 2018; Dankar, Pujolà, et al. 2018). Although mashed potato is itself extrudable, the use of additives (e.g. alginate, agar (Dankar, Pujolà, et al. 2018), or gums such as xanthan and *k*-carrageenan (Z. Liu, Zhang, and Bhandari 2018)) in concentrations as low as 1% has been found to improve rheological properties and print outcomes such as texture, surface quality, and shape stability. Other studies have evaluated the differences in texture and microstructure of printed and cast samples (Z. Liu, Bhandari, et al. 2018), or performance in dual extrusion with strawberry juice gel of lower mechanical strength as shown in Figure 5 (Z. Liu, Zhang, and Yang 2018). As mashed potato ink are easier to prepare and print than other plant-based inks, it is useful as a platform to improve our understanding of how to modify such inks for printing, especially the effects of additives on vegetable substances that contain multiple components, as opposed to hydrogels which generally have fewer and more well-defined ingredients.

The effect of different preparation methods on food inks was demonstrated by An et al., who studied algae-based inks produced from *Nostoc sphaeroides* biomass in both fresh and rehydrated forms, and found visible disparity in print quality for fresh inks from batches obtained on different dates, even though the water content of the two batches were not significantly different. Thus, drying and rehydrating plant material can minimise non-uniformity in the raw material while enabling better storage (An et al. 2018). However, even when dehydrated materials are used, the rheological differences between them has to be minimised through careful material selection. This was shown

by Kim et al., who tested inks made from a freeze-dried vegetable powder and a hydrocolloid in different combinations. Of the four hydrocolloids tested, only xanthan gum produced uniformly good results with all three vegetable powders even at high concentration (see Figure 5), as its high water hydration capacity enabled it to inhibit variations caused by powder swelling, producing inks with good extrudability and shape retention (Kim et al. 2018).

Finally, while all the studies above have used fresh or processed plant ingredients as the main component of the ink, other methods such as encapsulating live plant cells in edible bio-inks can also be used to create artificial plant tissue with good cell viability in the printed structure (Vancauwenberghe, Baiye Mfortaw Mbong, et al. 2017).

Meat

Perhaps surprisingly, meat in its various forms is by far the least well-researched among all the categories of food inks identified here, with only one study having extensively focused on it through the development of a printer with a peristaltic pump to extrude slurries of fibrous meat and gelatin (C. Liu, Ho, and Wang 2017). Other reports on the matter were brief, typically as one part of a study in printing of several materials; these included turkey and scallop purees with transglutaminase (Lipton et al. 2010), blended canned tuna (Kouzani et al. 2017), as well as pastes of shrimp or ground chicken with other ingredients such as egg white and cooking wine (Hertafeld et al. 2019). Given the proteins and other nutrients present in meat, it is imperative that further development be carried out in this area, especially if 3DFP is to supply complete and balanced meals rather than individual food items, as in the case of production of texturally-modified foods for dysphagia patients. Alternatively, 3DFP may even be used to produce meat-like products from other components, similar to how animal-free leather has been

created by biofabrication of collagen proteins (Modern Meadow 2019).

Summary of food inks for extrusion printing

An overview of the food inks for extrusion printing outlined above is shown in Table 1. For each category of food inks, general ingredient formulations have been identified with specific examples from the literature, followed by the main steps used from preparing to printing to post-processing (besides refrigeration for storage). Finally, sample values of rheological properties such as apparent viscosity, storage and loss moduli, and yield stress are presented for printable formulations where available. It should be noted that the table includes only data from the original studies that were given in numerical form; however, as most rheological data were presented in graphical forms and thus omitted here, the reader is encouraged to check the respective original publications for additional data.

Food inks in non-extrusion printing

Besides material extrusion, three other printing technologies have been used in 3DFP, namely inkjet printing, selective laser sintering, and binder jetting (Z. Liu and Zhang 2019). However, the usable food inks for the latter two technologies are often limited to powdered materials such as sugar for sintering and binder jetting (as well as a liquid binder in the latter case), or low-viscosity materials such as sauces for inkjet printing. Furthermore, these printing technologies have been used almost solely in commercial printers, such as by the CandyFab printer which uses a beam of hot air to sinter sugar powder (CandyFab 2019), the ChefJet Pro which prints sugar with water as a binder alongside an inkjet head to add food colouring (Gold 2014), or the FoodJet which prints designs onto food products using food inks with a solidifying agent to encourage layer merging and prevent structure collapse (Alec 2016).

Outside of commercial use, informal trials of different food inks have also been done by hobbyists and reported on online platforms, such as binder jetting with sake (Japanese rice wine) or other alcohol-based formulations as binders, though powder formulations were still limited to sugar or sugar mixtures (Holland, Foster, and Tuck 2019). Even in academic fields, little extensive research beyond preliminary testing has been reported on these printing platforms. For instance, researchers from TNO demonstrated laser sintering of the drinking chocolate powder Nesquik, which was able to fabricate chain links and company logos due to its mixture of sugar and fat content (Gray 2010), however no published reports on further exploration or characterisation of such material were found. Exceptions to this include the creation of an electrostatic inkjet printer for chocolate (as mentioned previously), and the development of a food-grade cellulose composite powder for binder jetting. Ball-milled amorphous cellulose was selectively recrystallised using a xanthan gum-based food ink, which was deposited through an inkjet printer onto a recessed plate holding the powder to create cohesive 2D structures (Holland et al. 2018). This was then extended to 3D structures, with more complex structures being enabled by the addition of konjac glucomannan to the powder (Holland, Tuck, and Foster 2018). As these ingredients are classified as dietary fibre, such inks could find applications in creating edible 3D designs with much lower caloric content than sugar or chocolate. However, more research is needed to better understand the relationship between material properties and process parameters to printing outcomes, and to potentially extend the range of food inks to more nutritious materials rather than just decorative substances.

Additives and their effect

Various additives have been mentioned in the previous sections, from common hydrocolloids like gums or starches, to specific ones such as sodium chloride in surimi

gel (Wang et al. 2018). Their main roles in food inks can be divided into two types: those that confer better printing outcomes, versus those that improve performance in other aspects. The former chiefly concerns texture modification to enhance printability and quality (e.g. precision, surface smoothness), as well as stability to withstand post-processing when required. The latter type includes benefits in a wider range of areas, such as health or nutrition, sustainability through use of alternative food sources, and improved flavouring of food inks. For instance, compared to traditional sources like meat or fish, insects not only provide similar high-quality protein and nutrients, but also sustainability improvements (e.g. more efficient feed-to-meat conversion rates, smaller land requirements, and reduced greenhouse gas production), although consumer acceptance still remains a major obstacle to its wider use. (Food and Agriculture Organization of the United Nations 2019). By incorporating insect protein into food inks, 3DFP could be used to create structures that look and taste appealing, potentially encouraging otherwise-resistant consumers to give these foods a try.

However, additives for health or flavour may also affect rheological or other properties of the ink, especially in interaction with other components or additives. This was shown in a study by Kim et al., who tested gels made using one of four hydrocolloids in combination with a food ingredient (salt, sucrose, or acetic acid) to study the effect of ions, sugars, or pH changes on ink viscosity, as these may be naturally present or added when seasoning food inks. Minimal change was observed for xanthan and guar gum, indicating their stability to such additions, but decreased viscosity was generally observed for locust bean gum and hydroxypropyl methylcellulose (Kim et al. 2018). Also, as in the case of chocolate, there may be maximum additive concentrations beyond which the ink is no longer printable (Liang Hao et al. 2019).

In addition, certain process modifications (i.e. pre/post-processing steps or printing parameter adjustment) may be usable with additives or in replacement of them to improve print quality, while minimising any cost or flavour changes in the printed products. These strategies include viscosity modification by dehydration of inks with high water content, adjustment of printing temperature, to fast-cooling after printing to increase geometric stability in post-processing (Fan Yang et al. 2019).

Finally, any simulations or models developed to predict the behaviour of food inks should ideally take into account any relevant effects of the additives used. An example of this is the regression model developed by Vancauwenberghe et al. for their pectin-based food ink, which determines the concentrations of each ingredient in order to yield a food ink with the desired flowability and mechanical/structural properties (Vancauwenberghe, Katalagarianakis, et al. 2017).

Challenges and opportunities

Much remains to be explored in the field of 3DFP, but considerable challenges still need to be overcome in order to facilitate future developments, especially in terms of materials and processes. Several of these aspects are highlighted below, including considerations in the choice of food inks and additives, disparity between current 3D printing technology and food applications, as well as preliminary considerations necessary for consumer use of 3DFP.

Development across the different categories of food inks has been greatly uneven, with chocolate and hydrogels having been studied far more extensively than others like dairy or meat. While this is unsurprising given the more easily printable nature of some foods over others, the question of general acceptability must also be considered when developing new food inks in addition to their rheological properties and printability, if 3DFP is to produce food that is not only printable but also desirable

for consumption. This concerns not only the question of which categories of food inks should be further developed, but also the selection of ingredients and processing methods to produce a food ink with appealing flavour and texture. The effect of the printing process on the texture or mouthfeel of the final product also needs to be considered, such as any microstructural changes in the final product compared to the unprinted ink due to the high shear and potential temperature changes in printing.

Priority should be given to food inks that confer additional benefits in areas like nutrition, whether by making inks directly from healthier food sources such as vegetables, or by enhancing other inks with healthy additives. In particular, 3DFP may be uniquely placed to produce enriched food items which would be difficult or impossible through conventional methods, such as ensuring the viability of probiotic baked foods even after post-processing (Zhang, Lou, and Schutyser 2018). Besides their nutritional content, other potential benefits include sustainability through utilising alternative nutrient sources, or texture customisation within a structure such as by dual extrusion of contrasting materials (Z. Liu, Zhang, and Yang 2018) or spatially-distributed dispersal of a second phase (Schutyser et al. 2018). However, in order to maximise the customisation with such additives, a better understanding of their effect on rheology and printability is needed. Furthermore, to minimise costs and textural or flavour changes associated with these additives, alternative ways to achieve good performance and printability should also be identified, such as dehydration or temperature control in pre-processing and printing.

In terms of processes, there is a clear mismatch between current printing technology with 3DFP especially in the software aspect, and there exists considerable opportunity to further improve current tools and adapt them for food inks. Multiple studies (see for instance (Severini, Azzollini, et al. 2018; Z. Liu, Bhandari, et al. 2018;

Severini, Derossi, et al. 2018) among others) have noted that printing parameters and path planning on existing slicing software such as Cura or Slic3r are optimised for thermoplastic materials like PLA or ABS which behave very differently from food inks, resulting in poor print quality if this difference is not sufficiently compensated for by optimising printing parameters such as extrusion rate and print speed. Together with other variables that may be fixed in the printer firmware, this causes discrepancies between the designed models and printed objects, as well as between estimated and actual printing times. As a result, considerable trial-and-error in adjusting the printing process is currently needed to achieve satisfactory outcomes, which needs to be minimised to shorten the process of testing and later printing food inks for consumption.

A similar though less severe lack of compatible technology can also be observed in the hardware aspect through the printer types used in 3DFP testing. While some studies used commercial food printers such as the FoodBot-S2 from Shiyin Tech or the FSE 2 from PORIMY, many others used open-source platforms such as the RepRap, or even custom printers built or modified from non-printers (usually CNC machines) equipped with syringe extruders. However, in most cases it is unclear whether the latter two types of printers fulfil requirements for food safety, which would be necessary if 3DFP is to be developed for actual consumption. One possible solution to these issues is to develop both printers and corresponding software designed specifically for extrusion printing of food inks; however, this is made difficult by the wide range in material properties across the range of food inks, and moreover would also reduce compatibility with the existing 3D printing ecosystem, presenting a barrier to entry into the field of 3DFP.

Finally, in order to develop 3DFP for practical consumption especially at the commercial level, improvements have to be made in at least three areas: printing time,

food safety, and repeatability between batches. Speed improvements can be achieved by adaptive algorithms that adjust printing parameters to balance print quality with time taken, especially since a high resolution may not be of utmost priority in most food applications. For inks where a gelation step is required, pre- or post-processing time can be reduced considerably through the use of in-situ gelation. In addition to the possible issues with non-food printers as highlighted above, the safety of food inks also needs to be quantified through measures such as monitoring the microbial content in the food during preparation and printing, and any changes in the printed food over time through post-processing and storage if it is not intended to be consumed immediately. Last but not least, variations between batches of raw material have already been shown to significantly affect printing outcomes; the extent of such effects and possible methods of mitigation will have to be considered in order to ensure consistent quality even outside lab conditions. Developing standards and quality control procedures should help to increase adoption of 3DFP especially in commercial applications (Tran 2017).

Conclusion

In this paper, the current status of food ink development for 3D printing was reviewed with respect to five categories of materials. These categories were classified according to the type of food represented by the ink and further grouped into two larger categories: natively extrudable (i.e. confectionery, dairy, hydrogels) versus non-natively extrudable materials (i.e. plants, meat). Focus was placed on inks for extrusion-based printing given its wider usage in research, as compared to other methods such as binder jetting or inkjet printing which have been used more in commercial or hobbyist fields and thus are less well-documented in comparison.

The extent of development was found to be uneven across the different ink categories, with natively extrudable inks generally being more well-researched, whereas

development of non-natively extrudable inks was more recent and still at a preliminary stage. Despite this disparity, several common trends in research could be identified: expansion in food inks beyond conventional materials of chocolate or gels (especially towards more nutritious inks that are made from or enriched with healthy ingredients), diversification of printing processes besides single material extrusion at ambient temperature, and finally development of quantitative or numerical models that are able to predict specific parameters for printing. Additionally, hydrocolloids and other additives were found to play important roles in the creation of printable gels, though these functions could in some cases be supplemented or replaced by modifications to the preparation or printing process. Finally, three key trends were identified in the development of food inks for 3DFP, namely:

- Diversification into non-natively extrudable materials;
- Nutritional improvements of printed food;
- Novel formulations tailored specifically for 3DFP.

Having emerged only within the past decade, 3D food printing is still a nascent field even compared to other applications such as bioprinting, and it is difficult to say if the directions of research outlined above will continue or further diverge as the field develops, depending on the intended application or target audience of a given ink. What is certain is that 3D food printing will not simply remain a novelty to construct edible structures, but instead move into producing practical food items that people may consume in daily life. The main challenge will be to achieve a better understanding of food inks in terms of how their components interact with each other and affect the printing process; this will ideally enable the minimisation of trial-and-error in optimising their formulation and printing conditions, and thus shorten development time. Moreover, the practical aspects of food safety, palatability, and public acceptance

among others will have to be thoroughly considered if 3D food printing is to reach consumers, whether in the commercial or personal setting.

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Table 1. Summary of the categories of food inks in extrusion-based printing. (η : viscosity, G^* : complex modulus, G' : storage modulus, G'' : loss modulus, $\tan\delta$: loss tangent, YS: yield stress, ω : angular frequency, LVR: linear viscoelastic range.)

Ink category	General formulations and examples	Main steps in pre-processing, printing, and post-treatment* (*besides refrigeration for storage)	Sample rheological properties
Confectionery	Dark or milk chocolate with cranberry powder and methylcellulose (Liang Hao et al. 2019), magnesium stearate or plant sterol powder (Mantihal et al. 2019)	<i>Pre</i> : Mix chocolate and additives by melting or grinding (if loaded as solid into hopper) <i>Print</i> : Syringe or screw extrusion above ambient temperature (approx. 30–40°C) <i>Post</i> : None	<i>Milk chocolate</i> : $\eta=16\text{Pa}\cdot\text{s}$ (at almost rest) and $2.35\text{Pa}\cdot\text{s}$ (at 50s^{-1}) (Lanaro et al. 2017) <i>Dark chocolate with plant sterol powder</i> (from hopper before printing): $\eta=4.58\pm 0.86\text{Pa}\cdot\text{s}$, YS= 54.25 ± 4.74 (Mantihal et al. 2019)
	Dough of flour and water/milk with other ingredients (sugar, butter, egg) and additives, e.g. probiotics (Zhang, Lou, and Schutyser 2018), xanthan gum (Kim et al. 2019), or mealworm powder (Azzollini et al. 2018)	<i>Pre</i> : Mix to form dough, rest <i>Print</i> : Syringe extrusion at ambient temperature, or screw extrusion above ambient ($>100^\circ\text{C}$) <i>Post</i> : Baking in oven (approx. 150–200°C for ≤ 16 minutes) or dried (60°C for 6 hr)	<i>Dough</i> (icing sugar, butter, low-gluten flour, egg, water): $\eta=1.602\text{Pa}\cdot\text{s}$ (at 1s^{-1}), $G'=33.201\text{Pa}$, $G''=11.704\text{Pa}$ (both at 10s^{-1}) (Fan Yang et al. 2018)
Dairy	Pastes/gels of milk products, e.g. semi-skimmed milk powder in water (Lille et al. 2018), milk protein concentrate with whey protein isolate (Y. Liu et al. 2018) or in sodium caseinate solution (Y. Liu et al. 2019)	<i>Pre</i> : Mix ingredients, homogenise (by extrusion through syringe), rest <i>Print</i> : Syringe or air pressure-driven extrusion at ambient temperature <i>Post</i> : None or drying (by oven or freeze-drying)	<i>60% semi-skimmed milk powder in water</i> : $G'=1550\pm 230\text{Pa}$, $G''=290\pm 41\text{Pa}$ (average in LVR), YS= $61.1\pm 0.3\text{Pa}$ (Lille et al. 2018) <i>Milk protein in sodium caseinate</i> (400g/L protein content): $G'=3936\omega^{0.3585}$, YS=500Pa (Y. Liu et al. 2019)
	Cheese (Kern, Weiss, and	<i>Pre</i> : Melt in syringe (in water bath)	<i>Processed cheese</i> (unaltered, before melting):

Ink category	General formulations and examples	Main steps in pre-processing, printing, and post-treatment* (*besides refrigeration for storage)	Sample rheological properties
	Hinrichs 2018; Le Tohic et al. 2018)	<i>Print:</i> Syringe extrusion above ambient temperature (approx. 75°C) <i>Post:</i> None	$G^*=66188\pm28229\text{Pa}$ (at 30°C), maximum $\tan\delta=0.29\pm0.03$ (at 64.2±4.14°C) <i>Processed cheese (after low-speed printing):</i> $G^*=32242\pm4301\text{Pa}$ (at 30°C), maximum $\tan\delta=0.35\pm0.02$ (at 62.9±1.17°C) (Le Tohic et al. 2018)
Hydrogels	Gels of one or more hydrocolloids and/or other ingredients in solution, e.g. pectin gel with sugar syrup and bovine serum albumin (Vancauwenberghe, Katalagarianakis, et al. 2017), sodium caseinate dispersion with sucrose, pectin, and starch (Schutyser et al. 2018), fish surimi gel with NaCl (Wang et al. 2018), egg white protein mixture with gelatin, starch, and sucrose (L. Liu et al. 2019)	<i>Pre:</i> Mix ingredients, homogenise, gelation by crosslinking or heating in water bath (if not done in situ or post-printing) <i>Print:</i> Syringe, screw, or air pressure-driven extrusion at ambient or elevated temperature (usually at or above gelation temperature), gelation (if done in situ) <i>Post:</i> None or gelation by incubating in crosslinker solution	<i>Lemon juice gel with potato starch:</i> $\eta=8079.3\text{Pa}\cdot\text{s}$ (at 0.1s ⁻¹), $G'=4924.2\text{Pa}$, $G''=760.8\text{Pa}$, $\tan\delta=0.155$ (at 0.63rad/s) (Fanli Yang et al. 2018) <i>Multi-component gel of k-carrageenan, xanthan, and starch:</i> $G'=814.8\pm34.6\text{Pa}$, $G''=189.8\pm16.3\text{Pa}$ (both at 10s ⁻¹), $\tan\delta=0.23$, $YS=60.5\pm3.1\text{Pa}$ (Z. Liu et al. 2019)
Plants	Mashed potato/MP (from raw potatoes or potato flakes in water) with additives, e.g. trehalose and potato starch (Z. Liu, Zhang, et al. 2018), alginate or agar (Dankar, Pujolà, et al. 2018), <i>k</i> -carrageenan and xanthan gums (Z. Liu, Bhandari, et al. 2018)	<i>Pre:</i> Prepare MP (cook and mash from raw, or mix potato flakes with hot water), homogenise, wrap and cook in water bath, rest <i>Print:</i> Syringe or screw extrusion at ambient temperature <i>Post:</i> None	<i>MP with 2% potato starch (from raw potatoes):</i> $\eta=36.36\text{Pa}\cdot\text{s}$ (at 0.01s ⁻¹), $\tan\delta=0.26$ (at 1Hz), $YS=312.16\text{Pa}$ (Z. Liu, Zhang, et al. 2018) <i>MP with carrageenan and xanthan (from flakes):</i> $\eta=42608\text{Pa}\cdot\text{s}$ (at 0.01s ⁻¹) and 1018Pa·s (at 1s ⁻¹), $\tan\delta=0.15\text{--}0.18$ (between 1–100rad/s) (Z. Liu, Bhandari, et al. 2018) $\tan\delta=0.177$ (average over 1–6.28rad/s) (Z. Liu,

Ink category	General formulations and examples	Main steps in pre-processing, printing, and post-treatment* (*besides refrigeration for storage)	Sample rheological properties
	Other plant-based inks from fresh produce or dehydrated powders with hydrocolloids, e.g. fruit-based “smoothies” with pectin (Derossi et al. 2018) or collagen (Severini, Derossi, et al. 2018), <i>Nostoc spharoides</i> algae with starch (An et al. 2018), plant protein concentrates or rye bran in water (Lille et al. 2018), freeze-dried vegetable powders with xanthan gum (Kim et al. 2018)	<i>Pre: (if fresh produce)</i> Blend, dehydrate (in juicer or oven), homogenise, mash/sieve, add hydrocolloid; <i>(if dehydrated)</i> Mix ingredients, homogenise, rest <i>Print:</i> Syringe, screw, or air pressure-driven extrusion at ambient temperature <i>Post:</i> None or drying (by oven or freeze-drying)	Bhandari, et al. 2018) <i>Fruit-based smoothie (from fresh produce):</i> $\eta=2560\text{Pa}\cdot\text{s}$ (at 0.14s^{-1}) and $35.04\text{Pa}\cdot\text{s}$ (at 28.34s^{-1}) (Derossi et al. 2018) <i>Vegetable inks (with 10% freeze-dried powder):</i> $G'=1640\text{--}30660\text{Pa}$, $G''=250\text{--}7880\text{Pa}$ (between 1–100rad/s) (Kim et al. 2018)
Meat	Pureed meat with texture-modifying or other additives, e.g. scallop or turkey with transglutaminase (Lipton et al. 2010), fibrous meat such as chicken with gelatin (C. Liu, Ho, and Wang 2017), ground chicken or shrimp with egg white (Hertafeld et al. 2019)	<i>Pre:</i> Puree raw meat, mix with additives <i>Print:</i> Syringe extrusion at ambient, possible in situ cooking with infrared lamp <i>Post:</i> Cooking (deep-frying, sous-vide in controlled-vapour oven) or none (if already cooked in situ)	N/A (no rheological data reported)

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