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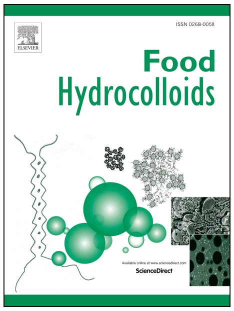

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Jetting-based 3D Printing of Edible Materials

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Abstract

3D printing or additive manufacturing is a fabrication technique gaining considerable interest across many disciplines owing to its dimensional precision and ability to produce novel geometrical shapes. Jetting-based 3D deposition is an important subset of 3D printing as it allows rather small units of deposition (i.e. droplets). Use of this technique for edible materials is relatively limited due to inability of piezoelectric inkjet printing to print inks with viscosity > 0.03 Pa.s. As a result, the technique is sometimes referred to as 2D food printing.

The present review summarises reported studies on jetting-based printing of edible formulations. It also discusses various approaches which could result in further progress of this field of study. They include: (i) advancements in printing techniques such as thermally, pneumatically and electrostatically aided deposition and (ii) innovative ink formulations in which supramolecular interactions, e.g. hydrophobic and electrostatic associations dominate the microstructure of the printed object. With an optimal combination of these two, novel microstructures can be produced which may find their applications beyond food, into pharmaceuticals/nutraceuticals. Where relevant, non-edible formulations have been discussed which have the underlying microstructural principles that can be translated to edible formulations.

Keywords

3D Printing; Additive Manufacturing; Jetting; Inkjet; Food; Edible.

1.0 Jetting-based Printing Techniques

The jetting-based 3D printing is a method for depositing liquid materials in a layer-by-layer fashion with each layer composed of distinct droplets dispensed on either a substrate, or a previous layer of the same material, or a powder layer. Several of such layers produce a 3D object when fused together. Such dispensing methods are typically characterised by relatively smaller units of deposition (drops as opposed to lines in extrusion), higher resolution and greater control over the amount being dispensed. These characteristics are shared by a diverse range of techniques such as piezoelectric inkjet printing, binder jetting, electrostatic jetting, hot-melt jetting as well as pneumatic jetting.

3D printing of food is a diverse field with many different techniques as shown in Figure 1. While many reviews have described recent advance in extrusion-based 3D printing (Godoi, Prakash, & Bhandari, 2016; Liu & Zhang, 2019; Liu, Zhang, Bhandari, & Wang, 2017; Pitayachaval, Sanklong, & Thongrak, 2018; Voon, An, Wong, Zhang, & Chua, 2019), a detailed literature review of jetting-based 3D printing of edible materials has not been reported to the best of our knowledge.

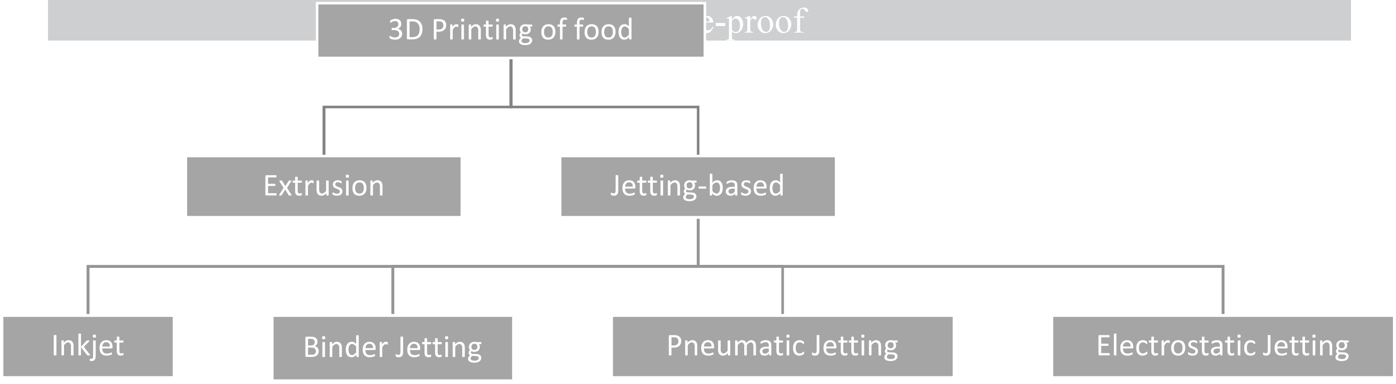

Figure 1. Classification of jetting-based techniques within 3D printing.

1.1 Piezoelectric Inkjet Printing

Piezoelectric inkjet printing refers to the low-viscosity (< 0.03 Pa.s) inks being dispensed using pressure generated by change in shape of a piezoelectric actuator in response to electric voltage. Vast majority of edible formulations which are able to support their shape against gravity have viscosity higher than 0.03 Pa.s at an ambient temperature. Moreover, many cross-linking agents frequently used in piezoelectric inkjet formulations which are capable to solidify a low-viscosity fluid (by means of covalent bond formation) are not edible.

As a result, this printing technique has been used mostly for printing on food substrates, i.e. filling surface cavities or creating images which usually involve low-viscosity inks. This limitation of conventional inkjet printing has caused it to be frequently regarded as a ‘2D printing’ technique as opposed to a ‘3D printing’ technique. Such uses of inkjet printing in food have been discussed in previously published reviews on the topic (Godoi, Prakash, & Bhandari, 2016; Liu & Zhang, 2019; Liu, Zhang, Bhandari, & Wang, 2017; Pitayachaval, Sanklong, & Thongrak, 2018; Voon, An, Wong, Zhang, & Chua, 2019). There have also been reports of printing drug solutions on edible substrates (Iftimi, Edinger, Bar-Shalom, Rantanen, & Genina, 2019) as well as making small cell-containing hydrogel structures using alginate (Negro, Cherbuin, & Lutolf, 2018).

The viscosity limitation of piezoelectric inkjet has been somewhat mitigated by printing at elevated temperatures. Beeswax is an FDA-approved food additive which has the potential to be used for delivering flavours and nutrients. Hot-melt inkjet printing has been used to produce a cylindrical geometry from a mixture of edible beeswax and a drug fenofibrate at 90°C. High precision of the technique allowed the inner bulk of cylinder to be patterned in shape of hollow honeycomb-like structures. Specimen with different surface area to volume ratio were printed, and provided a means to control in-vitro drug release profiles (Kyobula et al., 2017). The hot-melt approach has also been used for another drop-on-demand technique analogous to inkjet in which a precision positive displacement pump actuates droplets formation through a nozzle (Içten, Giridhar, Taylor, Nagy, & Reklaitis, 2015).

Other approaches for printing higher viscosity liquids include aiding the droplet ejection by means of air pressure (pneumatic jetting) (Gao, He, Fu, Qiu, & Jin, 2016; YanPu, 2016) or by introducing electrostatic charge difference between the printing material and the substrate (electrostatic jetting) (Suzuki, Takagishi, & Umezu, 2019; Takagishi, Suzuki, & Umezu, 2018).

The term ‘inkjet printing’ in food printing literature has often been used to describe binder jetting (Nachal, Moses, Karthik, & Anandharamakrishnan, 2019) and pneumatic jetting (Godoi et al., 2016; Gong et al., 2014; Jiang et al., 2019; Sun et al., 2015), thereby introducing occasional ambiguity. In this article, jetting techniques will be referred to as specifically as possible in order to avoid confusion.

1.2 Binder Jetting

In binder jetting, fabrication of each printed layer involves spreading of a thin layer of powder followed by jetting of a low-viscosity binder liquid in the shape of the respective cross-section of the object being printed. Such a liquid is capable of fusing together the powder particles. The post-processing steps comprise of removal of binder by heating as well as of unbound powder. Sugar (Southerland, Walters, & Huson, 2011) and many polymeric materials such as cellulose and xanthan gum (Holland, Foster, MacNaughtan, & Tuck, 2018), hydroxypropyl methylcellulose and ethylcellulose (Yu et al., 2009) as well as corn starch, dextran and gelatine (Lam, Mo, Teoh, & Hutmacher, 2002) have been reported to be 3D printed using binder jetting. The binders employed in such studies were either aqueous or alcohol based or both, sometimes with additives. Conventional piezoelectric inkjet printing is used for dispensing binder droplets.

There is significant overlap between the requirements for a material to be edible and for it to be used in pharmaceutical, nutraceutical and medical applications. As a result, binder jetting on edible powders has evolved further to find applications beyond food, into fabricating scaffolds for tissue engineering and controlled delivery of active compounds. It has been reported that within a doughnut-shaped pill containing paracetamol prepared by binder jetting, the in-vitro release times of paracetamol (Yu et al., 2009) can be controlled by changing the annular thickness as well as the amount of binder comprising a release-retardant substance (ethyl cellulose). The paracetamol dosage can be controlled by adjusting the pill height. These pills were found to have a near linear in-vitro release profile at many different inner/outer diameters, heights and concentrations of ethyl cellulose within the binder. Spritam©, a new pharmaceutical product which was approved in 2015 by the US Food and Drug Administration (FDA) has been the first product manufactured using binder jetting (and 3D printing in general) to obtain such approval (Maniruzzaman, 2019). The main advantage offered by binder jetting in this case was the high porosity of the printed pill, which allowed much faster oral disintegration.

Tissue engineering scaffolds containing corn starch, dextran and gelatin have been printed by (Lam et al., 2002) using binder jetting and were heated at 100°C for 1 hour after printing to remove the binder solvent. While the scaffolds already had sufficient mechanical strength to be handled manually, they were infiltrated with the co-polymer of polylactide and polycaprolactone in dichloromethane in order to improve stiffness and water resistance and to minimise porosity.

The use of inkjet printing to merely deposit binders as opposed to depositing the rest of the material is what makes binder jetting distinct from piezoelectric inkjet printing. The former has been used in literature much more frequently with 3D printing of edible materials than the latter.

1.3 Pneumatic Jetting

Pneumatic jetting (also known as valvejet) is a high-viscosity analogue of inkjet printing which is increasingly being used in both academic and commercial settings. Controllable and continuous air pressure is applied to the ink which is pushed through a dispensing valve opening and closing according to piezoelectric movement prompted by electric signals. The pneumatic pressure enables the use of inks with much higher viscosity compared to conventional inkjet printing, often hundreds of Pa.s depending on the nozzle diameter.

Fabrication of a colour candy has been reported using four multiple inks through pneumatic jet (YanPu, 2016). These inks were cream, honey, fruit gel and starch. Although viscosity values of these inks were not reported, the fact that the candy was able to retain its shape suggests that the viscosities of at least some of these inks were higher than 0.03 Pa.s.

Microparticles of calcium alginate were prepared by pneumatic jetting of sodium alginate solution in $CaCl_2$ solutions (Gao et al., 2016). The concentration of sodium alginate used was as high as 1.2% (w/v), likely to have viscosity substantially greater than 0.03 Pa.s. It was found that the parameters such as droplet velocity and $CaCl_2$ concentration have a great influence over the shape of the microparticles. Alginate gels are widely used in food, tissue engineering and delivery of active compounds.

1.4 Electrostatic Jetting

Besides pneumatic stress, electrostatic attraction between the ink and the substrate aids jetting allowing higher viscosity inks to be printed. Using this method, chocolate-based ink with ~0.6 Pa.s viscosity was printed. It was shown that the diameter of the printed droplet (and by extension, width of the printed line) was a function of applied voltage as well as the gap between the nozzle and the substrate. This was attributed to the difference in the so-called 'discharge state', i.e. the droplet formation at the tip of the nozzle under different conditions as observed using a high-speed camera (Takagishi et al., 2018). A follow-up study (Suzuki et al., 2019) to this showed that introducing an acrylonitrile butadiene styrene (ABS) fibre at the tip of the nozzle guided the droplet formation and made the printed lines more continuous and precise. The effect was found to be more profound at lower fibre diameters, or when the fibre tip was sharpened. However, only 4.25 µm of line height was achieved after printing 10 layers.

2.0 Hydrocolloids in Jetting-based Food Printing

Natural hydrocolloids including polysaccharides are an integral part of food formulations. Their properties are very diverse in terms of their viscosity, texture (liquid or gel), charge (charged or neutral) as well as surface activity. The use of hydrocolloids in piezoelectric inkjet printing is limited owing to the viscosity requirement. However, hydrocolloid powders such as dextran, starch, and cellulosic polymers are used heavily in powder binding owing to their high water-retention capability.

Since non-dilute solutions of vast majority of polysaccharides have viscosity not suitable for inkjet, they can be used in either pneumatic jetting, electrostatic jetting or jetting aided by positive displacement pump.

3.0 Developing New Formulations

Based on the above discussion of various jetting techniques, it emerges that the requirement for viscosity for piezoelectric inkjet printing severely restricts the range of formulations which can be adapted for inkjet-based 3D printing, particularly edible formulations. Low-viscosity inks are unlikely to form a self-supporting structure unless they contain some cross-linking agent. Many chemical cross-linking agents are not considered edible. Advancements such as hot-melt, pneumatic and electrostatic jetting allow the ink viscosity to be higher by several orders of magnitude. However, the edible printing formulations have not quite evolved at the same pace in such a way that full utilisation of such advancements in printing techniques is achieved.

The reason for this is that many of the above examples of printing edible materials simply involve precise spatial deposition of material. The microstructure of the materials which affects the sensory perceptions such as texture and taste remains largely the same before and after printing. While novel and more complex geometrical shapes are being achieved through 3D printing, novel microstructures are necessarily not. While exploiting the precise deposition of jetting, it is often ignored that the same small unit of deposition (i.e. a droplet) which can achieve a precise geometry, can also lead to a much more efficient and rapid mixing of two or more components owing to their larger surface area compared to volume. Such an efficient mixing at micron-scale can not only give rise to a microstructure much different to that of the individual components, but one that cannot be achieved by mixing these components using conventional mixing techniques.

Highly efficient mixing at micron-scale can induce colloidal self-assemblies, based on supramolecular interactions such as electrostatic attractions, hydrophobic associations and hydrogen bonds. One has to look beyond edible formulations for an example. An ink containing solution of poly(sodium 4-styrene sulfonate), an anionic polyelectrolyte was co-printed using inkjet with another ink containing solution of poly(diallyldimethylammonium chloride), a cationic polyelectrolyte (Limem, McCallum, Wallace, In Het Panhuis, & Calvert, 2011). Their efficient mixing led to a thick and homogeneous gel of polyelectrolyte complex which could not have been prepared by any other method during similar timescale. Edible equivalents of these polyelectrolytes can potentially be used in jetting-based printing to make such edible supramolecular complexes. This can also be extended to coacervates of edible polyelectrolytes and surfactants as well as emulsion droplets. Supramolecular complexes are responsive to changes in solvent conditions such as pH, ionic strength and polarity. They are also effective as delivery vehicles for active compounds, and have been used for food and pharmaceutical/nutraceutical-based applications.

The full potential of jetting-based printing of edible materials can only be realised if the innovation in formulation of inks as described above moves forward along with, and makes full use of, the developments in the printing techniques.

Jetting Technique	Working Principle	Advantages	Disadvantages	References
Inkjet	Jetting caused by piezoelectric pressure pulses	Highly precise deposition (both position and quantity)	Only suitable for low-viscosity inks; relatively slow and expensive	(Godoi et al., 2016; Iftimi et al., 2019; Kyobula et al., 2017; Liu & Zhang, 2019; Liu et al., 2017; Negro et al., 2018; Pitayachaval et al., 2018; Voon et al., 2019)
Binder jetting	Powder particles being fused together by desired jetting of binding liquid	Expansion of the range of printable materials compared to inkjet; relatively quicker	Only powdered materials can be printed; expensive equipment	(Holland et al., 2018; Lam et al., 2002; Southerland et al., 2011; Yu et al., 2009)
Pneumatic jetting	Liquid ink under air pressure jetted through piezoelectrically controlled valve	Suitable for inks with much higher viscosity; relatively quicker and cheaper	Less precise compared to inkjet	(Gao et al., 2016; YanPu, 2016)
Electrostatic jetting	Jetting aided by voltage-induced electrostatic attraction towards the substrate	Suitable for inks with much higher viscosity; width of printed line can be tuned by applied voltage	Less precise compared to inkjet; inks must be responsive to electric voltage; relatively slow	(Suzuki, Takagishi, & Umezu, 2019; Takagishi, Suzuki, & Umezu, 2018)

Table 1. Summary of jetting-based printing techniques.

4.0 Conclusion

Jetting-based techniques offer high levels of precision in spatial deposition of droplets. Conventional piezoelectric inkjet has limitations in terms of viscosity of the inks which can be printed using it. This limitation, especially with regards to edible formulation has been addressed by approaches such as pneumatic jetting and electrostatic jetting.

In order to fully exploit the advances in deposition techniques, edible ink formulations need to incorporate supramolecular interactions which enable a self-supporting structure upon printing. This is essential as vast majority of chemical cross-linking agents promoting covalent bond formation are not edible.

At the current state of technology, 3D printing in general and jetting in particular is not as scalable as the conventional manufacturing technique for edible materials. It is important to understand that the aim for research in this area is to make it complimentary to the conventional manufacturing by enabling easy customisation in terms of shapes, textures and delivery of active compounds. The right place for food printers is therefore not in factories, but at the point of sales (where customisation is sought) e.g. retail outlets or pharmacies as well as households.

Enhanced formulations printed using novel deposition techniques have the potential to transform 3D printing technique from a mere tool for precise deposition to an aid in effective mixing which induces colloidal self-assemblies. This furthers the scope of 3D printing of edible materials beyond food, into healthcare applications.

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Highlights

- Various jetting-based 3D printing techniques are discussed.
- Edible formulations printed using jetting-based techniques are reviewed.
- Suggestions are made over how the edible formulations should evolve.

Conflict of Interest

The authors hereby declare no conflicts of interest with regards to the manuscript.

Credit Author Statement

Saumil Vadodaria: Literature search and writing. **Thomas Mills**: Writing, reviewing and editing.