Università di Foggia

Department of Science of Agriculture, Food and Environment

Doctorate on **Management of Innovation in the agricultural and food system ofthe Mediterranean Region**

Adding unprecedented economic and social values to the side- and by-products of Mediterranean fruit and vegetables by reshaping them in novel sources of nutrients and tailored food products mediated by 3D printing technology.

Giancarlo Colelli 26.07.2023 12:15:22 GMT+01:00

Coordinator:
Prof. Dr. Giancarlo Colelli

Supervisor:
Prof. Dr. Carla Severini

Carla Severini

PhD candidate:
Mehmet Onur ORAL

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Abstract

The escalating interest in the systematic and logical design of food has recently emerged, driven by the evolving understanding of the intricate relationship between the 3D structure of food at both macro- and microscales and their nutritional, sensory, and physical properties. This approach has found significance in various areas, including the connection between food texture and mastication issues in the elderly, sensory perception during consumption, bio-accessibility of nutrients, and mass and heat transfer. Furthermore, this method allows for precise ingredient dosing to meet specific nutritional needs, paving the way for personalised sensorial and nutritional food products.

The rational design of food is directly related to promoting a healthier and more sustainable food system by amplifying health benefits, reducing food waste, and fostering sustainable consumption. 3D Food Printing (3DFP) emerges as a viable technology to achieve this ambitious goal, as it uniquely converts digital 3D models into tangible food products.

This doctoral thesis offers novel insights to unlock the full potential of 3DFP, a promising technology set to revolutionise food production and consumption. The research objectives are threefold:

1) To delineate the state of the art through advanced bibliometric methodologies in the past decade;
2) To enhance the control over 3D printing movements and utilise this technology as a fresh strategy for food prototyping;
3) To investigate, test, and validate the versatility of 3DFP in creating nutrient-enriched ink-gels from by-product ingredients.

Particularly, this thesis addresses the transformative potential of 3DFP in creating unprecedented economic and social values from the side- and by-products of Mediterranean fruits and vegetables. By reshaping these into novel sources of nutrients and tailored food products, we can maximise the utilisation of these often-discarded elements, thus contributing to a circular and sustainable economy.

This thesis comprises seven chapters, with the first providing an overview of the 3DFP landscape, its potential and challenges for industrial or individual applications. The remaining chapters include scientific papers already published in international peer-reviewed journals or submitted for potential publication.

Introduction

Current scenario and challenges for the food system

Food loss, waste, population growth, and global warming pose significant challenges to food sustainability and have extraordinary negative impacts on the environment, economy, and public health. As the global population continues to grow, with an estimated 9.7 billion people estimated by 2050 (United Nations, 2019), the demand for food is expected to increase continuously. At the same time, the loss and food waste (FLW) at various stages of the food system and the effects of global warming on agricultural productivity and natural resources are exacerbating the lack of the right to food.

Food loss, which refers to the reduction of edible food occurring during production, post-harvest handling, storage, and food waste, which refers to the edible food discarded or uneaten at the consumer level or during distribution and retail (Cohen et al., 2015), result in significant amounts of lost and wasted nutrients. These losses are particularly concerning in the context of malnutrition and the inadequate fulfilment of daily nutritious diets, which continue to affect millions worldwide. According to the Global Nutrition Report, a quarter of the global population is malnourished, with deficiencies in essential nutrients such as vitamins, minerals, and proteins (Global Nutrition Report, 2020). Moreover, the loss and waste of food contribute to the food system's inefficiency, further exacerbating malnutrition and nutrient deficiencies.

The effects of population growth and global warming compound food loss and waste. As the global population continues to rise, the demand for food increases, putting additional strain on agricultural production and natural resources (United Nations, 2019). Moreover, global warming, driven by the accumulation of greenhouse gases in the atmosphere, has detrimental consequences on agricultural productivity, water availability, and weather patterns, affecting food production, distribution, and storage (Kummu et al., 2012). Furthermore, these factors increase the challenge of providing adequate nutrition to the growing global population.

Agriculture and food production are vital sectors in the Mediterranean region, characterised by a diverse range of fruits and vegetables. However, traditional agricultural practices often generate substantial amounts of agricultural residues or by-products, including peels, seeds, stems, and other by-products, often discarded or underutilised, leading to economic losses and environmental waste (Zhang et al., 2020). Such materials pose resource utilisation, food waste, and sustainability challenges, requiring innovative solutions to reduce environmental pressure.

In the Mediterranean region, agricultural side- and by-products from fruit and vegetable crops present a unique yet underutilised resource. These materials, often discarded or neglected, contain valuable nutrients and compounds that can be harnessed to improve the nutritional profile of food products. Through 3D Food Printing technology, these side- and by-products can be transformed into innovative food inks, adding nutritional value and contributing to waste reduction, thereby enhancing sustainability. In this light, 3D Food Printing technology offers a compelling solution to capitalise on these resources. By reshaping these side- and by-products into novel, nutrient-rich food products, the technology can contribute to unprecedented economic and social values while addressing sustainability concerns and promoting healthier food options. This approach aligns perfectly with the global shift towards a circular economy, where resource efficiency and waste minimisation are critical. In the following chapters, this thesis will explore how 3D Food Printing technology can unlock the full potential of Mediterranean fruit and vegetable by-products, presenting a new frontier in food science and technology.

The Mediterranean region has a long history of food production, and the Mediterranean diet has been recognised for its health benefits, including reduced risks of chronic diseases such as cardiovascular diseases and cancer (Martinez-Gonzalez et al., 2019; Sofi et al., 2010). However, changing dietary patterns, urbanisation, and globalisation have led to shifts in food consumption patterns, resulting in increased consumption of highly processed and less nutritious foods (Mistretta et al., 2018; Burlingame et al., 2016). This situation has raised concerns about nutrition and health in the Mediterranean area.

In recent years, there has been increasing interest in finding ways to add value to agricultural residues and by-products to transform them into novel sources of nutrients and tailored food products.

3D food printing is an innovative technology that has been gaining considerable attention in recent years due to its unique opportunities, such as the capability to use different kinds of ingredients, also from by-products, to enhance the nutritional properties of the end-products (Ahn et al., 2016; Passos et al., 2019). The high flexibility of the 3DFP technology, which enables the employment of ingredients in different forms – i.e., powder, gels, paste, etc. – may contribute to better sustainability of the food sector. For instance, food powder from agricultural by-products or inedible parts of fruit and vegetable has been used to create printable food ink and generate innovative and more sustainable food products (Tatiana et al., 2021). This promising technology is increasingly being recognised as a sustainable solution that facilitates the incorporation of alternative ingredients in food processing, thereby contributing to the circular economy and waste reduction.

For example, insects have been widely investigated as potential substitutes for conventional protein sources in 3DFP, offering a sustainable and nutritionally enriched food alternative (Severini et al., 2018a). This approach addresses the growing demand for protein and mitigates the environmental impact of traditional livestock farming.

Moreover, considering its flexibility in using different materials, 3DFP has been leveraged to add value to food waste, often discarded due to aesthetic standards or by-products generated during processing. 3DFP can help reuse and recycle these materials by using edible powders to be included in the food ink (Bhandari, 2016). This potential capability enables the utilisation of alternative ingredients and waste resources and reshapes our food systems towards greater sustainability and nutritional enhancement.

The technical background of 3D food printing (3DFP)

3DFP consists of a layer-by-layer deposition process of edible materials capable of replicating, in the physical world, a 3D digital model. Considering the exploitation in the food sector, such technology enables the creation of complex food structures with a high level of customisation, offering opportunities for innovative food product development, personalised doses of nutrient and sensory properties. For instance, 3DFP can create customised shapes and textures, enhancing the final product's aesthetic appeal (Hussein S., 2021). Additionally, market innovations can be obtained by 3DFP by creating complex food structures that may be difficult or impossible to produce with traditional food manufacturing techniques (Dong et al., 2022). Dysphagia, or difficulty swallowing, is prevalent in older people and patients suffering from debilitating illnesses. These printers can improve the visual appeal of pureed diets, often required for dysphagic patients, and enable greater food uptake to prevent malnutrition (Pant et al., 2021). Considering the opportunity to better address the requirements for high nutritional quality based on individual requirements, 3DFP can create low-calorie or low-fat foods or, in addition, incorporate personalised doses of specific vitamins or minerals into the food product. It can be particularly beneficial for individuals with food allergies, sensitivities, or other health conditions that require strict dietary restrictions.

3DFP consists of several potential technologies with different printing movement methods and the approach utilised for material depositions. Among these, we want to remember different types of 3DFP.

1. **Extrusion-based printing (Fused Deposition Modeling):** This method is widely used in 3DFP. It involves pushing materials such as liquid or paste through a moving nozzle, which cools and solidifies to form layers (Figure 1). This method suits soft ingredients like chocolate, dough, mashed potatoes, cheese, and meat paste. However, it has limitations in creating complex and delicate forms due to the inherent risk of distortion (Lanoro et al., 2017; Liu et al., 2018).

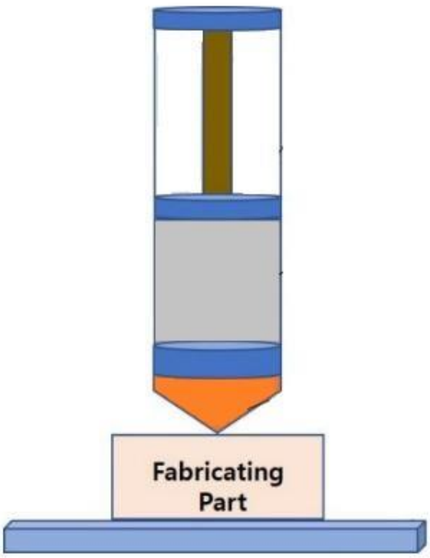

Figure 1: Extrusion-based 3DFP.

2. **Selective Laser Sintering (SLS):** This technique involves applying powder-type materials to a bed and then using a laser to solidify only the desired part. It forms a shape by hardening only the part exposed to the laser. This method is suitable for food ingredients in powdered form, such as sugar and starch. Adding food additives like artificial pigments and

fragrances can produce various colours and flavour outputs. However, there are safety considerations due to using lasers (Figure 2) (Kim et al., 2016; Kim et al., 2020).

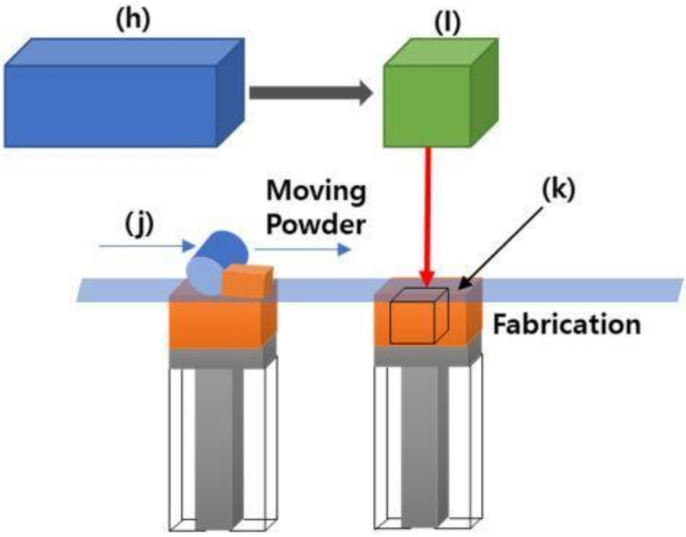

Figure 2: Selective Laser Sintering (SLS) 3DFP.

3. **Color Jet Printing (CJP):** This method uses a print head to distribute a binder into a powder layer selectively. It is cheaper than other 3D printers and can create complex geometries without artificial support structures (Figure 3). However, it requires the removal of the remaining ambient powder and hardening the prototype surface (Kim et al., 2016).

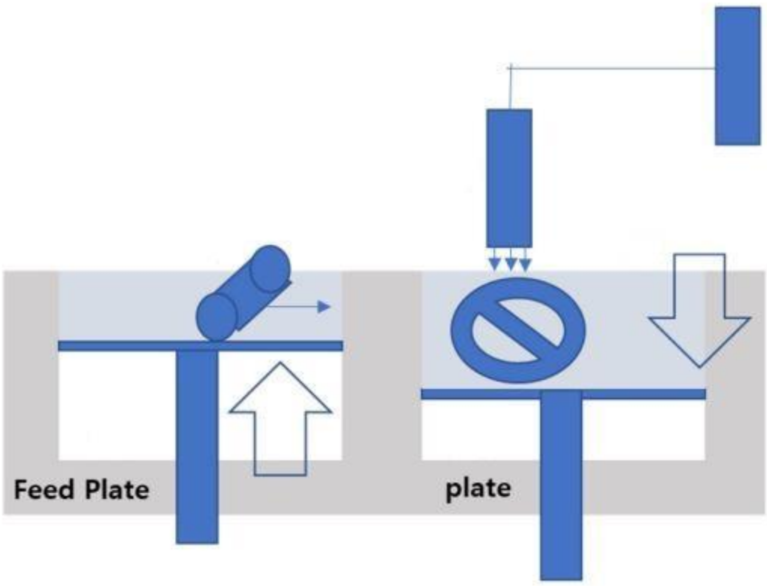

Figure 3: Color Jet 3DFP

4. **Stereolithography (SLA) and Digital Light Processing (DLP):** These methods use liquid photocurable resins and perform chemical reactions during light irradiation to produce solids. They can produce high-resolution and excellent surface-quality components. However,

they require dedicated hardening resins and are typically used to create small models requiring high precision (Figure 4) (Krkobabi'c et al., 2020).

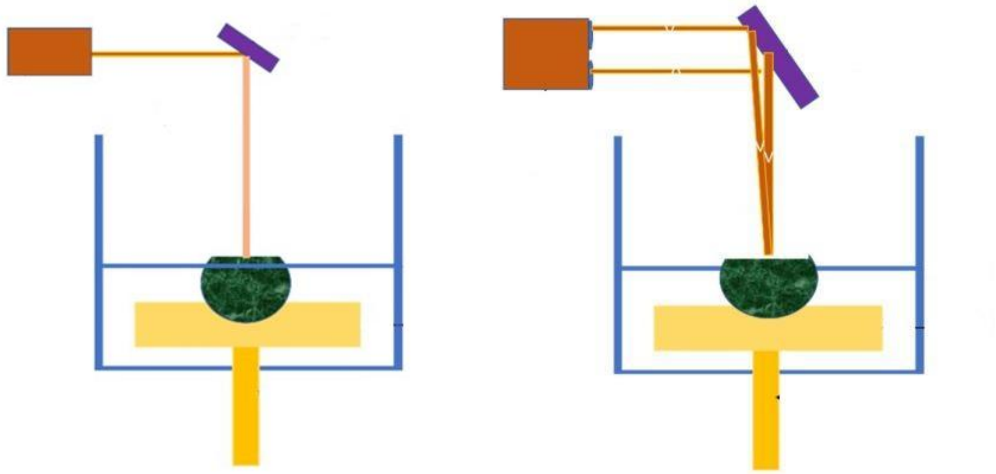

Figure 4: Stereolithography (SLA) (left) 3DFP and Digital Light Processing (DLP) (right) 3DFP.

However, the most common approach is using an extrusion-based printer consisting of a syringe filled with the printable food formula and the plunger, which applies the force necessary to deposit filament of the ink-food in the layer-by-layer modalities. On this basis, the design and development of these food inks is a critical step and a vast area of study, as they determine the rheological and mechanical properties of the food, thereby affecting the final quality of the printed food (Voon et al., 2019).

To obtain a precise replica of the 3D digital model, several steps of the 3D printing process needed to be designed, tested, and validated. At first, the pre-printing phase consisting of creating a suitable 3D digital model, is crucial (Sun et al., 2018), which is generally performed using common CAD software. Then, selecting food materials is another critical aspect to consider in 3D food printing. The properties of these materials, such as their rheological behaviour, nutritional content, and sensory attributes, significantly influence the quality and acceptability of printed food (Lipton et al., 2015). Therefore, a thorough understanding of these properties is essential for successful 3DFP. The printability of the food material is also defined as the ability to be printed without compromising the quality and the structural stability of the 3D-printed food structure (Derby, 2010), which is largely affected by its rheological properties. Identifying optimal printing conditions is also crucial in the 3D food printing process. Being controlled by several parameters such as printing speed, layer height, and nozzle diameter, which must be carefully calibrated to ensure the successful printing of the food products (Yang et al., 2018). In addition, generating the G-code, a language instructing the 3D printer on creating the desired object is another critical element in 3D food printing. The G-code dictates the printer's movements and the material deposition, thereby determining the shape and structure of the printed food (Ngo et al., 2018). Finally, the post-printing process, which may involve cooking or other forms of treatment, is an essential step that can significantly affect the quality and safety of printed food (Severini et al., 2016).

Here we want to analyse better the role and the importance of the printability of food formulas and, in addition, the need to control the printing movement precisely. These two points are largely analysed in the PhD thesis.

The rheological properties of food inks play a cardinal role in the success of 3D food printing, exerting a profound influence on both the printability and the ultimate quality of the printed food. Viscosity, yield stress, and thixotropy are highly significant among these properties.

Viscosity, defined as a fluid's resistance to shear or flow, is instrumental in determining the food ink's capacity to maintain its shape post-extrusion. It is a critical parameter that governs the flow behaviour of the food ink during the printing process. High viscosity ensures that the printed material retains its shape after deposition, but it also necessitates a higher force for extrusion from the printing nozzle. Therefore, an optimal balance is required to ensure efficient printing and shape retention (Dankar et al., 2018; Zhu et al., 2019). Yield stress, the minimum stress required to induce flow in a material, is indispensable for the material to hold its shape after printing. The yield stress prevents the printed layers from collapsing under their weight, thereby maintaining the structural integrity of the printed object. A material with a high yield stress behaves more like a solid until the yield stress is exceeded, after which it flows like a liquid. This property is particularly important in 3D food printing, where the printed structure needs to retain its shape until it is set or cooked (Sun et al., 2018; Chen et al., 2019). Thixotropy, the time-dependent shear thinning property, is also crucial in 3D food printing. It allows the material to be easily extruded during printing but regain its rigidity afterwards. This property benefits material that needs to be fluid during extrusion but solidifies quickly once printed. 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. The composition and concentration of ingredients, the interaction between them, and the processing conditions can all influence the rheological properties of the food ink (Godoi et al., 2016; Panda et al., 2018; Yang et al., 2018). Additives play a significant role in these inks, often used to modify their properties to meet specific requirements. For instance, thickeners can increase viscosity, gelling agents can enhance yield stress, and other additives can improve thixotropy. Understanding these additives' roles and interactions is crucial for developing food inks with desired properties.

Despite the advancements in 3D food printing, challenges must be addressed. These include the limited range of printable materials, difficulty controlling the texture and taste of printed food, and slow printing speed. However, the field also presents numerous opportunities, such as the ability to create personalised food, the potential for reducing food waste, and the possibility of creating novel food structures that are impossible with traditional manufacturing methods (Dankar et al., 2018; Zhu et al., 2019). The rheological properties of food inks are a critical aspect of 3D food printing, influencing the printability and final quality of the printed food. Further research in this area will pave the way for developing new food inks and advancing 3D food printing technology (Figure 5).

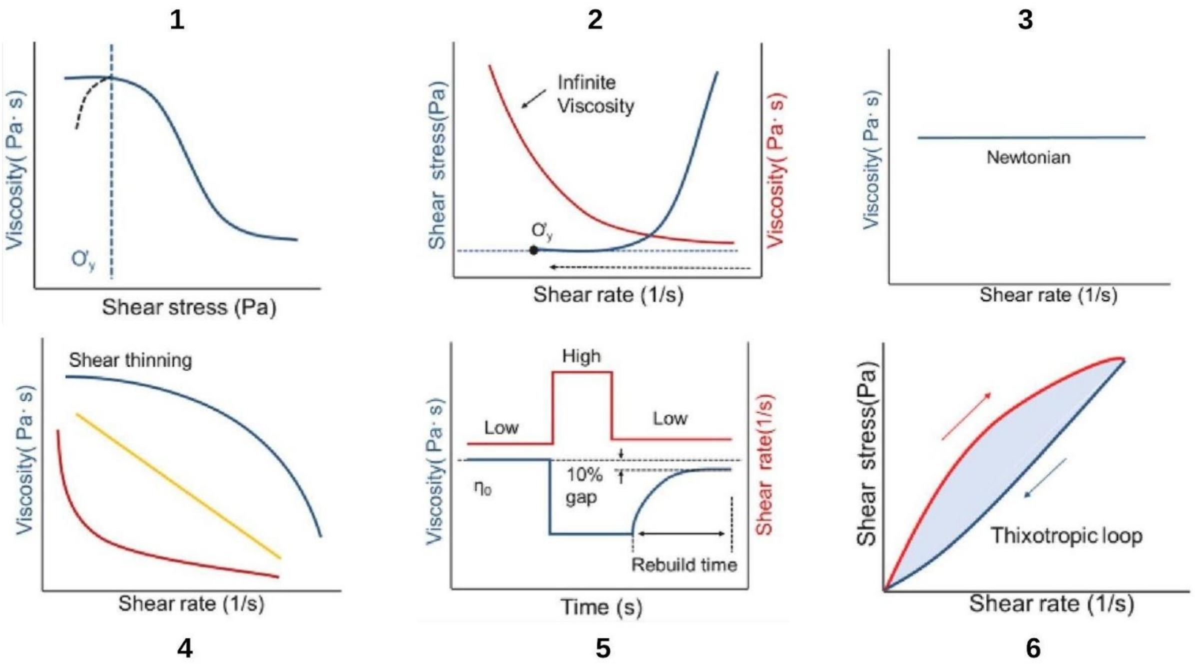

Figure 5: Rheology of edible inks for printing. Yield stress **(1-2),** non-Newtonian shear-thinning, shear-thickening and Newtonian behaviours **(3-4),** thixotropy test and thixotropic loop test **(5-6).**

Many printing variables drive/control the movements of the printing in the X, Y, and Z axes and, in addition, the amount of materials deposited per unit of time. For instance, the nozzle size plays a crucial role in determining the resolution and accuracy of the printed product, with smaller nozzles enabling finer details (Severini et al., 2016; Liu et al., 2018; Yang et al., 2018; Yang et al., 2018b; Severini et al., 2018; Derossi et al., 2019). Layer height, another critical parameter, affects the texture and structural integrity of the printed food with thinner layers often resulting in smoother surfaces and more robust structures (Severini et al., 2016; Liu et al., 2018; Yang et al., 2018a; Yang et al., 2018b; Severini et al., 2018; Derossi et al., 2019). The print speed, however, can significantly impact the printing process's efficiency and the final product's quality. Faster print speeds may lead to increased productivity but can also compromise the printed food's quality and accuracy (Kim et al., 2019; Vancauwenberghe et al., 2017; Lanaro et al., 2017; Le Tohic et al., 2017; Derossi et al., 2018a; Derossi et al., 2019). Infill level, which refers to the density of the interior of the printed object, is another key variable. Higher infill levels can enhance the structural strength and weight of the printed food but may also increase the printing time and material usage (Zhang et al., 2018; Derossi et al., 2019). A study by Alhnan et al. (2019) further elucidates the impact of these printing variables. The study found that the extrusion multiplier, an empirical value proportional to the extrusion rate and the speed of needle movement, was the most influential factor affecting the dispensed dose in 3D-printed oral dosages. The study also highlighted that the nozzle size significantly impacts the dose, with larger needles resulting in higher doses due to reduced resistance from the increased internal diameter of the needle. These findings underscore the importance of carefully calibrating printing variables to achieve optimal outcomes in 3D food printing. By understanding and manipulating these variables, it is possible to tailor the printing process to produce food products with desired attributes, thereby expanding the possibilities of this innovative technology (Callejo et al., 2019). Understanding and optimising these aspects is important for achieving desired printing outcomes, including accurate and consistent layer-by-layer deposition of the ink materials, resulting in well-defined and structurally sound food products. They can impact the ink material's flow behaviour and structural integrity during printing (Callejo et al., 2019). Therefore, careful consideration and optimisation of these factors are necessary for successful 3D food printing. The printability of food products can be influenced by various factors, including the rheological properties of the ink materials, the design of the digital models, and the printing process parameters (Derossi et al., 2016).

All such variables are generally individuated and defined using a slicing software that literally slices the 3D model in different layers, and it defines all printing movements and the conditions for depositing the food formula. Then, a G-Code consists of the information instructing the printer how to move during the manufacturing process. In 3D food printing, G-codes guide the printer's movements, such as the path to follow, the extrusion speed and the layer height, among other parameters.

The slicing software considers the printer's specific characteristics, such as the nozzle size and material type. This process is crucial for achieving the printed food item's desired shape, texture and other properties (Lipton et al., 2015). The G-codes guide the printer's movements, such as the path to follow, the extrusion speed and the layer height, among other parameters. Generating G-code for 3D food printing involves using specialised CAD software to create a digital model of the desired food product and converting it into G-code to provide instructions to the 3D printer. Parameters such as layer height, extrusion speed, temperature, and infill density need to be carefully defined in the G-code. Proper calibration and optimisation of printing parameters are critical for achieving the final food product's desired print quality and functionality. Popular software for creating digital models include Blender, Tinkercad, and Fusion 360, while slicing software such as Cura, PrusaSlicer, and Simplify3D are commonly used for generating G-code.

Three-dimensional (3D) food printing has emerged as a promising technology that has the potential to revolutionise the food industry by enabling the creation of complex food structures with high customisation. However, achieving high-quality and precise prints in 3D food printing can be challenging due to food materials' unique properties and requirements. In this context, using an innovative G-Code Designer for advanced G-code generation control can significantly improve printed food structures' quality and precision. FullControl Gcode Designer is a novel tool never used in food applications. It empowers the user to define every segment of the print path along with all printing parameters at every point along the print path. This novel approach enables the software to directly generate machine control code (Gcode) without the need for programming skills, CAD software, STL files, or slicing software. The user is free to create non-planar 3D print paths and overcome traditional restrictions of layer-wise print path planning (Gleadall, 2021). This approach also enables nozzle movements to be designed carefully, both during extrusion and while travelling between disconnected extrusion volumes, to improve the capabilities of the printing process or to overcome inherent limitations. Several case studies (Gleadall, 2021) have demonstrated the broad range of structures that can be designed using this software (Figure 6). For instance, figure 7 shows that the software can produce precisely controlled specimens for printer calibration, parametric specimens for hardware characterisation using hundreds of unique parameter combinations, novel mathematically defined structures, and previously inconceivable 3D geometries that are impossible to achieve using traditional slicing software (Gleadall, 2021).

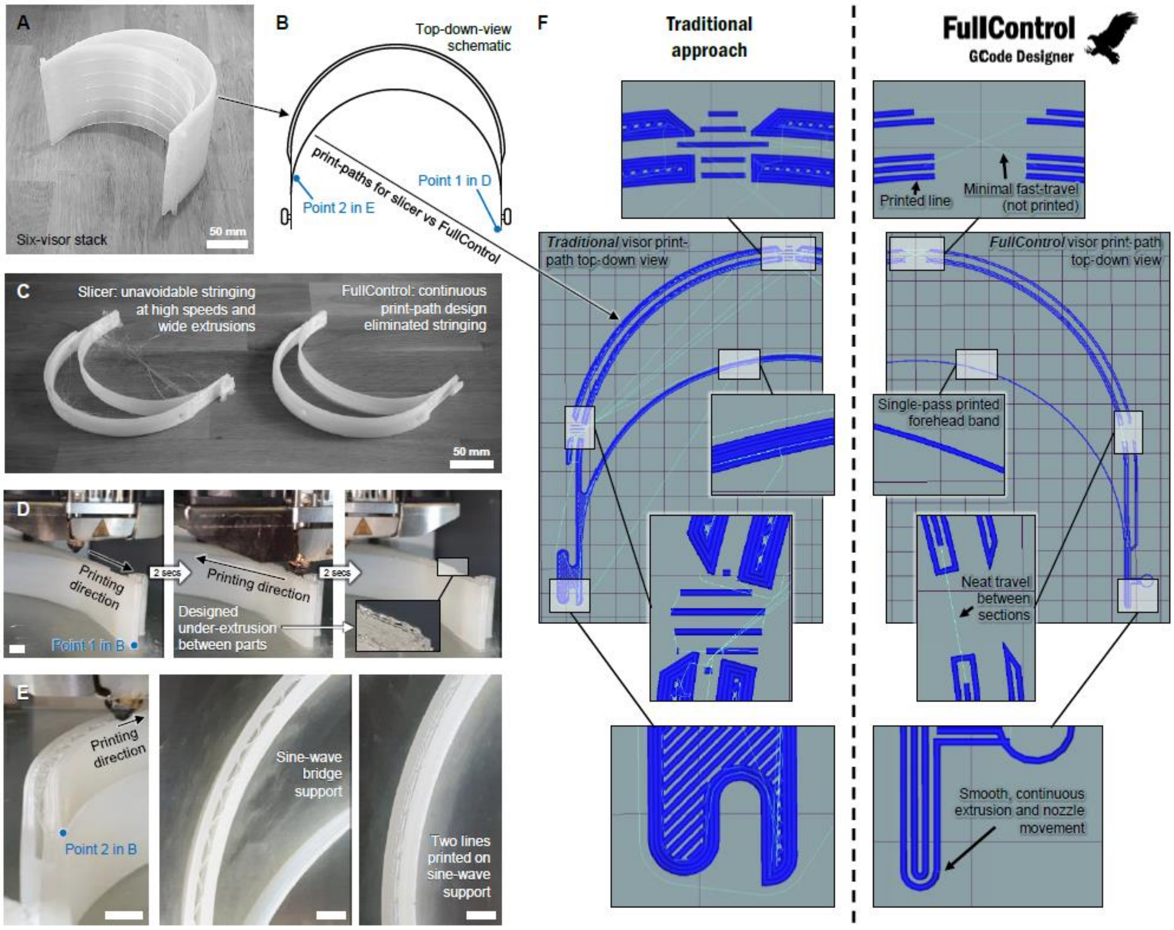

Figure 6: Comparison between traditional slicer software and FullControl print paths (Gleadall, 2021).

This new design approach is particularly useful to improve additive manufacturing operations, where traditional slicing software typically limits the user's capabilities to specific structures and printing parameters. The FullControl Gcode Designer allows the user unconstrained freedom to instruct every aspect of the printing procedure to achieve the desired part, including controlling auxiliary equipment or novel tool heads in multi-tool printers. This approach enables system calibration, design of defect-free parts, or development of currently inconceivable structures. Combining "FullControl Gcode Designer" with 3D food printing can offer several benefits in improving the quality and precision of printed food structures:

1. It allows precise control over the extrusion process, which is crucial for achieving uniform layering and consistent printing results.
2. It enables the optimisation of printing parameters to match the unique properties of food materials, such as viscosity and texture, resulting in improved print quality and structural integrity.

3. The customisation capabilities of “FullControl Gcode Designer” allow to the creation of personalised food structures with tailored designs, enhancing the aesthetics and the presentation of the final printed products.

Integrating “FullControl Gcode Designer” with 3D food printing can advance state of the art in food fabrication and open new possibilities for innovative applications in the food industry. By enabling higher quality and precision in 3D food printing, this integration can contribute to developing customised, visually appealing, and nutritionally optimised food products that cater to individual preferences and dietary needs. Further research and experimentation with this combined approach are warranted to explore its full potential and accelerate the adoption of 3D food printing in the food industry.

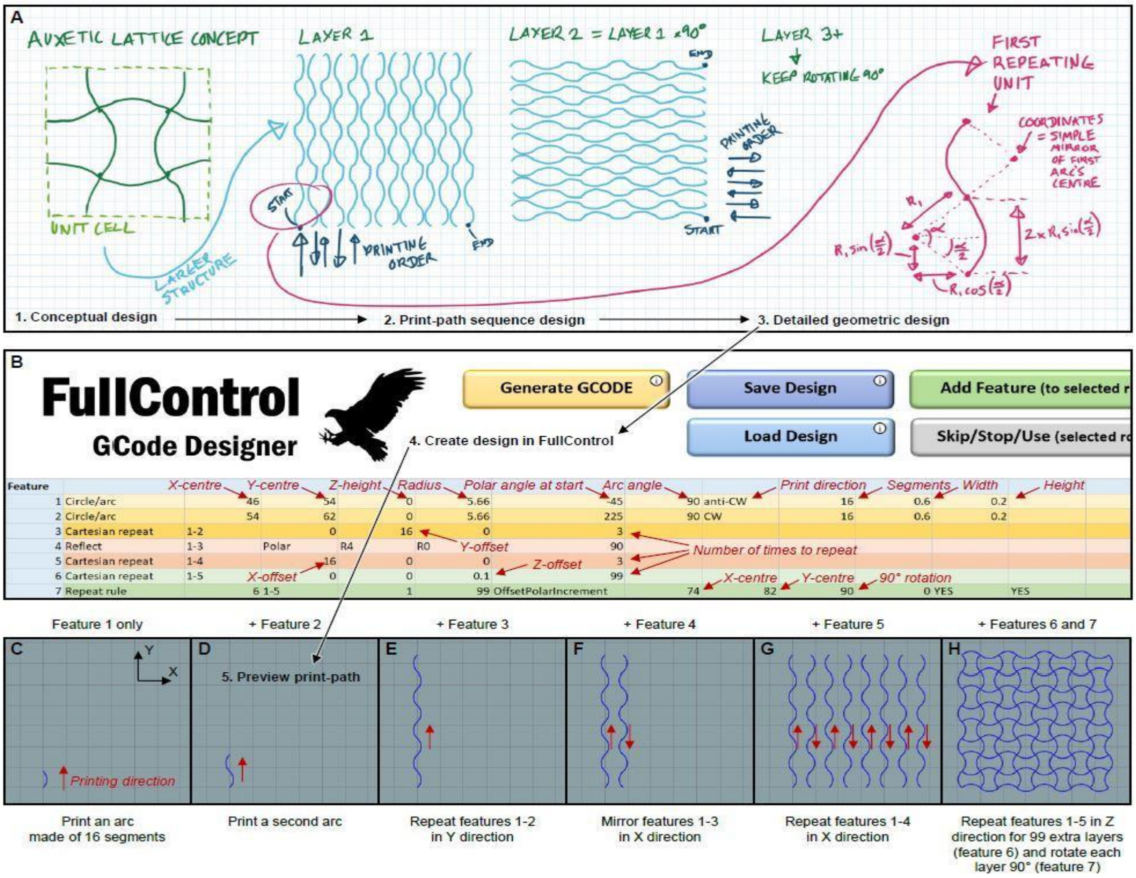

Figure 7: Comparison between traditional slicer software and FullControl print paths

Therefore, this process must be carefully managed to ensure the production of safe and high-quality 3D-printed food. In the preliminary stage of the development of 3DFP, the majority of the publications have been focused on the study of food formulas employable as food ink and the effect of their rheological properties (Cohen et al., 2009; Kim et al., 2019; Caporizzi et al., 2019). So, several types of food materials have been used as food inks, such as chocolate (Mantihal et al., 2019), doughs (Pulatsu et al., 2020), fruit and vegetable blends (Kim et al., 2018), fish and seafood products (Wang et al., 2018b), poultry (Wilson et al., 2021) and dairy products and derivatives (Le Tohic et al., 2018; Liu et al., 2019b). These materials are selected based on their rheological properties, sensory attributes, and nutritional characteristics to achieve the printed food products' desired texture, flavour, and nutritional composition (Figure 8).

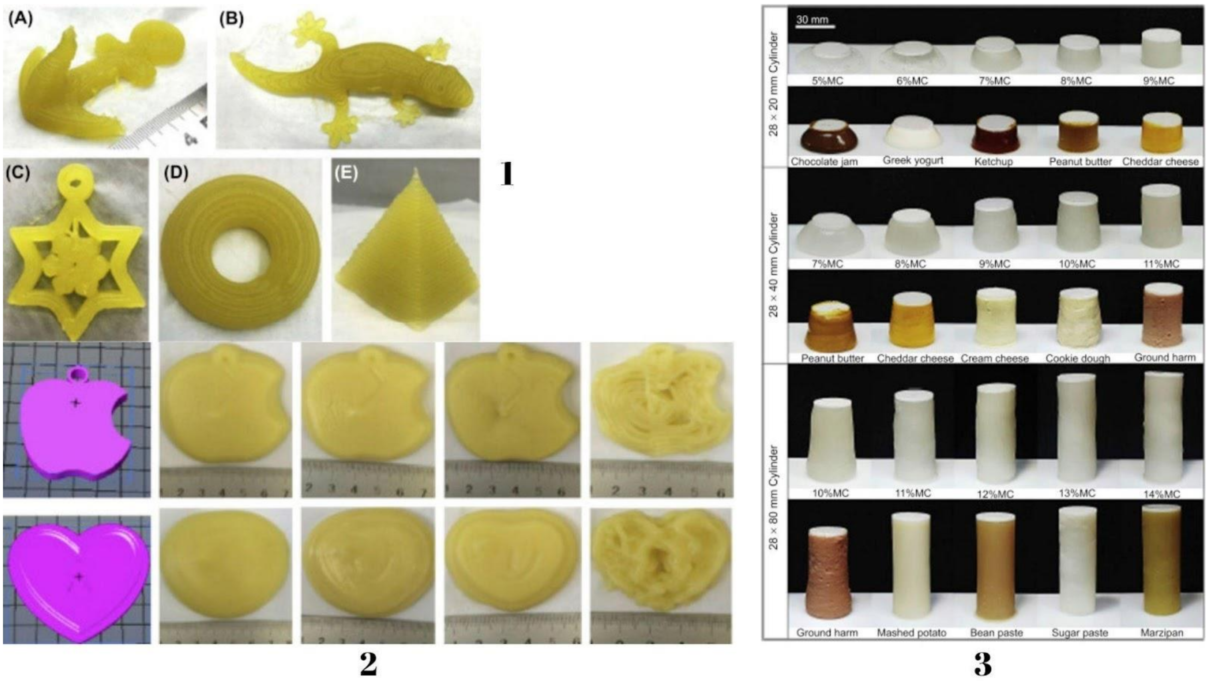

Figure 8: Lemon juice gel objects, Yang et al., 2018 **(1)**, dough objects, Liu et al., 2018 **(2)**, different methylcellulose concentration printed objects Kim et al., 2018 **(3).**

Research questions

Based on the above considerations, the following research questions arise:

- What are the key factors affecting the quality and precision of 3D-printed food structures?
- How can integrating specialised CAD software and G-code generation tools, such as "FullControl GCode Designer", optimise the printing conditions and customisation of food product design in 3D food printing?
- What are the potential benefits of using 3D food printing in creating customised and nutritional-optimised food products for individuals with specific dietary needs or preferences?
- How does using different ink materials and printing types impact the printability and quality of 3D-printed food structures?
- What are the challenges and limitations of 3DFP technology, and how can they be addressed to improve the quality and feasibility of printed food products?
- How can 3DFP contribute to reducing food waste and improving the efficiency of food production and distribution systems?

The research aims and objectives

The research activities are designed to improve scientific knowledge in 3DFP applications by developing innovative food formulations and processing techniques and utilising food waste and its by-products in the Mediterranean region. In addition, the potential of specialised computer-aided design (CAD) software and G-code generation tools, such as "FullControl GCode Designer," will be explored to optimise the printing parameters, printing conditions, and food product designs. The objective is to enhance the quality and precision of 3D-printed food structures. The proposed research activities will contribute to creating novel sources of nutrients and food products using 3D printing technology, reducing food waste, and increasing the value of Mediterranean food products.

The research consisted of three main sections of experimental activities.

Section A: A comprehensive analysis of the current state of the art of the 3DFP has been performed employing bibliometric analysis and data visualisation techniques. The aim was to identify emerging trends, collaborations, and knowledge gaps in the existing literature. This process highlighted new possibilities and opportunities in the field of 3DFP.

Section B: This section is dedicated to a better understanding of the effect of 3D food printing technology on some undervalued physical and technological aspects. More specifically, the impact of 3DFP on the 3D microstructure of food is examined, as well as its interrelation with the texture properties of cereal-based food products. Also, 3DFP was studied as a prototyping method to define the best 3D structure for reducing the kinetic of baking 3D printed biscuits. Furthermore, for the first time, an innovative GCode designer was tested in the food sector and compared with conventional slicing software by examining the quality of 3D-printed food and the efficiency of the printing process.

Section C: This section of the research was dedicated to the potential benefits of 3D food printing on the sustainability of the food sector through better exploitation of uncommon ingredients obtained from food waste and by-products. To do this, after describing the rheological properties of innovative ink-gel and testing its printability, such materials have been nutritionally enriched by introducing vegetable powders from different fruit and vegetable by-products. The aim was to understand how these by-products could be reintegrated into the food supply chain and how specific ingredients and the printing process influenced the properties of gel formulations.

The doctoral research endeavoured to pioneer methodologies in three-dimensional food printing, a potential paradigm shift that could revolutionise the culinary industry. This advancement may significantly contribute to overarching waste minimisation and sustainability objectives, profoundly impacting environmental stewardship. Despite facing limitations and challenges, the goal is to inspire further research and development in this field, contributing to the evolution of the food system towards greater sustainability, efficiency, and personalisation.

Expected outcome.

The integration of 3D food printing technology is expected to yield various benefits, such as enhanced precision and quality of printed food structures, improved printability, increased innovation, and creativity in food product design, and expanded market opportunities. This integration has the potential to transform the food industry by enabling the creation of unique and customised food products, driving innovation, and opening new market opportunities. Furthermore, 3DFP can address the issue of food waste and by-product utilisation, leading to the development of sustainable food products. Food manufacturers can adopt circular economy principles by optimising the use of by-products, reducing waste, and increasing sustainability. This situation can result in visually appealing, customisable, and nutritionally optimised value-added products that cater to the demand for healthy and sustainable food options. Further research in this area can lead to the development of novel food products that promote sustainable food practices in the Mediterranean region and beyond.

Research outline

- **Introduction**

- **Chapter 1: Drawing the scientific landscape of 3D Food Printing. Maps and interpretation of the global information in the first 13 years of detailed experiments, from 2007 to 2020.**

- **Chapter 2: Analyzing the most promising innovations in food printing. Programmable food texture and 4D foods. Drawing the scientific landscape of 3D Food Printing. Maps and interpretation of the global information in the first 13 years of detailed experiments, from 2007 to 2020.**

- **Chapter 3: Analyzing the effects of the 3D printing process per se on the microstructure and mechanical properties of cereal food products.**

- **Chapter 4: Accelerating the process development of innovative food products by prototyping through 3D printing technology.**

- **Chapter 5: Unconstrained freedom for designing and building 3D printed food.**

- **Chapter 6: Utilising Mediterranean food waste and by-products in 3D Food Printing.**

- **Chapter 7: Conclusion**

- **Chapter 8: References**

References

1. Ahn, J., Ahn, H. S., Cheong, J. H., & Dela Peña, I. (2016). Natural product-derived treatments for attention-deficit/hyperactivity disorder: Safety, efficacy, and therapeutic potential of combination therapy. Neural Plasticity, 2016, 1320423. https://doi.org/10.1155/2016/1320423

2. Alhnan, M. A., Okwuosa, T. C., Sadia, M., Wan, K. W., Ahmed, W., & Arafat, B. (2016). Emergence of 3D printed dosage forms: Opportunities and challenges. Pharmaceutical Research, 33(8), 1817-1832. https://doi.org/10.1007/s11095-016-1933-1

3. Bhat, Z.F., Bhat, H., Pathak, V., 2014. Prospects for *In Vitro* cultured meat – A Future Harvest.principles of tissue engineering (4th edition), 79, 1663 – 1683. https://doi.org/10.1016/B978-0-12-398358-9.00079-3.

4. Burlingame, G. M., Seebeck, J. D., Janis, R. A., Whitcomb, K. E., Barkowski, S., Rosendahl, J., & Strauss, B. (2016). Outcome differences between individual and group formats when identical and nonidentical treatments, patients, and doses are compared: A 25-year meta-analytic perspective. Psychotherapy, 53(4), 446–461. https://doi.org/10.1037/pst0000090

5. Caporizzi, R., Derossi, A., Severini, C., Azzollini, 2019. Chapter 4; Cereal-based and insect-enriched printable food: from formulation to postprocessing treatments, status and perspectives. Fundamentals of 3d food printing and applications, 93 – 116. https://doi.org/10.1016/B978-0-12-814564-7.00004-3.

6. Cohen, A., Laviv, A., Berman, P., Nashef, R., Abu-Tair, J., 2009. Mandibular reconstruction using stereolithographic 3-dimensional printing modelling technology. Oral surgery, oral medicine, oral pathology, oral radiology and endodontology, 5, 661 – 666. https://doi.org/10.1016/j.tripleo.2009.05.023.

7. Cohen, S., Martinez, H., & Schroder, A. (2015). Waste Management Practices in New York City, Hong Kong and Beijing. ALEP Waste Manage. Retrieved from https://www.swsu.ru/sbornik-statey/pdf/ALEP%20Waste%20Managent, 20.

8. Dankar, I., Haddarah, A., Omar, F.E.L., Sepulcre, F., Pujola, M., 2018. 3d printing technology: the new era for food customization and elaboration. Trends in food science & technology, 75, 231 – 242. https://doi.org/10.1016/j.tifs.2018.03.018.

9. de Andrade, G. A., Vazquez, R., Pagano, D. J., & Mascheroni, J. M. (2019). Design and implementation of a backstepping controller for regulating temperature in 3D printers based on selective laser sintering. In 2019 IEEE 58th Conference on Decision and Control (CDC) (pp. 1183-1188). IEEE. https://doi.org/10.1109/CDC40024.2019.9029473

10. Derby, B. (2010). Inkjet Printing of Functional and Structural Materials: Fluid Property Requirements, Feature Stability, and Resolution. Annual Review of Materials Research, 40, 395-414. https://doi.org/10.1146/annurev-matsci-070909-104502

11. Derossi, A., Caporizzi, R., Azzollini, D., Severini, C., 2018b. Application of 3d printing for customized food. A case on the development of a fruit-based snack for children. Journal of food engineering, 220, 65 – 75. https://dx.doi.org/10.1016/j.foodeng.2017.05.015.

12. Derossi, A., Severini, C., Kirill, M.G., Karsanina, M.V., Nicolai, B., Verboven, P., 2019. Mimicking 3d food microstructure using limited statistical information from 2d cross-sectional image. Journal of food engineering, 241, 116 – 126. https://doi.org/10.1016/j.foodeng.2018.08.012.

13. Derossi, A., Severini, C., Ricci, I., Caporizzi, R., Fiore, A., 2018a. Printing a blend of fruit and vegetables. New advances on critical variables and shelf life of 3D edible objects. Journal of food engineering, 220, 89 – 110. https://doi.org/10.1016/j.foodeng.2017.08.025.

14. Dong, H., Wang, P., Yang, Z., & Xu, X. (2023). 3D printing based on meat materials: Challenges and opportunities. Current Research in Food Science, 6, 100423. https://doi.org/10.1016/j.crfs.2022.100423

15. Einat, M., & Bar-Levav, E. (2015). 2D segmented large inkjet printhead for high speed 3D printers. Journal of Micromechanics and Microengineering, 25(5), 055012. https://doi.org/10.1088/0960-1317/25/5/055012

16. El-Ramady, H., Brevik, E. C., Elbasiouny, H., et al. (2021). Planning for disposal of COVID-19 pandemic wastes in developing countries: A review of current challenges. Environmental Monitoring and Assessment, 193, 592. https://doi.org/10.1007/s10661-021-09350-1

17. Fay, L.B., German, J.B., 2008. Personalizing foods: is genotype necessary?. Current opinion in biotechnology, 2, 121 – 128. https://doi.org/10.1016/j.copbio.2008.02.010.

18. Gan, J., German, J.B., Siegel, J.B., 2019. Molecular annotation of food – Towards personalized diet and precision health. Trends in food science & technology, 91, 675 – 680. https://doi.org/10.1016/j.tifs.2019.07.016.

19. Garcia – Herrero, I., Margallo, M., Laso, J., Batlle – Bayer, L., Bala, A., Fullana -i- Palmer, P., Vazquez-Rowe, I., Gonzalez, M.J., Amo-Setien, F., Dura, M.J., Sarabia, C., Abajas, R., Quinones, A., Irabien, A., Aldaco, R., 2019. Nutritional data management of food losses and waste under a life cycle approach: case study of the Spanish agri-food system. Journal of food composition and analysis, 82, 103223. https://doi.org/10.1016/j.jfca.2019.05.006.

20. German, J.B., Zivkovic, A.M., Dallas, D.C:, Smilowitz, J.T., 2011. Nutrigenomics and personalized diets: What will they mean for food?. Annual review of food science and technology, 2, 97-123. https://doi.org/10.1146/annurev.food.102308.124147

21. Gibson, I., Rosen, D., Stucker, B., & Khorasani, M. (2021). Additive manufacturing technologies (Vol. 17). Springer.

22. Johnson, L.K., Bloom, D.J., Dunning, R.D., Gunter, C.C., Boyette, M.D., Creamer, N.G., 2019. Farmer harvest decisions and vegetable loss in primary production. Agricultural systems, 176, 102672. https://doi.org/10.1016/j.agsy.2019.102672.

23. Kim, H., W., Lee, I., J., Parl, S.,M., Lee, J.,H., Nguyen, M.,H., Park, H., J., 2019. Effect of hydrocolloid addition on dimensional stability in post-processing of 3d printable cookie

dough. LWT – Food science and technology, 101, 69 – 75. https://10.1016/j.lwt.2018.11.019.

24. Krkobabić, M., Medarević, D., Pešić, N., Vasiljević, D., Ivković, B., & Ibrić, S. (2020). Digital Light Processing (DLP) 3D Printing of Atomoxetine Hydrochloride Tablets Using Photoreactive Suspensions. *Pharmaceutics*, *12*(9), 833. https://doi.org/10.3390/pharmaceutics12090833.

25. Kummu, M., de Moel, H., Porkka, M., Siebert, S., Varis, O., & Ward, P. (2012). Lost food, wasted resources: Global food supply chain losses and their impacts on freshwater, cropland, and fertiliser use. Science of The Total Environment, 438, 477-489. https://doi.org/10.1016/j.scitotenv.2012.08.092

26. Lanaro, M., Forrestal, D.,P., Scheurer, S., Slinger, D.,J., Liao, S., Powell, S.,K., Woodruff, M.,A., 2017. 3D printing complex chocolate objects: platform design, optimization and evaluation. Journal of food engineering, 215, 13 – 22. https://10.1016/j.foodeng.2017.06.029.

27. Le Tohic, C., O'Sullivan, J.J., Drapala, K.P., Chartrin, V., Chan, T., Morrison, A.P., Kerry, J.P., Kelly, A.L., 2018. Effect of 3d printing on the structure and textural properties of processed cheese. Journal of food engineering, 220, 56 – 64. https://dx.doi.org/10.1016/j.jfoodeng.2017.02.003.

28. Lebersorger, S., Schneider, F., 2014. Food loss rates at the food retail, influencing factors and reasons as a basis for waste prevention measures. Waste management, 34, 1911 – 1919. https://dx.doi.org/10.1016/j.wasman.2014.06.013.

29. Lin, C. (2015). 3D Food Printing: A Taste of the Future. Journal of Food Science Education, 14(3), 86-87. https://doi.org/10.1111/1541-4329.12061

30. Lipton, I.J., 2017. Printable food: the technology and its application in human health. Current opinion in biotechnology, 44, 198 – 201. https://doi.org/10.1016/j.copbio.2016.11.015.

31. Liu, Y., Liang, X., Saeed, A., 2019. Properties of 3D printed dough and optimization of printing parameters. Innovative food science & emerging technologies. 54, 9 – 18. https://doi.org/10.1016/j.ifset.2019.03.008.

32. Love, D.C., Fry, J.P., Milli, M.C., Neff, R.A., 2015. Wasted seafood in the United States: quantifying loss from production to consumption and moving toward solutions. Global environmental change, 35, 116 – 124. https://doi.org/10.1016/j.gloenvcha.2015.08.013.

33. Martínez-González, M. A., Gea, A., & Ruiz-Canela, M. (2019). The Mediterranean Diet and Cardiovascular Health. Circulation Research, 124(5), 779-798. https://doi.org/10.1161/CIRCRESAHA.118.313348

34. Mistretta, E. G., Davis, M. C., Temkit, M., Lorenz, C., Darby, B., & Stonnington, C. M. (2018). Resilience training for work-related stress among health care workers: Results of a randomized clinical trial comparing in-person and smartphone-delivered interventions. Journal of Occupational and Environmental Medicine, 60(6), 559-568. https://doi.org/10.1097/JOM.0000000000001285

35. Nishimura, K. (2018). Using 3D Printers to Create Teaching Materials and Support Tools for Children With Physical Disabilities. The Japanese Journal of Special Education, 56(2), 105–112. https://doi.org/10.6033/tokkyou.56.105

36. Pant, A., Lee, A. Y., Karyappa, R., Lee, C. P., An, J., Hashimoto, M., Tan, U., Wong, G., Chua, C. K., & Zhang, Y. (2021). 3D food printing of fresh vegetables using food hydrocolloids for dysphagic patients. Food Hydrocolloids, 114, 106546. https://doi.org/10.1016/j.foodhyd.2020.106546

37. Passos, I. C., Ballester, P. L., Barros, R. C., Librenza-Garcia, D., Mwangi, B., Birmaher, B., Brietzke, E., Hajek, T., Lopez Jaramillo, C., Mansur, R. B., Alda, M., Haarman, B. C. M., Isometsa, E., Lam, R. W., McIntyre, R. S., Minuzzi, L., Kessing, L. V., Yatham, L. N., Duffy, A., & Kapczinski, F. (2019). Machine learning and big data analytics in bipolar disorder: A position paper from the International Society for Bipolar Disorders Big Data Task Force. Bipolar Disorders, 21(7), 582-594. https://doi.org/10.1111/bdi.12828

38. Philippidis, G., Sartori, M., Ferrari, E., M'Barek, R., 2019. Waste not, want not: a bio-economic impact assessment of household food waste reductions in the EU. Resources, conservation & recycling, 146, 514 – 522. https://doi.org/10.1016/j.resconrec.2019.04.016.

39. Redlingshöfer, B., Coudurier, B., Georget, M., 2017. Quantifying food loss during primary production and processing in France. Journal of cleaner production, 164, 703 – 714. https://dx.doi.org/10.1016/j.clepro.2017.06.173.

40. Revilla, B.P., Salet, W., 2018. The social meaning and function of household food rituals in preventing food waste. Journal of cleaner production, 198, 320 – 332. https://doi.org/10.1016/j.clepro.2018.06.038.

41. Ricci, I., Derossi, A., Severini, C., 2019. Chapter 5; 3d printed food from fruits and vegetables. Fundamentals of 3d food printing and applications, 117 – 149. https://doi.org/10.1016/B978-0-12-814564-7.00005-5.

42. S., H. (2021). Overview of 3D Printing Technology. Bioequivalence & Bioavailability International Journal, 5(1), 1–3. https://doi.org/10.23880/beba-16000149

43. Severini, C., Derossi, A., Azzollini, D., 2016. Variables affecting the printability of foods: preliminary tests on cereal-based products. Innovative food science and emerging technologies, 38, 281 – 291. https://dx.doi.org/10.1016/j.ifset.2016.10.001.

44. Severini, C., Derossi, A., Azzollini, D., Albenzio, M., 2018. On printability, quality and nutritional properties of 3d printed cereal based snacks enriched with edible insects. Food research international, 106, 666 – 676. https://doi.org/10.1016/j.foodres.2018.01.034.

45. Severini, C., Derossi, A., Fiore, A.,G., 2017. Ultrasound-assisted extraction to improve the recovery of phenols and antioxidants from spent espresso coffee ground: a study by response surface methodology and desirability approach. European food research and technology, 243, 5, 835 – 847. https://10.1007/s00217-016-2797-7.

46. Sofi, F., Abbate, R., Gensini, G. F., & Casini, A. (2010). Accruing evidence on benefits of adherence to the Mediterranean diet on health: An updated systematic review and

meta-analysis. American Journal of Clinical Nutrition, 92(5), 1189-96. https://doi.org/10.3945/ajcn.2010.29673

47. Sun, J., Peng, Z., Zhou, W., Fuh, J. Y., Hong, G. S., & Chiu, A. (2015). A review on 3D printing for customized food fabrication. Procedia Manufacturing, 1, 308-319. https://doi.org/10.1016/j.promfg.2015.09.057

48. van Dooren, C., Janmaat, O., Snoek, J., Schrijnen, M., 2019. Measuring food waste in Dutch households: A synthesis of three studies. Waste management, 94, 153 – 164. https://doi.org/10.1016/j.wasman.2019.05.025.

49. Vancauwenberghe, V., Katalagarianakis, L., Wang, Z., Meerts, M., Hertog, M., Verboven, P., Moldenaers, P., Hendrickx, M.E., Lammertyn, J., Nicloai, B., 2017. Pectin based food-ink formulations for 3-d printing of customizable porous food simulants. Innovative food science & emerging technologies, 42, 138 – 150. https://doi.org/10.1016/j.ifset.2017.06.011.

50. Voon, S. L., An, J., Wong, G., Zhang, Y., & Chua, C. K. (2019). 3D food printing: a categorised review of inks and their development. Virtual and Physical Prototyping, 14(3), 203-218. https://doi.org/10.1080/17452759.2019.1603508

51. Voon, S. L., An, J., Wong, G., Zhang, Y., & Chua, C. K. (2019). 3D food printing: A categorised review of inks and their development. Virtual and Physical Prototyping, 14(3), 203-218. https://doi.org/10.1080/17452759.2019.1603508

52. Yang, F., Zhang, M., Fang, Z., Liu, Y., 2018b. Impact of processing parameters and post-treatment on the shpe accuracy of 3D-printed baking dough. International journal of food science and technology, 54, 68 – 74. https://doi.org/10.1111/ijfs.13904.

53. Zhang, L., Lou, Y., Schutyser, M.A.I., 2018. 3d printing of cereal-based food structures containing probiotics. Food structure, 18, 14 – 22. https://doi.org/10.1016/j.foodstr.2018.10.002.

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Contributors	Statement of contribution
Derossi, Antonio	Conceptualization, Investigation, Formal analysis, Methodology, Data curation, Visualization, writing - original draft
Caporizzi, Rossella	Formal analysis, Methodology, Visualization, Writing – review and editing
Paolillo, Maddalena	Visualization, Writing – review and editing
Memet, Onur Oral (Candidate)	Visualization, Writing – review and editing
Severini, Carla	Supervision, Methodology, Visualization, Writing – reviewand editing

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Contributors	Statement of contribution
Derossi, Antonio	Conceptualization, Investigation, Formal analysis, Methodology, Data curation, Visualization, writing - original draft
Caporizzi, Rossella	Formal analysis, Methodology, Visualization, Writing – review and editing
Oral, Mehmet Onur (Candidate)	Methodology, Visualization, Writing – review and editing
Severini, Carla	Supervision, Methodology, Visualization, Writing – reviewand editing

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Contributors	Statement of contribution
Derossi, Antonio	Conceptualization, Investigation, Formal analysis, Methodology, Data curation, Visualization, writing - original draft
Caporizzi, Rossella	Formal analysis, Methodology, Visualization, Writing – review and editing
Oral, Mehmet Onur (Candidate)	Methodology, Visualization, Writing – review and editing
Severini, Carla	Supervision, Methodology, Visualization, Writing – reviewand editing

Derossi, A., Corradini, M., Caporizzi, R., Oral, M., & Severini, C. (2023). **Accelerating the process development of innovative food products by prototyping through 3D printing technology**. *Food Bioscience, 52, 102417.* [*https://doi.org/10.1016/j.fbio.2023.102417*](https://doi.org/10.1016/j.fbio.2023.102417).

Contributors	Statement of contribution
Derossi, Antonio	Conceptualization, Investigation, Formal analysis, Methodology, Data curation, Visualization, writing - original draft
Caporizzi, Rossella	Formal analysis, Methodology, Visualization, Writing – review and editing
Corradini, Maria G.	Methodology, Writing, Review & Editing
Oral, Mehmet Onur (Candidate)	Methodology, Formal analysis, Visualization, Writing – review and editing
Severini, Carla	Supervision, Methodology, Visualization, Writing – reviewand editing

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Contributors	Statement of contribution

Derossi, Antonio	Conceptualization, Investigation, Formal analysis, Methodology, Data curation, Visualization, writing - original draft
Oral, Mehmet Onur	Conceptualization, Formal analysis, Methodology, Visualization, Writing – review and editing
Gleadall, Andy	Visualization, Writing – review and editing
Jeckle, Mario	Visualization, Writing – review and editing
Caporizzi, Rossella	Conceptualization; Investigation; Writing – review & editing
Severini, Carla	Supervision, Methodology, Visualization, Writing – reviewand editing

Chapter 1 Drawing the scientific landscape of 3D Food Printing; Maps and interpretation of the global information in the first 13 years of detailed experiments, from 2007 to 2020.

A. Derossi, R., Caporizzi, M., Paolillo, M.O., Oral, C., Severini,

Department of Agriculture, Food, Natural resources and Engineering (DAFNE), University of Foggia, Italy

Corresponding author: rossella.caporizzi@unifg.it

Abstract

3D Food Printing is a hot field of research in which many efforts are concentrating to unleash its potential for renewal. To facilitate this process we have drawn the global scientific landscape in these first 13 years of experiments by using bibliometric and data visualization approaches. We find a total of 170 documents between 2007 and 2020. China and Australia are the most productive countries followed by Italy. On a total of 582 co-authors, not more than 10 researchers are collaborating out of their research group/institution. This is a weakness that urges sharing information and experiences. Also, the networks of the keywords have showed some hidden opportunities. To reinforce the interrelations of digital design, microstructure and personalized food is needed to create unprecedented sensory perceptions and to alleviate the mastication and swallowing problems of the vulnerable consumers. Also, there is a shortage of papers on the printing of protein-rich inks.

Industrial relevance: The paper critically analyzes the global production of scientific experiments in 3D food printing. The results discovered that the level of collaborative researches is weak while the involvement of the industrial sector urge as crucial process to unleash the great ambitions of personalized food manufacturing. What emerged is the need to study parallel deposition methodologies such as selective laser sintering (SLS) or hot-air sintering (HAS) that would open for fast printing and the use of highly stable food powders. Others opportunity to make feasible to application of 3DFP could be the use protein-rich food inks and to make closer the topic of digital design and personalized consumers requirements.

Keywords: 3D food printing; bibliometric analysis, clustering, collaborative networks; novel research opportunities.

1. Introduction

Additive Manufacturing (AM) is a broad range of technologies having the capability to transform 3D digital images in real objects through a layer-by-layer deposition process. In the last 20 years, AM technologies – popularly known as 3D printing (3DP) – have triggered a process of renewal of the manufacturing of items, as we traditionally know. The potentials of designing complex structures with customized end-user properties such as desired shape, dimension and physical properties are the main reasons of its notable success in several industrial sectors and the interest of many scientific fields. While the use of 3DP for rapid prototyping (Rayna and Striukova, 2016) has been the first application, its usage currently occurs in pharmaceutical (Manoj et al., 2020; Beg et al., 2020), regenerative medicine (Agostinacchio et al., 2020; Bueno et al., 2019), biomedical implants (Murr, 2020) bioengineering (Nesic et al., 2020), architecture (Chan et al., 2020), jewelry (Stamati et al., 2011), aerospace (NASA, 2015, Jiang et al., 2020), construction (Buchanan and Gardner, 2019) etc. Moreover, after the deposition of thermoplastic materials, the printing of metal (Gibson et al., 2018), clothing (Want, 2020), glass (Li et al., 2018), concretes (Comminal et al., 2020), etc., have become popular. The awesome interest in 3DP is proved by over than 28k of scientific publications listed on Scopus database since 1987 (data from Scopus database). While 3D printing of thermoplastic materials is advanced and realizes highly complex structures, the 3D printing of food is taking its first steps even though it is also becoming of great interest with unparalleled level of innovation capable of transforming the way in which foods will be manufactured, stored and consumed. Cohen et al (2009) stated that '*after solving the main issues of slow printing and price, the remaining question is what the ways in which 3DFP will completely modify the food sector, while no doubts on whether 3DFP may affect food manufacturing and consumption*'. As reported by several authors the main ambitions of 3D food printing (3DFP) are personalized food manufacturing (Cohen et al., 2009; Lipton et al., 2015; Severini and Derossi, 2016; Derossi et al., 2020a; Pulatsu and Lin, 2021), on-demand production, food waste reduction and consumers-food co-creation (Godoi et al., 2016; Jiang et al., 2019; Pulatsu et al., 2020). Personalized food manufacturing by using 3D Printing has the potential of creating food not only with desired shapes and dimensions but also to get nutritional and functional properties right for people uniqueness (Godoi et al., 2016; Le-Bail et al., 2020). So, the level of innovation may also be augmented by interrelating personal medical data, tele-medicine, planned diet, lifestyle, gender, sex, etc. (Tagami et al., 2021). In addition, 3DFP has the potentials of creating customized sensory properties by using multi-food materials (Park et al., 2020), by modeling mechanical properties (Derossi et al., 2020a; Derossi et al., 2020b) or creating food with the desired shape (Severini et al., 2018; Schutyser et al., 2018; Pulatsu et al., 2021a), color (He et al., 2020) and flavor (Guo et al., 2021). To date, researchers focused their work on the effects of printing variables (Derossi et al., 2018; Perez et al., 2019), the printability of food formulas (Liu et al., 2018; Tian et al., 2021) through the use of hydrocolloids and analyzing the corresponding rheological properties (Kuo et al., 2021; Pant et al., 2020; Gholamipour-Shirazi et al., 2019; Zhu et al., 2019), on programmable 3D structures aiming to get desired texture properties (Derossi et al., 2021) and, more recently, on the time evolution of color, aroma and shape as effect of an external factors as first examples of 4D food printing (Guo et al., 2021; Phuhongsun et al., 2020; He et al., 2020b). The bibliography of 3D food printing is getting large with an exponential trend of the papers published in the last years as well as large is the number of the reviews aiming to delineate the most important features of 3DFP.

Scientometrics is a wide field of research aiming to analyze the quantitative aspects of science and technology and its dynamic nature seen on the angle of the process of communication (El Mohadab et al., 2020). While the advent of digital information products has generated a massive amount of scientific documents published online, the researchers are slow in searching, collecting, studying and interpreting this large mass of data. This is called a typical problem of information overload (Shao et al., 2021). Bibliometrics is a specific field of scientometrics defined as '*the application of mathematics and statistical methods to books and other media of communication*' (Otlet, 1934). In practice, bibliometric is a tool for clustering and mapping the information from scientific documents such as keywords, abstract, authors, countries, references, journals, citation, etc., delineating the salient traits of a scientific domain and discovering novel features, network, potentials and weakness/strengthens of that field. As reported by Waltman et al (2010), bibliometrics drives people in understanding how research topics are, or could be, how they are related to each other, or to delineate how scientific topics are evolving over time, to define the impact of some journals, institutions, authors, countries, etc. on the research topic under evaluation. For instance, Mingers et al. (2015) stated that the act of citing papers creates a link between ideas, points of view, methods, people, journals, and institutions generating a scientific network that may be analyzed. Furthermore, bibliometric methods allow creating invisible links between highly cited papers to research frontiers (Price, 1976; Mingers et al., 2015) paving the way for innovative ideas.

In the field of 3D food printing, several reviews have been published (Manthial et al., 2020; Baiano, 2020; Dankar et al., 2018; Jiang et al., 2020; He et al., 2020b; Zhao et al., 2020; Handral et al., 2020; Feng et al., 2019) but a complete quantitative analysis of the global scientific production – original papers, review, letter to Editor, books, proceeding of international conferences – have not been performed. Although the usefulness of the reviews is globally recognized, the obtained benefits remain limited if we are not able to manage this large mass of publications that increases monthly. The reading of reviews, often focused on specific hot-points, limits to a high extent the exploration of the research status of 3D food printing with missed opportunities in individuating strengthens and weakness points and innovative bridges between hidden sub-topics. Our belief is that 3D Food Printing technology will definitely transform the future of food production and consumption, but its level of maturity is too low to collect high level of investments needed for a direct employment in the food chain. For this, the interpretations of all global scientific information aiming to highlight novel networks and interrelations could act as driver to activate a scientific discussion on the weakness and strengthens on 3DFP and fuel new cross-contamination between different topics.

This is an original paper that uses as input-data the main information and indicators of the global production of scientific documents on the topic of 3D Food Printing with the aim to draw the scientific landscape of 3DFP highlighting weakness and opportunities. With this aim we have combined bibliometric and statistical methods, mapping and clustering analysis, and visualization approaches to manage the large body of scientific documents in the field of 3D Food Printing.

2. Material and Methods

2.1. Collecting bibliographic information

We used SciVerse Scopus as the main database because it contains the largest number – 20,000 items - of indexed journals, books and conference proceedings (Mingers and Laydesdorff, 2015); also, although Scopus retrieves back till 1996, 3D Food Printing is a relatively young field of research and there are no reasons to search scientific documents before 1996. A schematic representation of the strategy used to collect the most relevant scientific documents belonging to the field of 3DFP, is reported in Figure 1. The search was conducted on 25 November 2020.

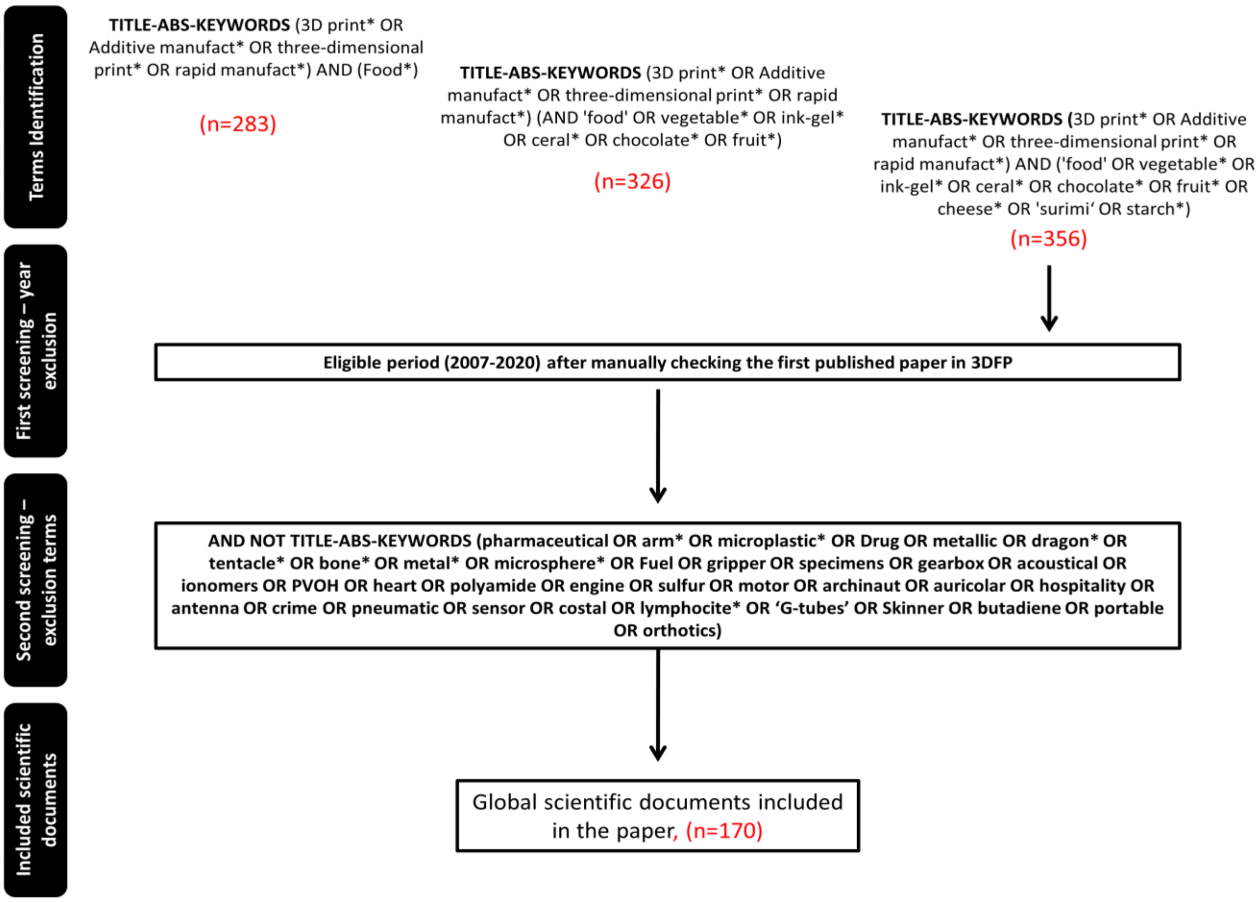


Fig. 1. Strategy employed to collect scientific documents in the field of 3D Food Printing.

First, different techniques and operators have been used to improve the accuracy of our search. For instance, asterisks have been used to collect related words as in the case of using *3D print** which enlarge the searching for combined words such as *3D printing*, *3D printed*. Also, the operators [AND], [OR] and [AND NOT] were used for including or excluding specific terms from the search. The type and number of terms used in the search query of Scopus was decided by preliminary tests aiming to collect the maximum number of papers belonging to the field of 3DFP.

First, using specific terms in a step-by-step approach of searching we retrieved a maximum of N=356 documents. Then, the obtained documents were limited first for the 2007-2021 and, second, a manual screening to avoid some 'false-positives' was performed. For instance, the

recent paper publication by Jereman et al. (2021) which contains in the abstract and as keywords the terms '*food*', '*additive manufacturing*' and '*food processing*' is focused on the field of abrasive waterjet technology with only negligible interrelation with the food sector.
Finally, a total of 170 documents have been retrieved and the following information was downloaded in format files .CSV: citation information; bibliographical information, abstract & keywords. Further properties of the documents such as the time distribution, subject area, countries and the types of documents have been obtained directly from the Scopus website. All the bibliometric information has been used in the following analyses: 1. Analysis of overall publication indicators; 2. Analysis of the co-authorship; 3. Analysis of the co-occurrence for the author's keywords.
While the analysis and visualization of some general indicators was performed by using the software Power BI (Microsoft) the other techniques were performed by using the software VosWiever, ver. 1.6.16 initially realized and released by van Eck and Waltman (2009) and widely used in many fields of research (Park and Nagy, 2018; Mascarenhas et al., 2018; Sweileh et al., 2018; Perianes-Rodriguez et al., 2016; Shao et al., 2021).

2.2.Co-authorship analysis

The analysis of co-authorship was performed by using as input data the authors and countries information of the retrieved papers with the aim to create networks of active collaborations in the field of 3D Printing. For the analysis, an entry-level of one document per country have been used while no entry-levels have been assigned for the number of citations received for each country. The fractional counting method was used by assigning to each author/country weight of 1/N, with N the number of total authors or countries involved in the paper (Perianes-Rodriguez et al., 2016).

2.3.Co-occurrence analysis of the author's keywords

The generation of the map for the author's keywords involved three different steps. A first analysis was carried out without any entry-level value for the number of occurrences. This approach allowed extracting a global list of 526 keywords and the corresponding list of the number of occurrences. This data have been used to define a relative weight for each keyword. We assumed a maximum weight of 1 for the keyword showing the highest occurrence which, in our case, was the term '*additive manufacturing*' with 66 occurrences. Then, the weight of the other keywords was defined by computing the fraction of the occurrences on the total of 66; relative weight = highest number of occurrence/number of occurrences of each keyword. Next, we decided to include in the network visualization only the keywords showing a weight > 0.05 corresponding to a minimum number of occurrences of 3. Finally, a thesaurus file was used for data cleaning consisting in the capability of merging similar words - i.e. '*food printing*', '*3D food printing*' and '*3D food manufacturing*' - or to delete potentially unrelated words. Figure 2 schematically describes the approach used to analyse the author's keywords.

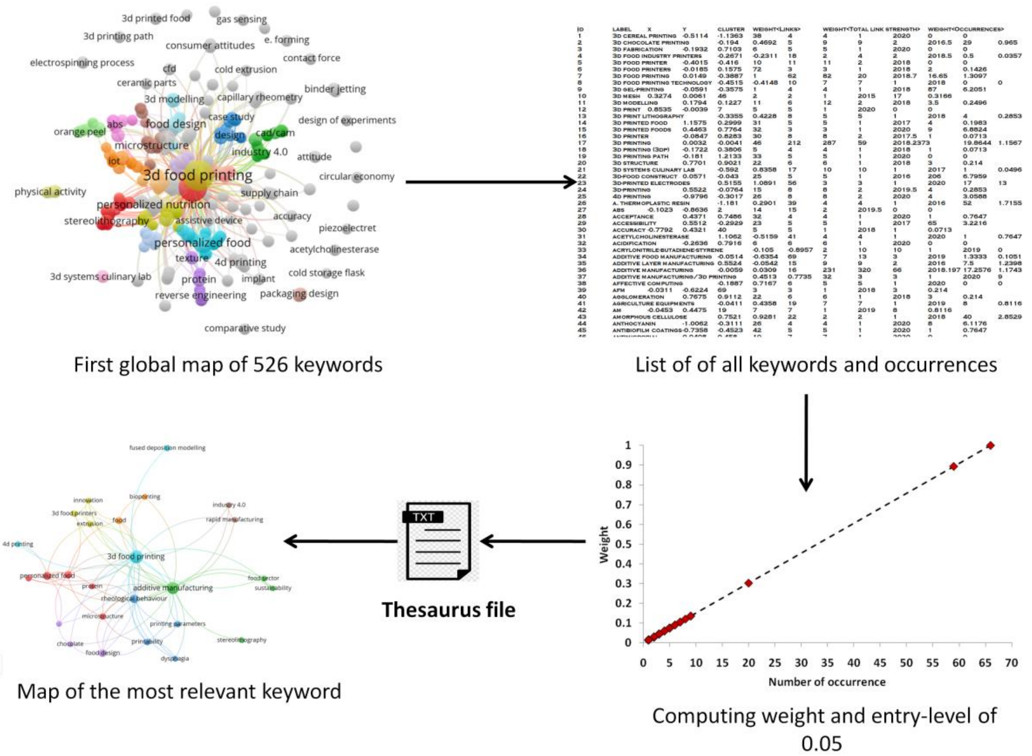


Fig. 2. Schematic representation of the method used for the analysis of the author's key-words

3. Result and Discussion

3.1.Distribution of publications over time, space and subject area

The evolution over time of scientific documents clearly shows a rapid increase in the interest of the scientific community in 3D Food Printing. While fewer than 9 papers were published between 2007 and2015, an average of 29 papers/per year was obtained from 2016 to 2020 with a peak of 48 documents in 2018 (Figure 3a). At the moment of writing this, paper only 6 papers result in 2021 that is lower than what is possible to observe in the general field of 3D printing. This suggests a shortage of new data on 3D food printing but during 2020, COVID-19 has been a calamity for scientific society with the closure of universities, research centers, laboratories and the cancellation of many scientific congresses and workshop (Subramanya et al., 2020). Also, COVID-19 has collected extraordinary efforts from all scientific sectors limiting the development of other themes of research. For instance, Kambhampati et al. (2020) retrieved 1638 publications on COVID-19 in the first 17 weeks of 2020.

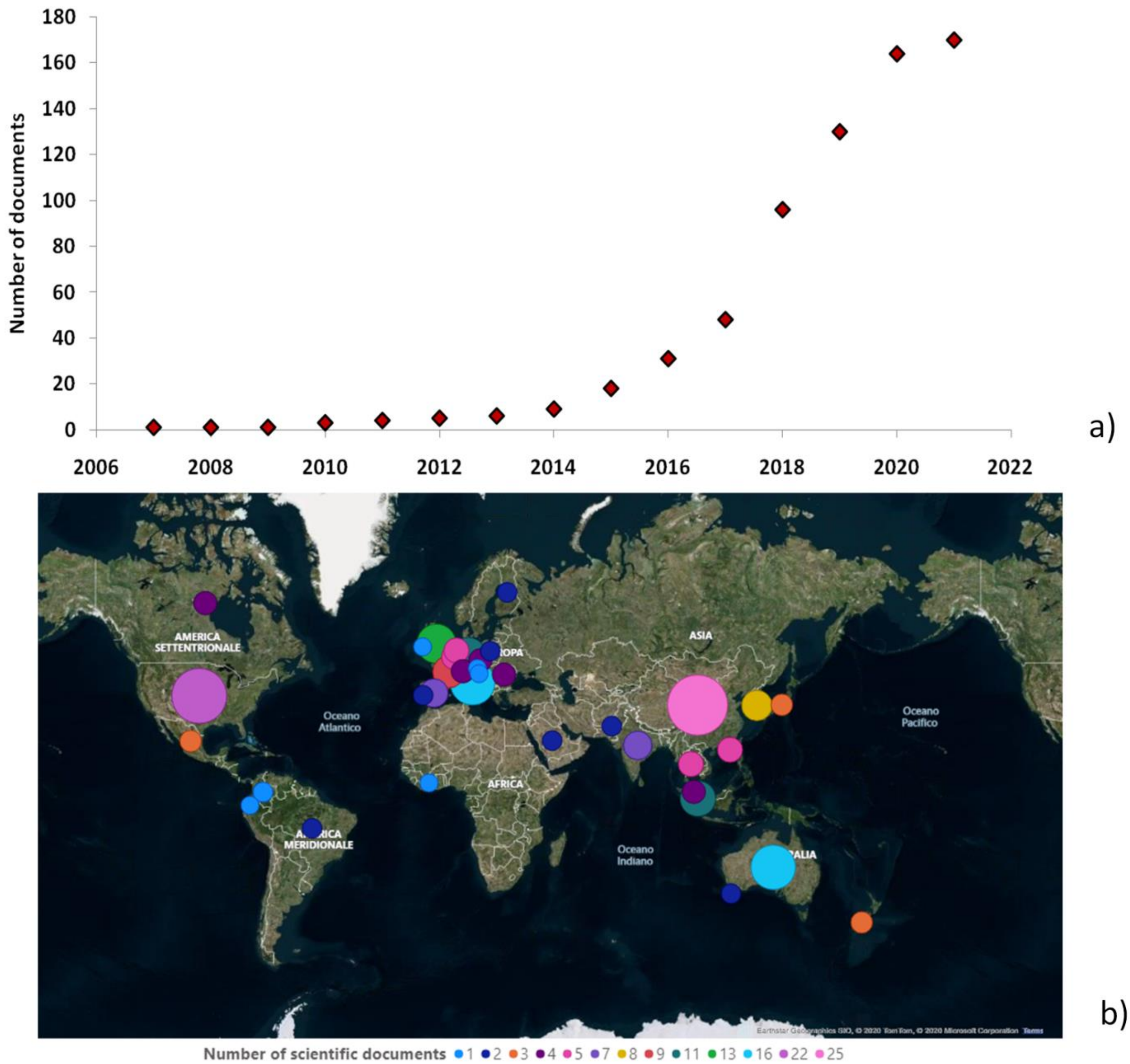


Fig. 3. Evolution over time (a) and geographical distribution (b) of the global publications on the emerging topic of 3D Food Printing.

Additional information may be obtained from the geographical distribution of the published documents (Figure 3b). China, USA, Australia and Italy are listed as the first productive countries in the field of 3D food printing with 25, 22, 16 and 16 scientific documents, respectively. However, when analyzing in detail the type of journals, significant differences have emerged with USA that has disseminated data mainly in the scientific areas of intelligent systems, computer application, medicine and 3D digital design and fabrication while only 2 papers on a total of 25 were published in food science and technology (Jiang et al., 2019; Lipton et al., 2015). On the other hand, more than 80% of the papers coming from China, Australia and Italy have been published in the food science and technology area (data not shown). Table 1 delineates the main features of the global scientific production on 3DFP. As expected for a young research topic, the majority of the documents are classified as original articles (N=90; 53%) and conference papers (N=38; 22.3%) but it is worth noting an interesting number of reviews (N=20; 11.7%). Of these reviews, some are generally focused on the main potentials and ambitions of 3DFP (Manthial et al., 2020; Le-Bail et al., 2020; Nachal et al.,

2019) while others have analyzed specific topics such as the development of food-inks (Voon et al., 2019; Feng et al., 2019; Gholamipour-Shirazi et al., 2020; Jiang et al, 2019), 3D printing of meat (Dick et al., 2019), regulatory and economic issues (Baiano, 2020) and materials and machines used for food printing (Tan et al., 2018). The analysis of the subject category shows as the majority of the papers have been published under '*engineering*' and '*agricultural and biological science*' areas, but another important category is '*computer science'*. This is in accordance with the nature of 3DFP that allows creating foods from digitalized images obtained by computer aided design (CAD). The multi-disciplinary features of 3DFP allow it to grow in closer fields of research such as Human-Computer-Interaction (HCI) which recently begun to pay interest in the food sector with the birth of the Human-Food-Computer-Interaction (HFCI) area that promises to reshape how food is produced, transported, prepared and consumed. Relevant examples have been published by Chaudry et al. (2012); Hashimoto et al., (2012); Comber et al., (2014) Betran et al. (2019). Tools to support the nutritional assessment such as the size portions for diet intake have been published by Chaundry et al., (2012) while Hashimoto et al., (2012) studied smart kitchen capable to help people of preparing unfamiliar food recipe – or nutritionally personalized food formula – by using a complex system providing recipe information by using visual and sound data to recognize ingredients.

Type of documents		**Subject category**		**References for Subject Category**
Article	91	Engineering	87	Fahmy et al. (2020)
Conference Paper	38	Agricultural and biological science	71	Portanguen et al., (2019)
Review	20	Computer science	40	Ramundo et al., (2016
Book Chapter	16	Materials science	27	D'angelo et al., (2016
Conferece Review	2	Chemistry	26	Gholamipour-Shirazi et al., (2019)
Book	1	Chemical engineering	19	Dianez et al., (2019)
Short Survey	1	Business, management and accounting	13	Charlebois and Juhasz (2018)
Note	1	Decision sciences	11	Steenhuis et al., (2018)
Editorial	0	Biochemistry, genetics and molecular biology	9	Sun et al., (2015)
Erratum	0	Mathematics	9	Kim et al., (2018)
Letter	0	-		

Table 1 – Distribution of 3D Food Printing papers for type of document and subject category.

The scientific productivity of the top 10 institutions is shown in Figure 4. A total of 56 documents, 33% of the global production, were published. Of these, the most productive institutions have been the University of Queensland, Jiangnan University and the University of Foggia, respectively with 9, 8 and 8 publications. The top ten institutions produced their papers from 2016 to 2020 suggesting they strongly believe in the high potential of 3D Food Printing deciding to invest significant research efforts in a limited period of time. In addition, they published a total of 32 different journals while the total number of journals publishing 3DFP experiments has been 116 with high diversity in their main scopes. This proves that editors now consider 3DFP an important topic in food engineering, food polymers application, computer in agriculture, computers in industry, etc.

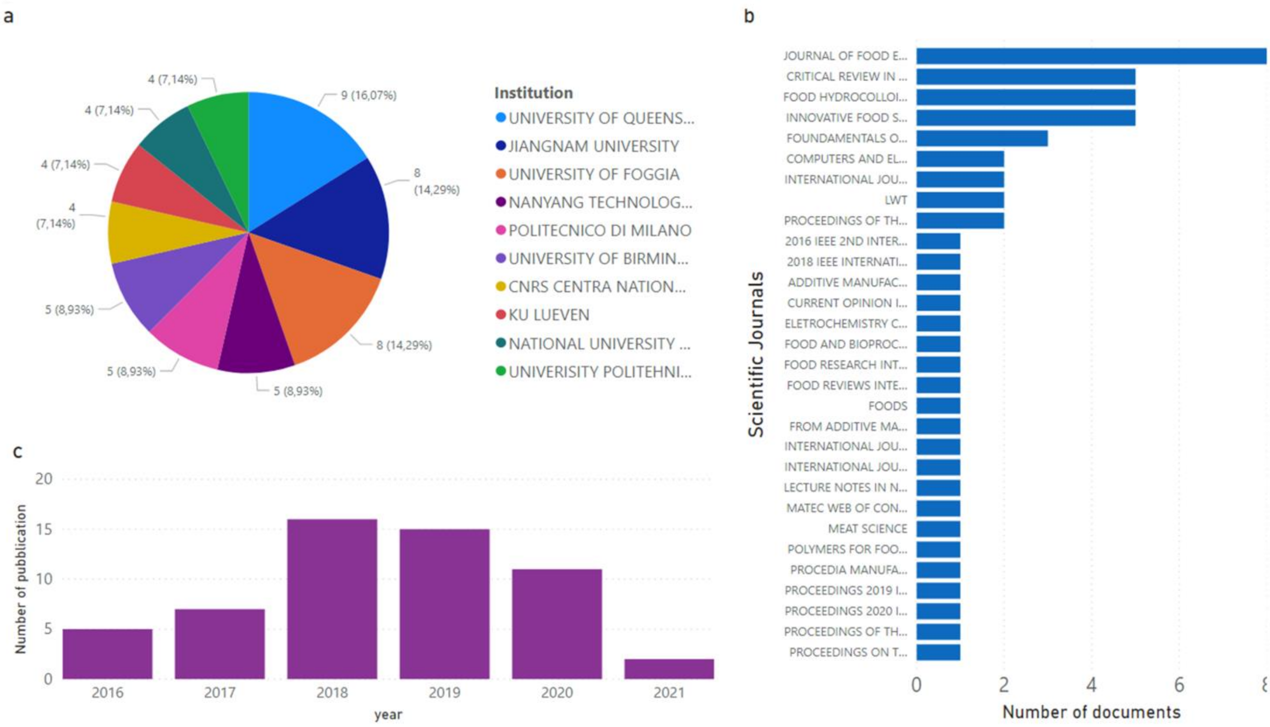


Fig. 4. Results of the first 10 istitutions publishing on 3D Food Printing. A) fraction of the number of pubblications. B) distribution over time; c) publications by source.

3.2.Analysis of co-authorship

With the aim to examine the active collaborations among countries, the network visualization of the active countries in 3DFP gives great opportunities for immediate responses and novel discussion. In the map, the size and color of data respectively represent the number of occurrences and the cluster type. Two countries co-occur if they co-authored a publication on 3DFP. Moreover, a link between two countries is created when they co-occur in the same documents and the greater the strength link is, the higher the number of the papers co-authored by the countries is or, in other words, they show a strong collaboration. The global network of the co-occurrence is featured by 38 countries indicating that 3D food printing is broadly considered an interesting and promising field of research all around the world; however, 15 countries look like 'isolated island' without active international collaborations (Figure 5A). The other 27 countries create the largest cluster with a total of 41 links (Figure 5B) of which China and Australia count the higher number of links toward other countries and respectively of 9 and 4. Also, China and Australia formed the highest links strength - respectively, 18 and 10 on a total of 55 - proving they have very productive international collaborations with many published papers in the field of 3D food printing. More specifically, with link strength of 7, China and Australia are the countries with the most prolific collaborative endeavors (Yang et al., 2018; Wang et al., 2018; Feng et al., 2019; Guo et al., 2019). In addition, the map shows that China and Australia might be considered the bridge between USA, Europe and Asia enlarging the global network of collaboration and making visible to the researchers the potential countries to invite for novel collaborations. Italy, for instance, that is one of the most productive countries (Figure 5b), shows only 2 links with Switzerland and Brazil while its involvement in other international collaborations could help to improve the quality of the 3D food printing technologies. It is important to note that the label of some countries is not clearly visualized in the map due to the default scale used by the software VosViewer but they actively participate in the network of active countries in the field of 3D Food Printing. Some of these are for instance Germany (Kern et al., 2018), Portugal (Sartal et al., 2019) and India (Piyush et al., 2019).

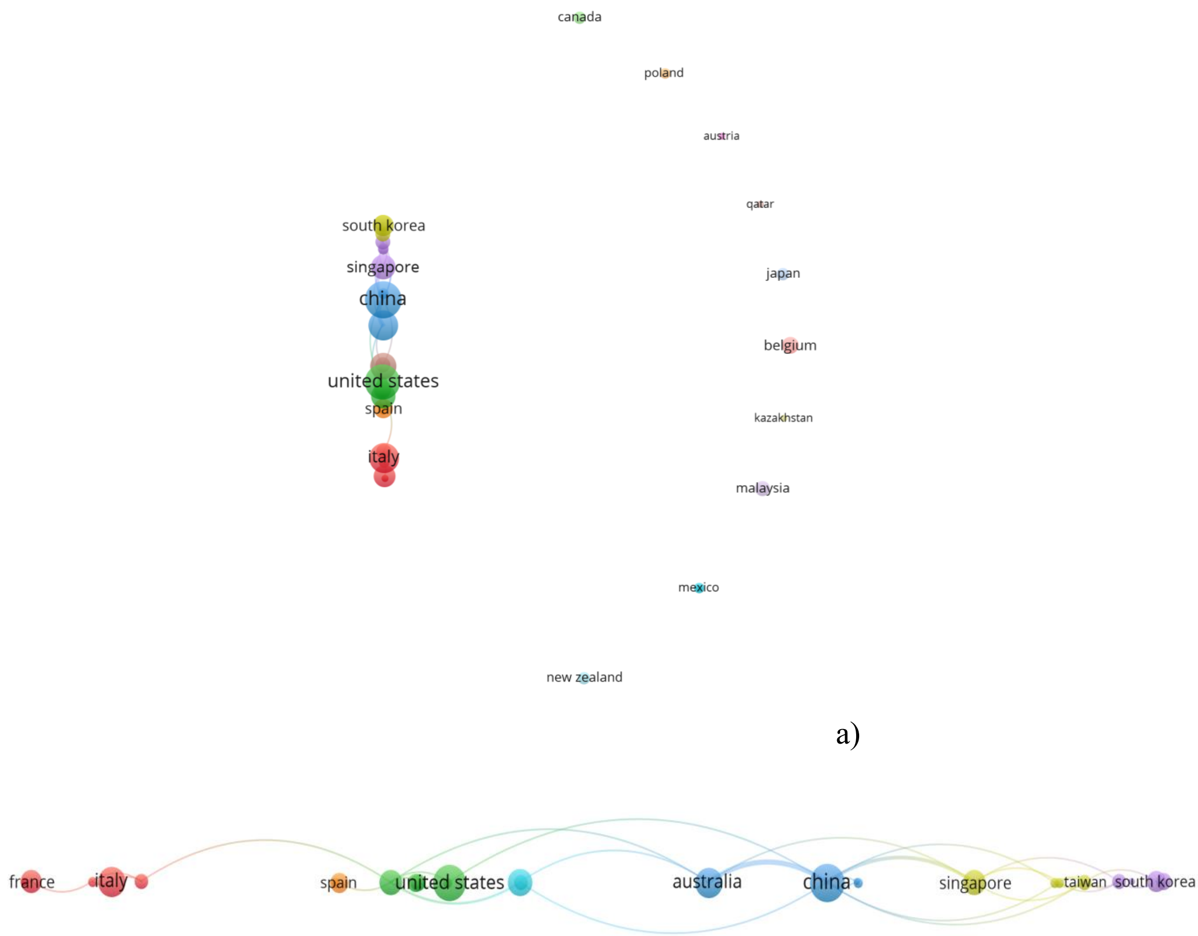


Fig. 5. Map of the active countries in the field of 3D food printing. A) Global map of countries contributing in the field of 3D food printing; b) Largest cluster of interconnected countries.

When analyzing the network of the authors a total of 582 names have been visualized. The map of the co-authored papers indicated a large number of colored clusters suggesting that all researchers are working in small groups (data not shown). More specifically, the most productive clusters appeared under the main label of the following authors: ‘Bhandari, B.’, ‘Lipson, H.’, ‘Lammertyn, J.’, ‘Hao, L.’, ‘Lanaro, M.’, and ‘Liu, Y’. Also, only the clusters of ‘Bhandari’, ‘Hao’ and ‘Liu’ are linked stating that they are sharing results, data, and efforts in 3D food printing activities. Furthermore, the visualization of many islands of authors proves that the majority of the researchers are collaborating mainly in their own research groups or institutions.

The largest set of connected authors, as shown in Figures 6a and 6b, is characterized by 51 authors, 6 clusters, 194 links and a total strength link of 208. The maps also report color scales indicating the average year of publication (figure 6a) and the average number of citations received by each author (figure 6b). With the aim to avoid multiple and repetitive figures we have used colored dot circles to highlight the clusters including authors who have strictly collaborated in co-authoring papers. On the left hand is located the largest cluster with 15 linked authors (red) who singularly show at least 14 links and publishing papers in the last years 2019-2020. Of these authors, the larger fraction showed a limited average citation value of 2 while only Liu, Y., who clearly has collaborated outside his ‘birth cluster’, received an excellent number of average citations value of 52.5 with only two papers (a total of 105 citations). Also, this cluster relates to the grey cluster by three links between Liu, Y. and Zhang, M., Yang, F., and Bhandari B. On the opposite side is located the green cluster including 9 authors who published between 2014 and 2018 obtaining average citations values between 1 and 23. Another important cluster is highlighted in grey color, located in the middle of the map, and containing the most collaborative researchers being they linked out with others 4 clusters. Also, to this cluster belong the most productive and cited authors such as Zhang, M., who published 8 documents and showed an average citation of 40.12 while Bhandari, B., signed a total of 9 papers and received an average citation of 66.78. In addition, these two authors have created the strongest collaboration with a strength link of 7 between the years 2018 and 2019. So, they are tightly collaborating resulting very productive with 7 co-authored documents in two years. Similarly, to the grey cluster, the light blue cluster is located in the middle of the map representing conjunction with other clusters of authors. The light blue is featured by the highest diversities with authors who are collaborating in a wide period of time – from 2016 to 2020 – and receiving very different average citations with values between 0 (Kobun R. and Lee, B.B.) and 132.50 of Godoi, F.C. that has been the first editor of the only book on 3D Food Printing titled ‘*Fundamentals of 3D Food Printing and Applications*’ (Godoi et al., 2018). So, Godoi, F.C. shows the highest average citations resulting from a total of 265 citations and 2 published papers. Finally, when analyzing the total number of citations, the highest cited authors follow the next order: Bhandari, B. (601), Prakash, S. (326), Zhang, M., (321), Godoi, F.C. (265), Yang, F. (163) and Manthial, S. (97).

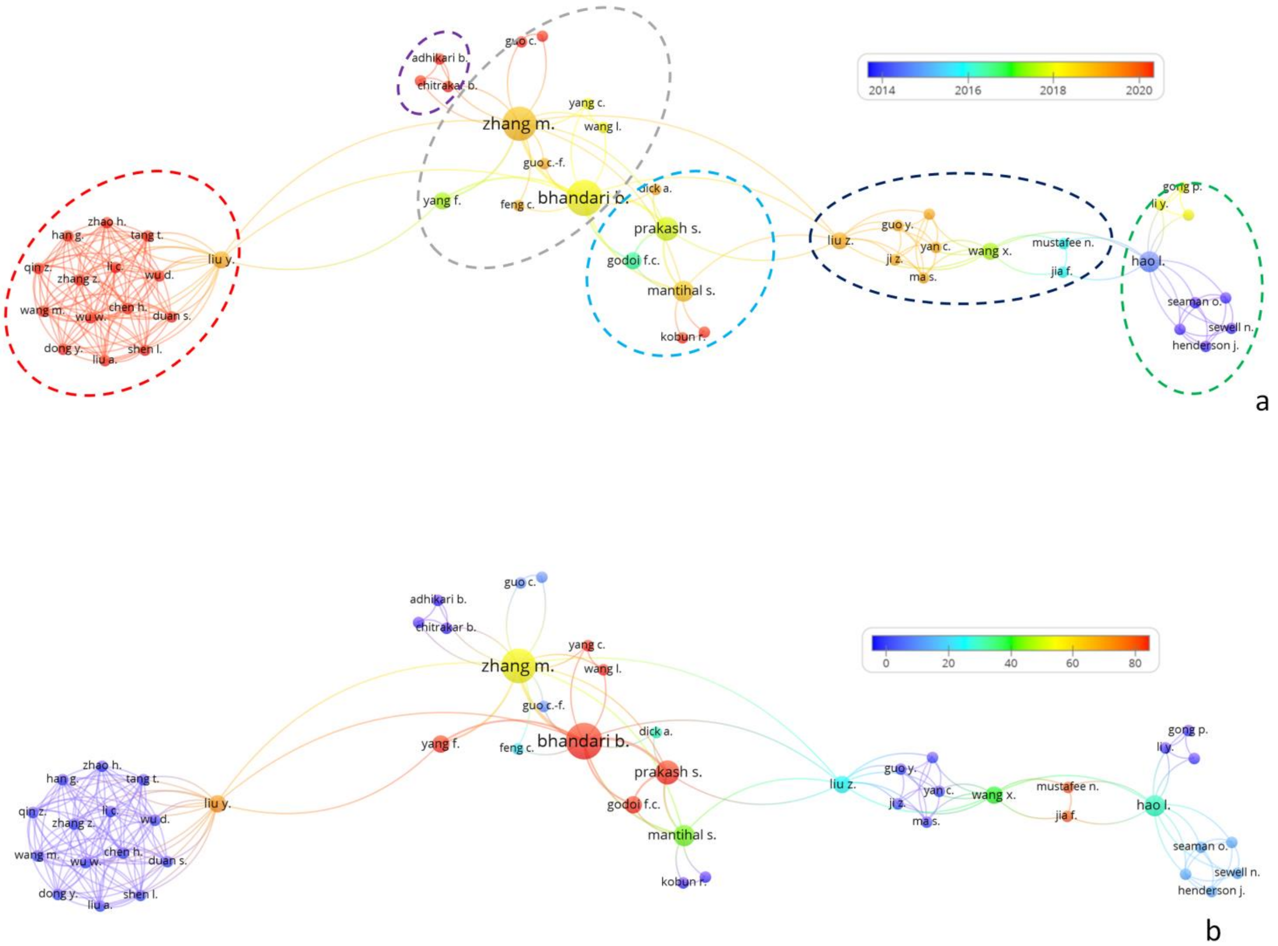


Fig. 6. Map of the largest set of interconnected authors. Color map indicates the year distribution (a) and the average number of received citation (b).

3.3.Analysis of co-occurrence

Being the keywords the backbone of a scientific paper, used to resume the main issues and ambitions of the research, we have performed the analysis of the co-occurrence by using as input data the author's keywords. After the preliminary analysis – without entry-level for the number of occurrences – a total of 491 keywords have been retrieved and mapped (data not shown). Then, according with the method described in the previous section of the paper, we have limited the number of keywords building the map of the most representative terms by using an entry-level of n.3 occurrences corresponding to the relative weight of 0.05. The most characterizing keywords for the field of 3D food printing – a total of 28 terms - are graphically illustrated in Figure 7 with the corresponding number of occurrences. Some keywords appear to be interchangeable such as 'food printing', '3D food printing', '3D printing' and all of them were replaced with the term '3D Food printing' by the cleaning approach of Figure 2. However, before discussing the main results, we want to note the repeated usage of keywords such as 'personalized nutrition', 'customized food', 'customized food fabrication' which prove as the 3DFP is considered one of the most promising technologies to make real the fabrication of nutritionally and sensorial personalized food products.

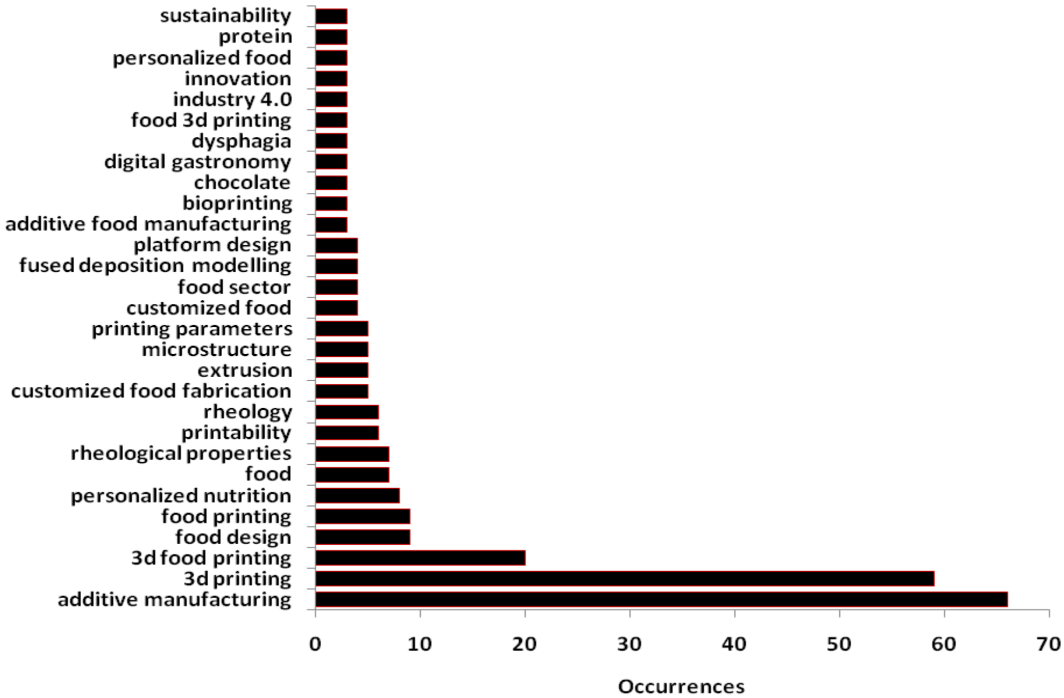


Fig. 7. The first 28 keywords with co-occurrences greater than of 3.

The maps of the most representative keywords in the field of 3DFP are reported in figure 8 where the number of occurrences is represented by the size of the data point and the color scale indicates the average year of publication (figure 8a) and the normalized average number of citation (Figure 8b). Only a period of time between 2016 and 2020 is reported because the representative keywords fall later than 2016 with the only except of '*platform design*' with the average year of 2015. Again, the thickness and the length of the links between two keywords indicate the coupling strength. So, the thicker and shorter is the link, the closer the linkage between two keywords. The specific keywords '*rheological properties*', '*protein*', '*extrusion*' and '*fused deposition modeling*' indicate recent hot-points in 3D food printing area with an average years between 2019 and 2020. This is consistent with the increasing efforts of many research group for the creation of food pastes exhibiting shear thinning behavior that makes easy the extrusion through a narrow nozzle, and other additional properties such as adhesiveness, viscosity and consistency which provide the capability to not collapse under the weight of the overlying layers enabling a good fidelity of printing (Gholamipour-Shirazi et al., 2019; Zhu et al., 2019). Also, the development of methods to estimate rheological properties with the aim to get a high printability is one of the most challenging points for the practical applications of 3D food printing at the industrial level or for home use (Liu et al., 2019; Zhu et al., 2019). The importance of this hot point is also proved by the high number of occurrences of the keywords '*rheological properties*', N=13, placed as third in the list of the keyword's occurrence after general terms of '*additive manufacturing*', N=91, and '*3D food printing*', N=32 (Table 2). This amplifies the importance of defining the essential rheological information and their ranges by which it is possible to expect a good printability; moreover, not less important will be to discover methods to design and create ink-foods that match the proper rheology.

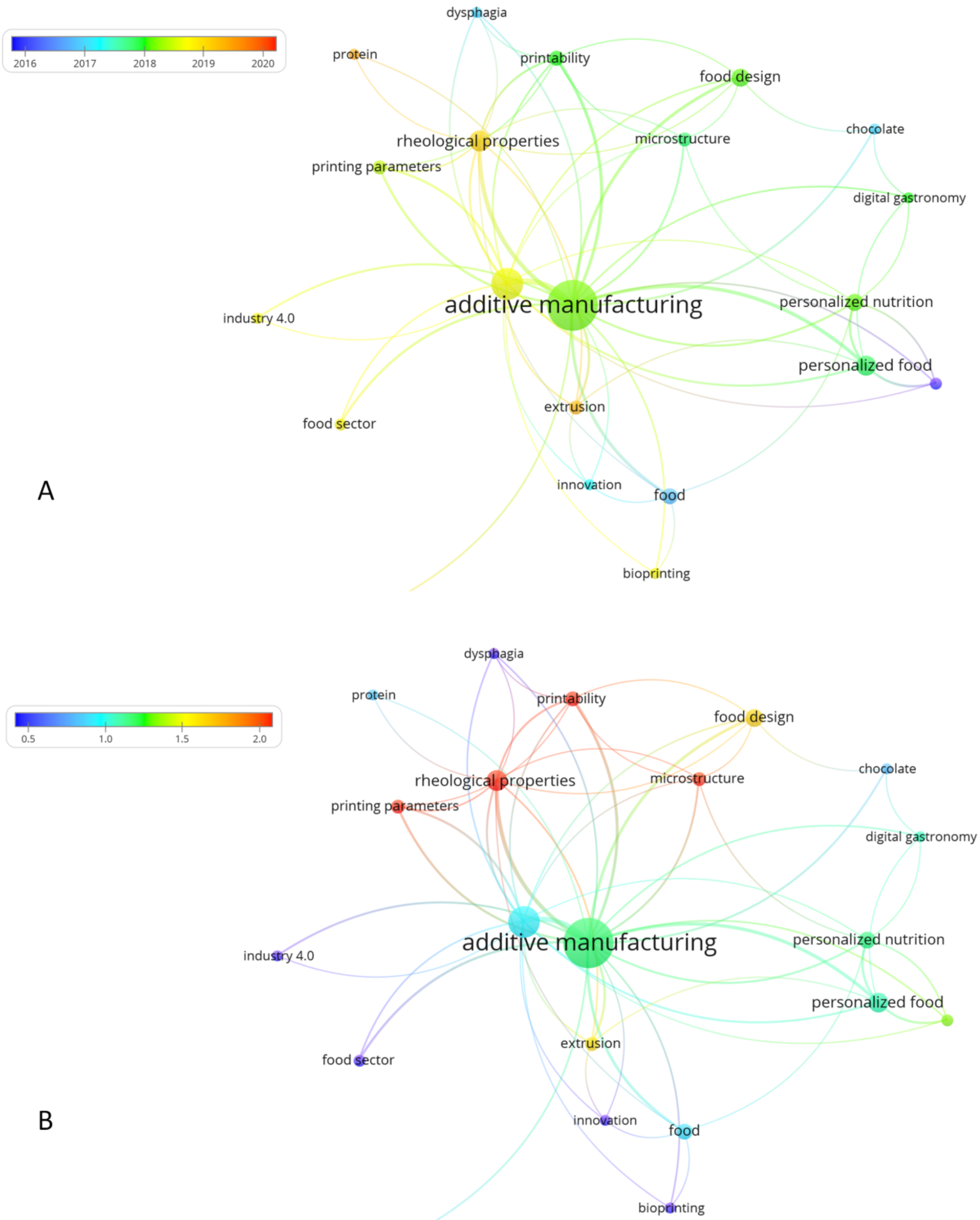


Fig. 8. Network visualization of the author's keywords used in the field of 3D food printing. A) color scale indicates the average year of publication; B) color scale indicate the average normalized citations.

The time evolution of 3D food printing reveals as the keyword's '*dysphagia*', '*platform design*', '*chocolate*' show an average year of publication of 2017. Mainly, the first experiments on 3D food printing have been performed by testing the chocolate due to its similarities with the thermoplastic materials used in fused deposition modeling (Jia et al., 2016; Mantihal et al., 2017). Also, the keyword '*dysphagia*' that seems no longer used in the recent year it is rather included in the most general theme of the creation of personalized food structure capable of mitigating mastication or swallowing problem of elderly people (Derossi et al., 2020a; Derossi et al., 2020b). Finally, we can observe the keywords '*printability*', '*microstructure*', '*food design*', '*personalized food*' and '*personalized nutrition*' that fall approximately in 2018.

Table 2 – Main results of the co-occurrences analysis performed using author's keyword as input data.

Author's keywords	Links	Total Link Strength	Occurrences	Avg. Pub. Year	Avg. Citations	Avg. Norm. Citations
3d food printing	15	44	32	2018	12.31	0.95
additive manufacturing	20	91	91	2018	18.60	1.16
bioprinting	3	4	3	2018	3.00	0.40
chocolate	3	5	3	2017	16.33	0.81
digital gastronomy	4	5	3	2018	18.00	1.08
dysphagia	4	6	3	2017	7.00	0.48
extrusion	5	10	5	2019	13.20	1.57
food	5	10	7	2016	12.85	0.89
food design	6	12	9	2018	31.66	1.62
food sector	2	6	4	2018	5.50	0.19
fused deposition modelling	1	2	3	2019	9.66	0.88
industry 4.0	2	4	3	2018	5.33	0.41
innovation	4	4	3	2017	4.00	0.15
microstructure	6	8	5	2017	33.20	1.96
personalized food	6	21	12	2017	32.50	1.09
personalized nutrition	7	13	8	2018	29.50	1.14
platform design	4	9	4	2015	78.00	1.32
printability	7	15	6	2018	45.00	2.07
printing parameters	4	11	5	2018	46.40	3.46
protein	2	2	3	2019	10.00	0.84
rheological properties	10	26	13	2019	28.92	2.50

With the aim to dig out the most important points of 3D food printing, it is useful to analyze the normalized average citations of the author's keywords as reported in Figure 8B. Also, other indexes of the co-occurrence's analysis are listed in Table 2. The keywords '*printing parameters*', '*rheological properties*', '*printability*' and '*microstructure*' exhibited the highest average normalized citations with values of 3.46, 2.51, 2.07 and 1.97 respectively. So, the most read and cited papers in the field of 3D food printing share information about the optimization of printing parameters and the printability of food paste which are the biggest obstacles to overcome for the practical application of 3DFP at a large scale. In addition, the study of the '*microstructure*' of 3D printed samples is gaining great importance because it gives deep information on the structural weakness of the printed samples (Severini et al., 2018a; Derossi et al., 2020) allowing the improvement of 3D printed food stability. Next, the keywords '*food design*' and '*extrusion*' reached an average normalized citation respectively of 1.62 and 1.57. This allows making two main considerations: 1. The use of extrusion units to deposit food filaments through syringe-based or screw-based systems is the most used method in 3D food printing; 2. The food design is and must be considered in a broad sense not only indicating the potential of forming complex food shapes but the workflow of making tangible a unique project thought on the specific nutritional and sensorial needs of the consumers.

Contrarily, these data highlight a weak point of the current research in 3D food printing: the lack of experiments and information on different deposition methods such as selective laser sintering (SLS) or hot-air sintering (HAS), binder jetting, inkjet printing (Sun et al., 2015). For instance, SLS or HAS that uses powder material and laser or hot air to fuse particles in a layer-by-layer building process, has the great advantages to being very fast allowing to create a massive amount of products. But these techniques are currently undervalued and only tested on sugars or sugar-rich formula (Mantihal et al., 2020; Gray, 2010). Furthermore, being the powders highly stable and because the un-sintered powder may be easily re-used, SLS or HAS would be a very efficient technique to study in detail.

Finally, we want to analyze the keywords '*chocolate*' that by three occurrences and an average normalized citation of 0.81 apparently is a field in which the interest of researcher is decreasing although some papers have been recently published mainly by Mantihal and co-authors (Mantihal et al., 2019a; Mantihal et al., 2017; Karyappa and Hashimoto, 2019; Mantihal et al., 2020; Rando et al., 2021). The reasons why the keywords appear with a weak importance in the map of Figure 9 is that the authors decided to not include the terms 'chocolate' in the choice of the keywords, but they classified the papers in a more general manner by using keywords such as '*3D food printing*', '*printable material*', '*texture modification*', '*infill percentage*', '*sensory*' etc. (Mantihal et al., 2019a; Manihal et al., 2019b; Mantihal et al., 2020). This would suggest to all authors a surgical choice and use of the keywords avoiding general terms – often already included in the title - that limits the capability to make visible and to share their results and, in turn, to improve the level of understanding of 3D food printing technology.

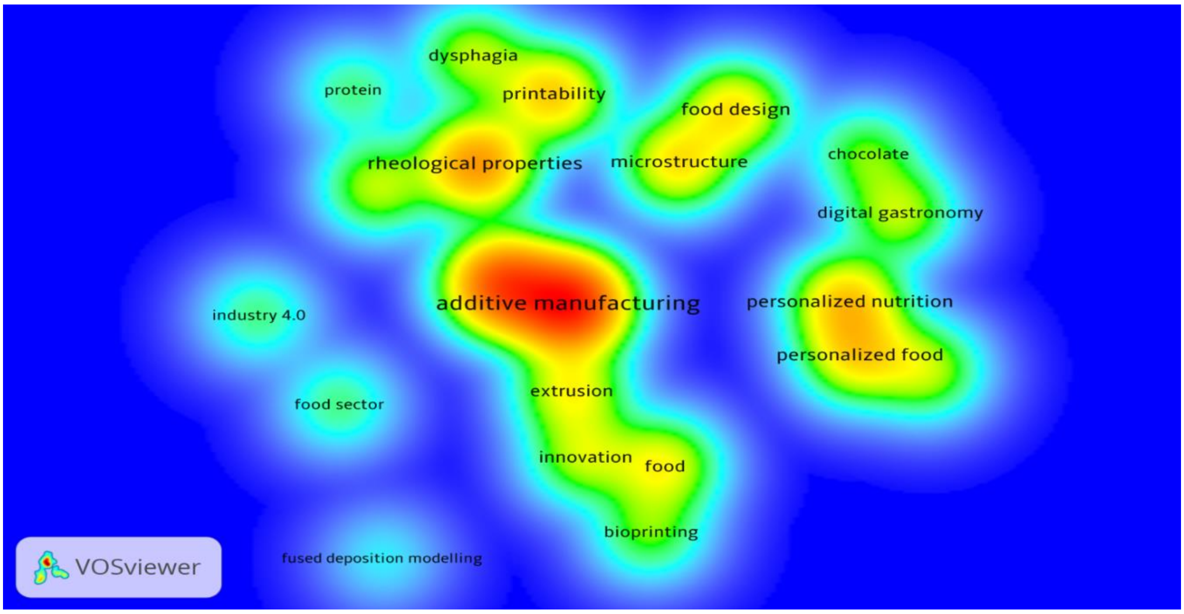


Fig. 9. Visualization of the density of link of the map of the author's keywords used in 3FDP literature.

To conclude, we want to analyze the interrelations of 3D food printing topics. The density visualization of Figure 9 displays for any point of the map the density of the links at that point. In other words, we can assume the figure as a geographical map with islands in the sea of 3D food printing. While the contours of the island are shaped by the interrelation of the keywords that co-occur in the same papers, the altitude of the island represents the number of links of each keyword. The density visualization has been obtained by setting the parameter Kernel width of the software VosViewer at 1.22. Four main islands can be observed.

The largest is shaped by the keywords '*additive manufacturing*', '*3D food printing*', '*extrusion*', '*innovation*', '*food*' and '*bioprinting*' and we want to label it with the name of *Innovative 3D Food Printing by Extrusion*. The second island of research may be labeled as *Rheology Control and Printing Fidelity Optimization* being featured by topics such as 'printability', 'rheological properties', 'printing parameters and 'dysphagia'. These two first islands are physically interconnected but mainly through the topic of rheological properties of food ink which of course is central for the innovation in 3D food printing. Another interesting result is given from the single island *Protein* that is located very close to the previous island, but it shows only two links (Table 1) with the keywords '*rheological properties*' and '*additive manufacturing*'. Here we can figure out a weak point of current research or – contrarily – a good opportunity to study in detail the use of food formula rich in proteins for 3D food printing experiments aiming to get novel personalized food and improving the knowledge about effects of printing parameters, microstructure, dysphagia. The use of novel sources of proteins also coming from the recovery of other food manufacturing processes would be of great relevance for many current challenges of food security and nutrition (FAO, 2020). However, apart from some previous studies on the use of cereal-based food formula (Zhang et al., 2018; Derossi et al., 2020a; Derossi et al., 2020b; Derossi et al., 2021) and insect powder (Severini et al., 2018a), the body of the knowledge of 3D printed food rich in protein is fragile (Uribe-Wandurraga et al., 2020). Others islands in the left part of the map that are worth mentioning can be labeled as *Design at Macro- and Microstructure* and *Personalized Food*. These are themes of crucial importance for the future development of 3DFP but they appear only weakly linked with the others research topics allowing to highlight additional opportunities such as to study the creation of food with personalized mechanical properties that based on the deep information on digital design, microstructure properties and rheological feature of food-ink may unleash novel potentials of 3DFP by creating unprecedented texture perceptions and helping to mitigate the mastication and swallowing problems of elderly or patients.

4. Conclusions

3D Food Printing, 3DFP, is an emerging field of research with unprecedented ambitions of creating foods with sensorial and nutritional personalized properties from digital images. This paper creates a physical map of the global production of scientific documents aiming to contribute to the future development of 3DFP highlighting weaknesses and discovering hidden latent opportunities.

We find a total of 170 documents from 38 Countries of which China and Australia are the most productive and collaborative. In addition, only 51 authors on a total of 582 researchers shape the largest map of the active collaboration although more limited – not more than 10 - is the number of authors directly connected out of their own group/institution. This is a weakness in the actual field of 3DFP and novel international collaborations should be activated to speed up the real implementation of 3D printing in the food sector. When mapping the most representative author's keywords fall in the period between 2016 and 2020. Current hot points, in which the majority of the efforts are concentrated, are the '*rheological properties*', '*printability*', '*printing parameters*' and '*microstructure*' which show the highest average normalized citations. In fact, the hottest and urgent topic regards the capability to design and develop ink-food with excellent rheological properties and the ability to customize the printing parameters on the features of the ink-food. Also, the analysis of the co-occurrences revealed the shortage of experiments on alternative printing methods such as SLS and HAS which could introduce two main novelties: fast printing and the use of dehydrated food-ink with a prolonged shelf life. Finally, four main islands of research has been individuated in the field of 3D food printing such as *Innovative 3D Food Printing by Extrusion, Rheology Control and Printing Fidelity Optimization, Design and Macro- and Microstructure* and *Personalized Food.* While the first two are connected by the study of rheological properties of food-ink, the others appear weakly connected. This result highlights the opportunity to reinforce the relationship between digital design, microstructure and novel personalized food products with unprecedented sensory perception or capability of mitigating the mastication and swallowing problems of vulnerable consumers. Finally, the study of food-inks rich in protein – which appears few and isolated - should be enlarged and linked to other aspects such as the digital design, the effects on microstructure and the creation of personalized food products.

5. References

1. Agostinacchio, F., Mu, X., Dirè, S., Motta, A., & Kaplan, D.L. (2020). In situ 3D printing: Opportunities with Silk Inks. *Trends in Biotechnology*, in press, https://doi.org/10.1016/j.tibtech.2020.11.003 .
2. Baiano, A. (2020). 3D printed Foods: A comprehensive Review on Technologies, Nutritional value, safety, consumer, attitude, nutritional value, safety, consumer attitude, regulatory framework, and economic and sustainability issues. *Food Reviews International*, in press, https://doi.org/10.1080/87559129.2020.1762091.
3. Beg, S., Almalki, W.H., Malikm A., Farhan, M., Aatif, M., Rahman, Z., Alruwaili, N.K., Alrobaian, M., Tarique, M., & Rahman, M. (2020). 3D printing for drug delivery and biomedical applications. *Drug Discovery Today*, 25, 9, 1668-1681. https://doi.org/10.1016/j.drudis.2020.07.007.
4. Betran, F.A., Jhaveri, S., Lutz, R., Isbister, K., & Wilde, D. (2019). Making Sense of Human-Food Interaction. CHI 2019, may 4-9, Slasgiw, Scotland, UK. https://doi.org/10.1145/3290605.3300908.
5. Bueno, G., Altes, P., Carnovale, L., Hernandez, N., Sancho, J., Zamora, G., Esteban, C., & Llagostera, S. (2019). Present or Future: Rapid 3d Printing Prototyping Technology for Aortic Aneurysm Surgery Planning and Its Utility in Open and Endovascular Treatment. *European Journal of Vascular and Endovascular Surgery*, 58, 6, e271-e272, https://doi.org/10.1016/j.ejvs.2019.06.869 .
6. Buchanan, C., & Gardner, L., (2019). Metal 3D printing in construction: A review of methods, research, applications, opportunities and challenges. *Engineering Structures*, 180, 332-348. https://doi.org/10.1016/j.engstruct.2018.11.045
7. Chan, S.S.L., Pennings, R.M., Edwards, L., Franks, G. (2020). 3D printing of clay for decorative architectural applications: effect of solids volume fraction on rheology and printability. Additive manufacturing, 35, 101335, https://doi.org/10.1016/j.addma.2020.101335
8. Charlebois, S., & Juhasz, M. (2018). Food futures and 3D printing: Strategic market foresight and the case of structure 3D. *International Journal on Food System Dynamics*, 9 (2), 138-148.
9. Chaundry, B.M., Connelly, K., Siek, K.A., & Welch, J.L. (2012). Formative evaluation of a mobile liquid portion size estimation interface for people with varying literacy skills. *Journal of Ambient Intelligence and Humanized Computing*, https://doi:10.1007/s12652-012-0152-9 .
10. Cohen, D., Lipton, J., Cutler, M., Coulter, D., Vesco, A., & Lipson, H. (2009). Hydrocolloid printing: a novel platform for customized food production. Proceedings of the 20th Annual International Solid Freeform Fabrication Symposium, pp. 807–818. Austin, TX.
11. Comber, R., Choi, J.H.j. Hoonhout, J., & O'Hara, K. (2014). Designing for human-food-interaction: an introduction to the special issue on 'food and interaction design'. Int. J. Human-Computer Studies, 72, 181-184. https://doi.org/10.1016/j.ijhcs.2013.09.001
12. Comminal, R., Leal da Silva, W.R., Andersen, T.J., Stang, H., & Spangenberg, J. (2020). Modelling of 3D concrete printing based on computational fluid dynamics. *Cement and Concrete Research*, 138, 106256. https://doi.org/10.1016/j.cemconres.2020.106256

13. D'Angelo, G., Hansen, H.N., & Hart, A.J. (2016). Molecular Gastronomy Meets 3D Printing: layered Construction via reverse Spherification. *3D Printing and Additive Manufacturing*, 3, 153-159.
14. Dankar, I., Haddarah, A., Omar, F.E.L., Sepulcre, F., & Pujolà, M. (2018). 3D printing technology: The new era for food customization and elaboration, *Trends in Food Science & Technology*, 75, 231-242, ISSN 0924-2244, https://doi.org/10.1016/j.tifs.2018.03.018 .
15. Derossi, A., Caporizzi, R., Ricci, I., & Severini, C. (2018). Critical variables in 3D food printing. In: Godoi, F.C., Bhandari, B., Prakash, S., Zhang, M. *Fundamentals of 3D Food Printing and Applications*, 2018, 41-91. https://doi.org/10.1016/B978-0-12-814564-7.00003-1
16. Derossi, A., Caporizzi, R., Oral, M.O., & Severini, C. (2020a). Analyzing the effects of 3D printing process per se on the microstructure and mechanical properties of cereal food products, *Innovative Food Science & Emerging Technologies*, 66, 102531, https://doi.org/10.1016/j.ifset.2020.102531 .
17. Derossi, A., Paolillo, M., Caporizzi, R., & Severini, C. (2020b). Extending the 3D food printing test at high speed. Material deposition and effect of non-printing movements on the final quality of printed structures. *Journal of Food Engineering*, 275, 109865. https://10.1016/j.jfoodeng.2019.109865
18. Derossi, R. Caporizzi, M. Paolillo, & C. Severini (2021). Programmable texture properties of cereal-based snack mediated by 3D printing technology. *Journal of Food Engineering*, 289, 110160, https://doi.org/10.1016/j.jfoodeng.2020.110160.
19. Dianez, L., Gallegos, C., Brito-de la Fuente, E., Martinez, I., Valencia, C., Sanchez, M.C., Diaz, M.J., & Franco, J.M. (2019). 3D printing in situ gelification of k-carrageenan solutions: effect of printing variables on the rheological response. *Food Hydrocolloids*, 87, 321-330.
20. Dick, A., Bhandari, B., & Prakash, S. (2019). 3D printing of meat. *Meat Science*, 153, 35-44. https://doi.org.10.1016/j.meatsci.2019.03.005
21. El Mohadab, M., Bouikhalene, B., & Safi, S. (2020). Bibliometric method for mapping the state of the art of scientific production in Covid-19. *Chaos, Solitons and Fractals*, 139, 110052. https://doi.org/10.1016/j.chaos.2020.110052
22. FAO (2020). The state of Food Security and Nutrition in the World. http://www.fao.org/3/ca9692en/CA9692EN.pdf
23. Fahmy, A.R., & Becker, T., (2020). 3D printing and additive manufacturing of cereal-based materials: Quality analysis of starch-based systems using a camera-based morphological approach. *Innovative Food Science and Emerging Technologies*, 63, 102384
25. Feng, C., Chang, M., & Bhandari, B. (2019). Materials properties of printable edible inks and printing parameters optimization during 3D printing: a review. *Critical review in food science and nutrition*, 59 (19), 3074-3081, https://doi.org/10.1080/10408398.2018.1481823
26. Gholamipour-Shirazi, A., Norton, I.T., & Mills, T. (2019). Designing hydrocolloids based food-ink formulations for extrusion 3D printing. *Food Hydrocolloids*, 95, 161-167. https://doi.org/10.1016/j.foodhyd.2019.04.011 .

27. Gholamipour-Shirazi, A., Kamlow, M.A., Norton, I.T., & Mills, T. (2020). How to formulate for structure and texture via medium of additive manufacturing-A review. *Foods*, 9 (4), 497, https://doi.org/10.3390/foods9040497.
28. Gibson, M.A., Mykulowycz, N.M., Shim, J., Fontana, R., Schmitt, P., Roberts, A., Ketkaew, J., Shao, L., Chen, W., Bordeenithikasem, P., Myerberg, J.S., Fulop, R., Verminski, M.D., Sachs, E.M., Chiang, Y.M., Schuh, C.A., Hart, J., & Schroers, J. (2018). 3D printing metals like thermoplastics: Fused filament fabrication of metallic glasses. *Materials Today*, 21, 7, 697-702, https://doi.org/10.1016/j.mattod.2018.07.001.
29. Godoi, F.C., Prakash, S., & Bhandari, B. (2016). 3d printing technologies applied for food design: status and prospects. *Journal of Food Engineering*, 179, 44-54. https://doi.org/10.1016/j.jfoodeng.2016.01.025
30. Godoi, F., Bhandari, B., Prakash, S., & Zhang, M. (2018). Fundamentals of 3D Food Printing and Applications. Academic press. ISBN 9780128145647. https://doi.org/10.1016/B978-0-12-814564-7.00003-1
31. Gray, N. (2010). Looking to the future: creating novel food using 3D printing. http://www.foodnavigator.com/Science/Looking-to-the-future-Creating-novel-foods-using-3D-printing.
32. Guo, C.F., Zhang, M., & Bhandari, B. (2019). A comparative study between syringe-based and screw-based food printers by computational simulation. *Computers and Electronics in Agriculture*, 162, 397-404. 10.1016/j.compag.2019.04.032
33. Guo, C., Zhang, M., & Devahastin, S. (2021). Color/aroma changes of 3D-Printed buckwheat dough with yellow flesh peach as triggered by microwave heating of gelatin gum Arabic complex coacervates. *Food Hydrocolloids*, 112, 106358. https://doi.org/10.1016/j.foodhyd.2020.106358
34. Handral, H., Hua Tay, S., Wan Chan, W., & Choudhury, D. (2020). 3D printing of cultered meat products. *Critical reviews in food science and nutrition*, 21, 1-10. 10.1080/10408398.2020.1815172
35. Hashimoto, A., Funatomi, T., Ueda, M., & Yamakata, Y. (2012). Recognizing ingredients at cutting process by integrating multimodal features. CEA'12. November, 2, doi:10.1145/2390776.2390780.
36. He, C., Xhang, M., & Guo, C. (2020a) 4D printing of mashed potato/purple sweet potato puree with spontaneous color change.. *Innovative Food Science and Emerging Technologies*, 59, 102250. https://doi.org/10.1016/j.ifset.2019.102250
37. He, C., Zhang, M., & Fang, Z. (2020b). 3D printing of food: pretreatment and post-treatment of materials. *Critical reviews in food science and nutrition*, 60 (14), 2379-2392. https://doi.org/10.1080/10408398.2019.1641065
38. Jerman, M., Zelenak, M., Lebar, A., Foldyna, V., Foldyna, J., & Valentincic, J. (2021). Observation of cryogenically cooled ice particles inside the high-speed water jet. *Journal of Materials Processing Technology*, 289, 116947. https://doi.org/10.1016/j.jmatprotec.2020.116947
39. Jia, F., Wang, X., Mustafee, N., Hao, L. (2016). Investigating the feasibility of supply chain-centric business models in 3D chocolate printing: A simulation study. Technological Forecasting and Social Change, 102, 202-213. DOI: 10.1016/j.techfore.2015.07.026
40. Jiang, H., Zheng, L., Zou, Y., Han, S., & Wang, S. (2019). 3D food printing: main components selection by considering rheological properties. *Critical Reviews in Food*

Science and Nutrition, 59 (14), 2335-2347. https://doi.org/10.1080/10408398.2018.1514363
41. Jiang, J., Zhang, M., Bhandari, B., & Cao, P. (2020). Current processing and packing technology for space foods: a review. *Critical reviews in Food Science and Nutrition*, 60 (21), 3573-3588. https://doi.org/10.1080/10408398.2019.1700348
42. Julkarnyne M. Habibur Rahman, MD Nahin Islam Shiblee, Kumkum Ahmed, Ajit Khosla, Masaru Kawakami, & Hidemitsu Furukawa (2020). Rheological and mechanical properties of edible gel materials for 3D food printing technology. *Heliyon*, 6, 12, e05859, https://doi.org/10.1016/j.heliyon.2020.e05859.
43. Kambhampati, S.B.S., Vaishya, R., & Vaish, A. (2020). Unprecedented surge in publications related to COVID-19 in the first three months of pandemic: A bibliometric analytic report. *Journal of Clinical Orthopaedics and Trauma*, 11, 3, S304-S306, https://doi.org/10.1016/j.jcot.2020.04.030 .
44. Karyappa, R., & Hashimoto, M. (2019). Chocolate-based ink three-dimensional printing (Ci3DP). *Scientific Report*, 9 (1), 14178. https://doi.org/10.1038/s41598-019-50583-5
45. Kim, N.P., Cepeda, B., Kim, J., Yue, G., Kim, S., & Kim, H. (2018). IoT controlled screw-type 3D food printer using single line design technique. International Conference on Computational Science and Computational Intelligence, CSCI 2018, December 2018, Las Vegas.
46. Kern, C., Weiss, J., & Hinrichs, J. (2018). Additive layer manufacturing of semi-hard model cheese: effect of calcium levels on thermo-rheological properties and shear behavior. *Journal of Food Engineering*, 235, 89-97.
47. Kuo, C.C., Qin, H., Cheng, Y., Jiang, X., & Shi, X. (2021). An integrated manufacturing strategy to fabricate delivery system using gelatin/alginate hybrid hydrogels: 3D printing and freeze-drying. *Food Hydrocolloids*, 111, 106262, https://doi.org/10.1016/j.foodhyd.2020.106262 .
48. Le-Bail, Maniglia, B.C., & Le-Bail, P. (2020). Recent advances and future perspective in additive manufacturing of foods based on 3D printing. *Current Opinion in Food Science*, 35, 54-64, https://doi.org/10.1016/j.cofs.2020.01.009 .
49. Li, N., Zhang, J., Xing, W., Ouyang, D., & Liu, L. (2018). 3D printing of Fe-based bulk metallic glass composites with combined high strength and fracture toughness. *Materials & Design*, 143, 285-296, https://doi.org/10.1016/j.matdes.2018.01.061 .
50. Lipton, J.I., Cutler, M., Nigl, F., Cohen, D., & Lipson, H. (2015). Additive manufacturing for the food industry. *Trends in Food Science & Technology*, 43, 114-123. https://doi.org/10.1016/j.tifs.2015.02.004
51. Liu, Z., Zhang, M., Bhandari, B., & Yang, C. (2018). Impact of rheological properties of mashed potatoes on 3D printing. *Journal of Food Engineering*, 220, 76-82. https://doi:10.1016/j.foodeng.2017.04.017 .
52. Liu, Z., Bhandari, B., Prakash, S., Mantihal, S. & Zhanga, M. (2019). Linking rheology and printability of a multicomponent gel system of carrageenan-xanthan-starch in extrusion based additive manufacturing. *Food Hydrocolloids*, 87, 413-424. https://doi.org/10.1016/j.foodhyd.2018.08.026 .
53. Manoj, A., Bhuyan, M., Raj Banik, S., & Ravi Sankar, M. (2020). 3D Printing of Nasopharyngeal Swabs for COVID-19 Diagnose: Past and Current Trends. *Materials Today*: Proceedings. doi: 10.1016/j.matpr.2020.11.505

54. Mantihal, S., Prakash, S., Godoi, F.C., Bhandari, B. (2017). Optimization of chocolate 3D printing by correlating thermal and flow properties with 3D structure modeling. Innovative Food Science and Emerging Technologies, 44, 21-29. DOI: 10.1016/j.ifset.2017.09.012
55. Mantihal, S., Prakash, S., & Bhandari, B. (2019a). Texture-modified 3D printed dark chocolate: sensory evaluation and consumer perception study. *Journal of Texture Studies*, 50 (5), 386-399. DOI: 10.1111/jtxs.12472
56. Mantihal, S., Prakash, S., Godoi, F.C., Bhandari, B. (2019b). Effect of additives on thermal, rheological and tribological properties of 3D printed dark chocolate. *Food Research International*, 119, 161-169. DOI: 10.1016/j.foodres.2019.01.056
57. Mantihal, S., Kobun, R., & Lee, B.-B (2020). 3D food printing of as the new way of preparing food: A review. *International Journal of Gastronomy and Food Science,* 22, 100260. https://doi.org/10.1016/j.ijgfs.2020.100260
58. Mascarenhas, C., Ferreira, J.J., & Marques, C. (2018). University-industry cooperation: a systematic literature review and research agenda. *Science and Public Policy*, 45, 708-718. https://doi.org/10.1093/scipol/scy003
59. Mingers, J., & Leydesdorff L. (2015). A review of theory and practice in scientometrics. European *Journal of Operational Research*, 246, 1-19. https://doi.org/10.1016/j.ejor.2015.04.002
60. Murr, L.E. (2020). Metallurgy principles applied to powder bed fusion 3D printing/additive manufacturing of personalized and optimized metal and alloy biomedical implants: an overview. *Journal of Materials Research and Technology*, 9, 1087-1103. https://doi.org/10.1016/j.jmrt.2019.12.015
61. Nachal, N., Moses, J.A., Karthik, P., & Chinnaswamy, A. (2019). Application of 3D printing in Food processing. *Food Engineering Reviews*, 11, 3, https://doi:10.1007/s12393-019-09199-8
62. NASA. Funds system for 3D printing in space (2017). *Metal Powder Report*, 72, 6, Pages 434-435, https://doi.org/10.1016/j.mprp.2017.10.024.
63. Nesic, D., Durual, S., Marger, L., Mekki, M. Sailer, I., & Scherrer, S.S. (2020). Could 3D printing be the future for oral soft tissue regeneration? *Bioprinting*, 20, e00100, https://doi.org/10.1016/j.bprint.2020.e00100 .
64. Otlet, P. (1934). Traité de documentation: le livre sur le livre, théorie et pratique. Brussels: Editions Mundaneum.
65. Pant, A., Lee, A.Y., Karyappa, R., Lee, C.P., An, J., Hashimoto, M., Tan, U.X., Wong, G., Chua, C.K., & Zhang, Y. (2021). 3D food printing of fresh vegetables using food hydrocolloids for dysphagic patients. *Food Hydrocolloids*, 114, 106546, ISSN 0268-005X, https://doi.org/10.1016/j.foodhyd.2020.106546 .
66. Park, J.Y., & Nagy, Z. (2018). Comprehensive analysis of the relationship between thermal comfort and building control research – A data-driven literature review. *Renewable and sustainable energy reviews*, 82, 2664-2679. https://doi.org/10.1016/j.rser.2017.09.102
67. Park, S.M., Kim, H.W., & Park, H.J. (2020). Callus-based 3D printing for food exemplified with carrot tissues and its potential for innovative food production. *Journal of Food Engineering*, 271, 109781, https://doi.org/10.1016/j.jfoodeng.2019.109781 .

68. Perianes-Rodrguez, A., Waltman, L., & van Eck, N.J. (2016). Constructing bibliometric networks: A comparison between full and fractional counting. *Journal of Informetrics*, 10, 4, 1178-1195, https://doi.org/10.1016/j.joi.2016.10.006 .
69. Perez, B., Nykvist, H., Brogger, A.F., Larsen, M.B., & Falkeborg, M.F. (2019). Impact of macronutrients printability and 3D-printer parameters on 3D-food printing: A review. *Food Chemistry*, 287, 249-257. https://doi.org/10.1016/j.foodchem.2019.02.090
70. Phuhongsung, P., Zhang, M., & Bhandari, B. (2020). 4D printing of products based on soy protein isolate via microwave heating for flavor development. *Food Research International*, 137, 109605. https://doi.org/10.1016/j.foodres.2020.109605
71. Piyush, Kumar, R., & Kumar, R., (2019). 3D printing of food materials: A state of art review and future applications. Materials Today: Proceedings of the 1st International Symposium on Synthesis, Characterization and Processing of Inorganic, Bio and Nano Materials, 33, 1463-1467. https://doi.org/10.1016/j.matpr.2020.02.005
72. Portanguen, A., Tournayre, P., Sicard, J., Astruc, T., Mirade, P.S. (2019). Toward the design of functional foods and biobased products by 3D printing: A review. *Trends in Food Science and Technology*, 86, 188-198.
73. Price, D.J.d.S. (1976). A general theory of bibliometric and other cumulative advantage processes. *Journal of the American Society for Information Science*, 27, 292-306. https://doi.org/10.1002/asi.4630270505
74. Pulatsu, E., Su, J.W., Lin, J., & Lin, M. (2020). Factors affecting 3D printing and post-processing capacity of cookie dough. *Innovative Food Science & Emerging Technologies*, 102316. https://doi.org/10.1016/j.ifset.2020.102316
75. Pulatsu, E., & Lin, M. (2021a). A review on customizing edible food materials into 3D printable inks: Approaches and strategies. *Trends in Food Science & Technology*, 107, 68-77, https://doi.org/10.1016/j.tifs.2020.11.023 .
76. Pulatsu, E., Su, J.W., Kenderes, S.M., Lin, J., Vardhanabhuti, B., & Lin, M. (2021). Effects of ingredients and pre-heating on the printing quality and dimensional stability in 3D printing of cookie dough. *Journal of Food Engineering*, 294, 1104, https://doi.org/10.1016/j.jfoodeng.2020.110412 .
77. Ramundo, L., Taisch, M., Terzi, S. (2016). State of the art of technology in the food sector value chain towards the IoT. *IEEE 2nd International Forum on Research and Technologies for Society and Industry Leveraging a Better Tomorrow*, Bologna; Italy; 7 September.
78. Rando, P, & Ramaioli, M. (2021). Food 3D printing: effect of heat transfer on print stability of chocolate. *Journal of Food Engineering*, 294, 110415. https://doi.org/10.1016/j.jfoodeng.2020.110415
79. Rayna, T., & Striukova, L., (2016). From rapid prototyping to home fabrication: How 3D printing is changing business model innovation. *Technological Forecasting and Social Change*, 102, 214 224, https://doi.org/10.1016/j.techfore.2015.07.023.
80. Sartal, A., Carou, D., Dorado-Vicente, R., & Mandayo, L. (2019). Facing the challenges of the food industry: Might additive manufacturing be the answer?. Proceedings of the Institution of Mechanical Engineers, Part B: *Journal of Engineering, Manufacture*, 233, (8), 1902-1906.
81. Schutyser, M.A.I., Houlder, M., de Wit, M., Buijsse, C.A.P., & Alting, A.C. (2018). Fused deposition modeling of sodium caseinate dispersions. *Journal of Food Engineering*, 220, 49-55. https://doi.org/10.1016/j.jfoodeng.2017.02.004

82. Severini, C., & Derossi, A. (2016). Could the 3D printing technology be a useful strategy to obtain customized nutrition?. *Journal of clinical Gastroenterology*, 50, S175-S178. DOI: 10.1097/MCG.0000000000000705
83. Severini, C., Azzollini, D., Albenzio, M., & Derossi, A. (2018a). On printability, quality and nutritional properties of 3D printed cereal based snacks enriched with edible insects. *Food Research International*, 106, 666-676. https://doi.org/10.1016/j.foodres.2018.01.034
84. Severini, C., Derossi, A., Ricci, I., Caporizzi, R., & Fiore, A. (2018b). Printing a blend of fruit and vegetables. New advances on critical variables and shelf life of 3D edible objects. *Journal of Food Engineering*, 220, 89-10, https://doi.org/10.1016/j.jfoodeng.2017.08.025 .
85. Shao, B., Li, X., & Bian, G. (2021). A survey of research hotspots and frontier trends of recommendation system from the perspective of knowledge graph. *Expert Systems with applications*, 165, 113764. https://doi.org/10.1016/j.eswa.2020.113764
86. Stamati, V., Antonopoulos, G., Azariadis, Ph., Fudos, I. (2011). A parametric feature-based approach to reconstructing traditional filigree jewelry (2011). Computer-Aided Design, 12, 1814-1828, https://doi.org/10.1016/j.cad.2011.07.002
87. Steenhuis, H.J., Fang, X., & Ulusemre, T. (2018). Strategy in 3D printing of Food. PICMET - Portland International Conference on Management of Engineering and Technology: Managing Technological Entrepreneurship: The Engine for Economic Growth, Proceedings4 October 2018, Honolulu; United.
90. Subramayna, S.H., Lama, B., & Acharya, K.P. (2020). Impact of COVID-19 pandemic on the scientific community. *Qatar Med J.*, 1, 21. doi: 10.5339/qmj.2020.21
91. Sun, J., Zhou, W., Huang, D., Fuh, J.Y., & Hong, G.S. (2015). An overview of 3D printing technologies for food fabrication. *Food Bioprocess Technology,* 8 (8), 1605e1615. http://dx.doi.org/10.1007/s11947-015-1528-6 .
92. Sun, J., Peng, Z., Yan, L., Fuh, J.Y.H., & Hong, G.S. (2015). 3D food printing-An innovative way of mass customization in food fabrication. *International Journal of Bioprinting*, 1 (1), 27-38.
93. Sweileh, W.M., Al-Jabi, S.W., Zyoud, A.H., Sawalha, A., & Abu-Taha, A. (2018). Global research output in antimicrobial resistance among uropathogens: A bibliometric analysis (2002-2016). *Journal of Global Antimicrobial Resistance*, 13, 104-114. DOI: 10.1016/j.jgar.2017.11.017
94. Mantihal, S., Prakash, S., Godoi, F.C., Bhandari, B. (2017). Optimization of chocolate 3D printing by correlating thermal and flow properties with 3D structure modeling. *Innovative Food Science & Emerging Technologies*, 44, 21-29, https://doi.org/10.1016/j.ifset.2017.09.012 .
95. Mantihal, S, Kobun, R, Lee, B.B. (2020). 3D food printing of as the new way of preparing food: a review. International journal of Food Gastronomy and Food Science, 22, 100260, https://doi.org/10.1016/j.ijgfs.2020.100260.
96. Tagami, T., Ito, E., Kida, R., Hirose, K., Noda, T., & Ozeki, T. (2021). 3D printing of gummy drug formulations composed of gelatin and an HPMC-based hydrogel for pediatric use. *International Journal of Pharmaceutics*, 594, 120118, https://doi.org/10.1016/j.ijpharm.2020

97. Tan, C., Toh, W.Y., Wong, G., & Lin, L. (2018). Extrusion-based 3D food printing – Material and machines, International Journal of Bioprinting, 4 (2). doi: 10.18063/IJB.v4i.143.
98. Tian, H., Wang, K., Lan, H., Wang, Y., Hu, Z., & Zhao, L. (2021). Effect of hybrid gelator systems of beeswax-carrageenan-xanthan on rheological properties and printability of litchi inks for 3D food printing. *Food Hydrocolloids*, 113, 106482. https://doi.org/10.1016/j.foodhyd.2020.106482 .
99. Uribe-Wandurraga, Z.N., Zhang, L., Noort, M.W.J., Schutyser, M.A.I., Garcia-Segovia, P., & Martinez-Monzo, J. (2020). Printability and physicochemical properties of microalgae-enriched 3D printed snacks. *Food and Bioprocess technology*, 13, 2029-2042. https://doi.org/10.1007/s11947-020-02544-4
100. van Eck, N.J., & Waltman, L. (2009). How to normalize co-occurrence data ? An analysis of some well-know similarity measures. *Journal of the American Society for Information Science and technology*, 60, (8), 1635-1615. https://doi.org/10.1002/asi.21075
101. Voon, S.L., An, J, Wong, G., Zhang, Y., & Chua, C.K., (2019). 3D food printing a categorized review of inks and their development. *Virtual and Physical Prototyping*. 10.1080/17452759.2019.1603508
102. Waltman, L., van Eck, N.J., & Noyons, E.C.M. (2010). A unified approach to mapping and clustering of bibliometric networks. *Journal of Infometrics*, 4, 629-635. https://doi.org/10.1016/j.joi.2010.07.002
103. Wang, L., Zhang, M., Bhandaru, B., & Yang, C. (2018). Investigation of surimi gel as primising food material for 3D printing. *Journal of Food Engineering*, 220, 101-108. https://doi.org/10.1016/j.jfoodeng.2017.02.029
104. Want, S. (2020). 3D Printing clothing design based on wireless sensors and FPGA. *Microprocessors and Microsystems*, 103407, https://doi.org/10.1016/j.micpro.2020.103407 .
105. Yang, F., Zhang, M., Bhandari, B., & Liu, Y. (2018). Investigation on lemon juice gel as food material for 3D printing and optimization of printing parameters. *LWT, Food Science and technology*, 87, 67-76. https://doi.org/10.1016/j.lwt.2017.08.054
106. Zhang, L., Lou, Y., & Schutyser, M.A.I. (2018). 3D printing of cereal-based food structures containing probiotics. *Food Structure*, 18, 14-22. https://doi.org/10.1016/j.foostr.2018.10.002
107. Zhao, L., Zhang, M., Chitrakar, B., & Adhikari, B. (2020). Recent advances in functional 3D printing of foods: a review of functions of ingredients and internal structures. *Critical Reviews in Food Science and Nutrition*, in press. https://doi.org/10.1080/10408398.2020.1799327
108. Zhu, S., Stieger, M. A., van der Goota, A. J., & Schutyser, M. A. I. (2019). Extrusion-based 3D printing of food pastes: Correlating rheological properties with printing behaviour. *Innovative Food Science & Emerging Technologies*, 58, 102214. https://doi.org/10.1016/j.ifset.2019.102214.

Chapter 2 Analyzing the most promising innovations in food printing. Programmable food texture and 4D foods

Authors: Oral, M.O., Derossi, A., Caporizzi, R., Severini, C.

Department of Agriculture, Food, Natural resources and Engineering (DAFNE), University of Foggia, Italy

Corresponding author: **antonio.derossi@unifg.it**

Abstract

With the increase of the published scientific results, 3D food printing is suddenly growing with many opportunities and possibilities to contribute to the current challenges of the food system. We present a brief review focused on two promising applications that are drawing where and how food printing is currently evolving: 4D food printing and the creation of programmable food texture. We found an interesting number of scientific documents regarding 4D food printing that generates a time evolution of colour, shape, dimension, aroma, and nutritional content. Among others, these changes are activated by many external stimuli such as acid/alkaline conditions, the anisotropic behaviour of printed materials during drying, but further experiments and discussions are still needed considering the effect of such stimuli on other important points such as safety and quality parameters. Contrarily, very few results have been published on the programmable food texture, but these few allow us to glimpse breakthrough innovations capable to completely renew the texture perception and mechanical properties of food products with extreme benefits for market innovation and consumer's desires and needs such as the creation of a fragile structure for people suffering dysphagia.

Keywords: Food properties; dynamic changes; 4D printing; digital design; human-food-computer interaction; food texture.

1. Roadmap of the 3D Food Printing

To tackle the current challenges for a world where all people can easily access to enjoyable, sustainable, safe, and healthy food, there is the need for innovative technologies capable to renew how foods are produced, transported, distributed, and consumed. After the success of the applications of additive manufacturing (AM) in many industrial fields, the idea of 3D food printing (3DFP) has motivated the interest of the research community and food industry for its many potentials of innovation. Indeed, Cohen et al. (2009) stated that '*after solving the main issues of slow printing and price, the remaining question is what the ways in which 3DFP will completely modify the food sector, while no doubts on whether 3DFP may affect food manufacturing and consumption*'. Among the AM techniques, the deposition of food formulas while the printer moves in the *X, Y, Z* space replicating a digital model, is the prevalent methodology (Nijdam et al., 2021; Tomasevic et al., 2021). Such principles make the 3DFP the only technique having possibilities for a profound renewal of how food is produced. The intricated movements of the printer add a huge number of degrees of freedom allowing feasible the architectural building of complex structures never realized before. At the moment of writing this paper, the state of art of 3DFP shows approximately 170 scientific documents published from 2007 to 2020 (Derossi et al., 2021a) of which 53% of original papers, 22% of conference papers and 11.7% of reviews.

Undoubtedly, the first goal motivating the designed experiments was the creation of innovative and fascinating shapes thanks to the design of dozens of digital models which are replicated through layer-by-layer deposition of thin filaments of food formulas. To accomplish these shapes, also considering the huge diversities of the edible materials, researchers have dedicated most of the experimental activities to study the printability of several food materials and the effects of printing variables on the quality of the end products. As an example, experiments have been performed by using wheat dough, fruit and vegetables, cheese, meat and also fish (Tomasevic et al., 2021; Zhao et al., 2021). Likewise, many efforts have been committed to improving the printability of food formulas by using traditional hydrogel (Paolillo et al., 2021; Zhao et al., 2021) or less common oleogels and hybrid gels (Tian et al., 2021). Moreover, a large body of papers has been focused on the effects of the most important printing variables such as printing speed, layer height, nozzle diameter, infill density, infill patterns, etc., on the overall quality parameters of printed food, primarily on the printing fidelity and structural stability (Derossi et al., 2018a; Feng et al., 2020; Guo et al., 2019; Severini et al., 2016; Pulatsu et al., 2020; Wang et al., 2018; Xu et al., 2020). In this review, going beyond this basic information widely discussed, we want to point out where and how the 3DFP technology is evolving and what type of impact such growth may have on the current challenges of the food system.

While growing the number of scientific experiments, several hinders limiting the practical application of 3DFP at the industrial level or for home use have been highlighted (Derossi et al., 2021b) but, in addition, new clear opportunities and possibilities have been identified and are stimulating the research activities. Indeed, all actors of the food chain may stand to benefit from 3DFP applications, with on-demand food production, adoption of a consumers-centric approach, innovative food perceptions, satiety control, food loss reduction, personalized nutrition, customized sensory products, digital transition in food manufacturing, and novel business creation as well.

So, 3DFP has ambitions and potential to contribute to the transformation of the food system and accelerating the progress under the United Nations Development Goals (SDGs), the UE Farm to Fork Strategy, the EU Green Deal. 3DFP may help for a better food system capable of producing in a sustainable way, foods safe, highly enjoyable, made of a balanced mixture of nutrients and functional compounds for a healthy and active life. To accomplish all this, a roadmap consisting of four main key-priorities area (KPs) from which many actions resulting in a set of achievements allowing to realize aforementioned goals may be described as reported in Figure 1.

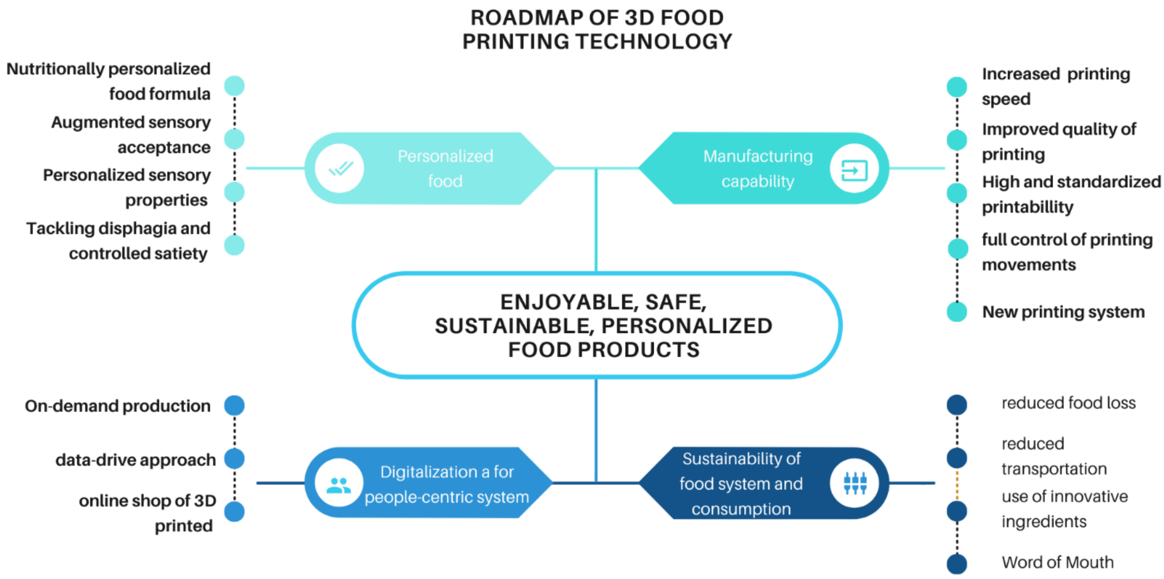


Fig. 1. Schematic representation of the roadmap of 3D food printing.

For instance, the exclusive properties of 3DFP release a set of opportunities for the creation of nutritional and sensorial personalized food (Pulatsu and Lin, 2021) matching nutritional requirements of small consumer's group with profound benefits on the mitigation of non-communicable diseases (NCD) and childhood obesity (KP1). This made 3DFP capable of positively impacting UN SDG 2.2 and UN SDG 3.4 targets. Also, the benefits on people's health could be reinforced by a multi-disciplinary approach connecting 3DFP with the data-driven approach as well as artificial intelligence and machine learning (KP 1 and KP2). If medical data and other information regarding the health status, lifestyle, gender, habits, were used and modeled to get accurate designs of the food formula, the 3DFP could be employed to drastically enhance the level of food customization (Derossi et al., 2018a). On this specific point, the approach of artificial intelligence (AI) and machine learning could be very useful for designing novel personalized food both in terms of nutritional needs and sensory perceptions (Camarena, 2020; Pini et al., 2019). AI tools could learn from consumers' habits and drive the nutrient recommendations (Eftimov et al., 2017) employable for printing personalized food products. Also, considering novel food perception and consumer's acceptance, Pini et al. (2019) used an AI approach to generate novel combinations of existing parameters of different kinds of raw foods – i.e., colour, nutrients, sounds, etc. – capable to sustain innovation and creativity in food preparation. These realizations could impact also the ongoing pandemic situation by a great level of precision capable to reinforce the immune response to viruses, reducing the risk of malnutrition of hospitalized patients or for people in quarantine, or alleviate mastication and swallowing problems of elderly (Derossi et al., 2021b). Recent experiments created innovative products enriched with probiotics aiming to modulate the immune systems by the inhibition of pathogenic microorganisms (Liu et al., 2020; Yoha et al. 2021; Zhang et al., 2018). Furthermore, 3D food printing may facilitate the consumption of healthy food and the control of how much food is eaten. The opportunities of maximizing sensory enjoyment and of controlling/modulating satiety would be of profound interest for mitigating several diseased strictly liked to the consumption behaviours (Spence, 2021). A fully controlled deposition of food materials would allow creating new shapes, dimensions, overlapping colour, taste and texture. So, healthy food, or single ingredients, traditionally rejected from specific consumers group because considered unpleasant – as occurring for broccoli or spinach in child age – could be used to create 3D printed product with maximized enjoyment of the consumer's experience (Sun et al., 2015; Tomasevic et al., 2021). Examples of customized snacks though for children age, between 3 and 10 years, have been realized by designing a food formula capable of matching their dietary requirements (Derossi et al., 2018b). However, the heating behaviour is extremely affected by many factors including and primarily, visual appealing and multisensory taste and flavour experience. On this, 3DFP could significantly contribute maximizing sensory acceptance by the creation of new shapes, depositing layers with different colours, complex taste and odour, creating asymmetric food structures, and by the 4D printing that allows triggering changes in sensory properties as a function of time.

To go beyond the basic aspect of 3DFP, this review is focused primarily on the two most recent and intriguing hot-points - the 4D food printing and the creation of programmable food texture – which, looking to the future, could guide innovative solutions contributing to current EU strategic agenda of the food system. More specifically, Section 2 analyzes and discusses the state of the art of 4D printing while Section 3 discusses the possibility to create a programmable food structure with desired texture properties. Finally, Section 4 resumes and highlights the main points.

2. *4D Food Printing: using external stimuli to activate changes over time of 3D printed food.*

4D food printing, 4DFP, occurs when 3D printed edible structures undergo physical and chemical changes as a function of time. To do this a proper and accurate use of an external source of stimuli (i.e., light, temperature, pH, pressure, etc.) and material stimuli (i.e. gels, puree, cereal dough, etc.) may activates, in a controlled manner, the changes over time of the colour, shape, and flavour of 3D printed food. At the time of writing this paper, some pioneering experiments have been performed and the obtained results extend the potential applications of 3DFP as well as the potential interest of the food industry. Colour change over time triggered by different stimuli is the most studied application of 4DFP. He et al. (2020a) printed mashed potato (MP) and purple sweet potato puree (PSP) to use the property of anthocyanins - of which sweet potatoes are rich - to undergo colour change when exposed to different pH values. The authors prepared MP in a range of pH between 2.5 and 7.8. When 3D printed cylinders of MP and PSP were stacked, the authors observed significant colour changes of MP as a function of time while the antochyanins diffused through MP layers at different pH (Figure 2a). This preliminary experiment was then extended by designing complex pyramids with interchanged layers of MP at different pH and PSP (Figure 2b). From these results, we can speculate that further advances could be obtained by modulating the thickness of both the MP and SPS layers influencing the first diffusion rate of the anthocyanins thus obtaining a huge variety of colour changes. Another interesting approach to obtain the colour evolution was proposed by Chen et al. (2021a) who prepared a gel emulsion of lotus root containing curcumin powder. In this case, the colour change – from yellow to pink – was activated by exposing the samples to alkaline conditions generated by microwave heating at 280 W for 1-3 min (Figure 2c). In addition to the colour change, after microwave heating, the transition from semi-solid to solid was obtained, thus changing its textural properties. Such change generated an increase of hardness and gumminess resulting in augmented force to chew the samples. So, the use of microwaves could deliver a completely different sensory experience modulable for different consumer groups or individuals. Similarly, microwave heating was utilized by Guo et al. (2021) to induce colour and aroma change in buckwheat dough. The authors prepared complex coacervate capsules as stimulus-response material containing, among other ingredients, encapsulated red pepper pigment and cinammaldehyde essential oil. After microwave heating at 200-W from 1 to 4 min, the cinnamaldehyde content increased more than two times, and the red index of the food structure increased, suggesting the possibility of combining 3D printing and microwave to promote desired quality changes over time. More recently, Ghazal et al. (2021) studied the spontaneous colour change during storage activated by different pH from 2 to 8. Specifically, the authors put in contact printed layers made a mixture of red cabbage juice (RCJ), vanillin powder (V) and potato starch (PS) and layers consisting of lemon, orange or apple juice gels at different pH. They observed a colour change in the next three hours after printing at room temperature and for six days of storage in refrigerated conditions. So, the colour change could be modelled by designing and printing a structure in which the area in contact of two layers at different pH, would be programmed/controlled. Under this idea, the authors printed 3D digital models designed to accelerate the colour change by modifying the exposure of RJC-V-PS with fruit gels at different pH. However, when 4D printing involves the change of the pH attention should be posed to the safety of food products. Indeed, increasing the pH during storage – as in the six days of storage reported by Ghazal et al. (2021) – could raise the risk for safety due to microbial growth. So, when external stimuli are used to activate the desired changes during the time, any potential drawbacks, mainly in terms of food safety, should be carefully considered. Similarly, the changes of different quality parameters under the activation of heating methods should also consider the potential degradation of nutritional compounds.

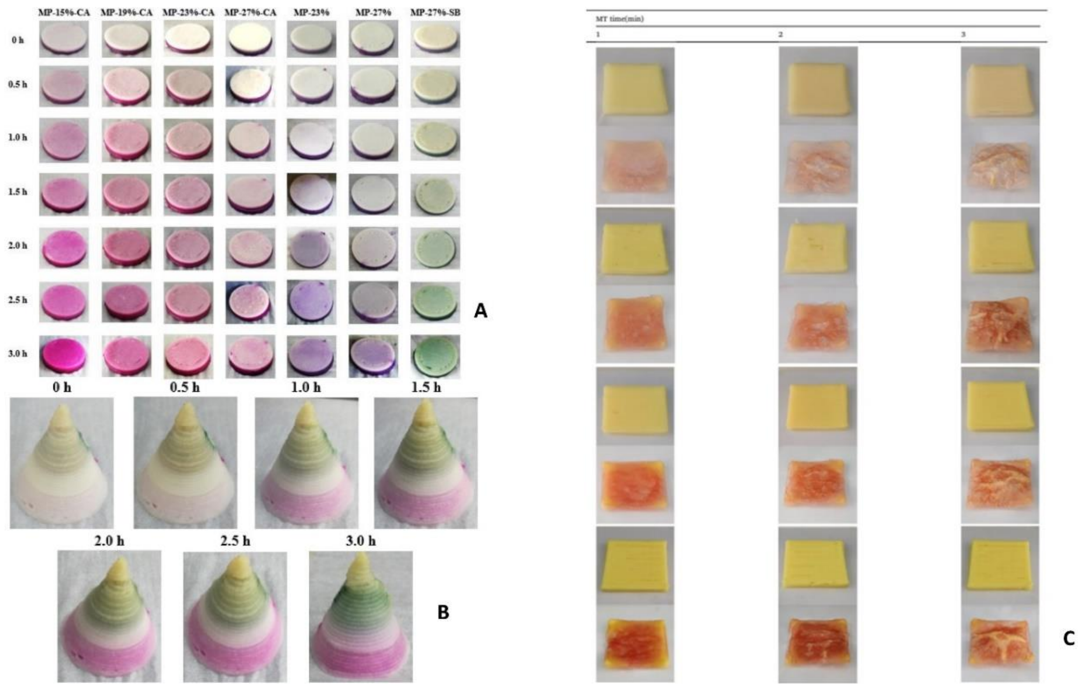


Fig. 2. 4D Food Printing: evolution of color over time of 3D printed structures under different external stimuli: (a) and (b) use pH as external stimuli (from He et al., 2020a); (c) use of microwave heating and alkaline conditions as external stimuli (from Chen et al., 2021a).

2.1. Approaches to obtain shape and dimension changes over time of 3D printed food.

Another interesting goal of 3DFP is to generate changes in the shape of 3D printed food. This option could largely expand the applications and the potential success of 3DFP on bars, restaurants, vending machines, lab of gastronomy, and industry, due to the great implications on the visual aspect, texture, capability to entrap sauce and seasonings, etc. In addition, we want to point out the possibility of producing flat food products activating the desired shape by external stimuli. Thus, 4DFP could be used to save volume during transportation or storage with great impact on the sustainability of food system. Dehydration, for instance, depending on the mechanical properties of the food materials, may drive shape change under appropriate conditions. Chen et al. (2021b) printed a pumpkin puree prepared at different moisture content and a mass fraction of alginate of 2% on a paper layer located on the bed of the printer. Interestingly, the differences in moisture content of food formulas, the mechanical properties and their behavior during dehydration were utilized to trigger the shape change. The double-layer products, indeed, has anisotropy in any plane's thickness direction. Accurately managing the layers, the infill pattern and printing thickness, the authors could activate the bending of the printed pumpkin puree obtaining diversity in shape, texture, and colour changes. The study has also reported that customers could have a better idea of the final shape, and reducing or prolonging the cooking time could offer innovative sensory experiences to the customers (Figure 3a). He et al. (2020b), used microwave heating as external stimuli and purple sweet potato as material stimuli to create innovative food structure. Microwave heating was employed for its fast heating and high drying capacity while the purple potato was preferred for its nutritional content. The researchers showed the high capability of the MW to activate the shape change of the samples particularly for the food formula prepared with higher amount of salt as a result of the increased dielectric properties. Also, the MW power significantly affected the rate of bending with times to reach the maximum bending from 42 to 12 min respectively when applying power of 2 and 8 W/g. (Figure 3b). Similarly, Liu et al. (2021) compared the effects of different drying methods - microwave (MD), infrared (ID), and air-drying (AR) - on the shape changes of 3D printed starch-based ink gel. In this case, although the MD could activate the shape change of the sample, it resulted in the lower bending angle while the highest shape change was observed for sample dried by air and infrared. It was hypothesized that the fast heat transfer occurred by the MW with the generation of internal vapour induced a great expansion of the structure that limited the shrinkage and the shape changes of the samples. Also, for air-drying the increase of temperature at 35°C, 50°C, and 65°C, produced the raise of the bending angle as a function of dehydration time. This interesting information paves the way for future applications based on better control of the shape changes of 3D printed food to different levels of application (industries, restaurants, etc.) integrating the chemical and physical properties of the food ink and its interrelation with the materials used as the bed for filament deposition (Figure 3c).

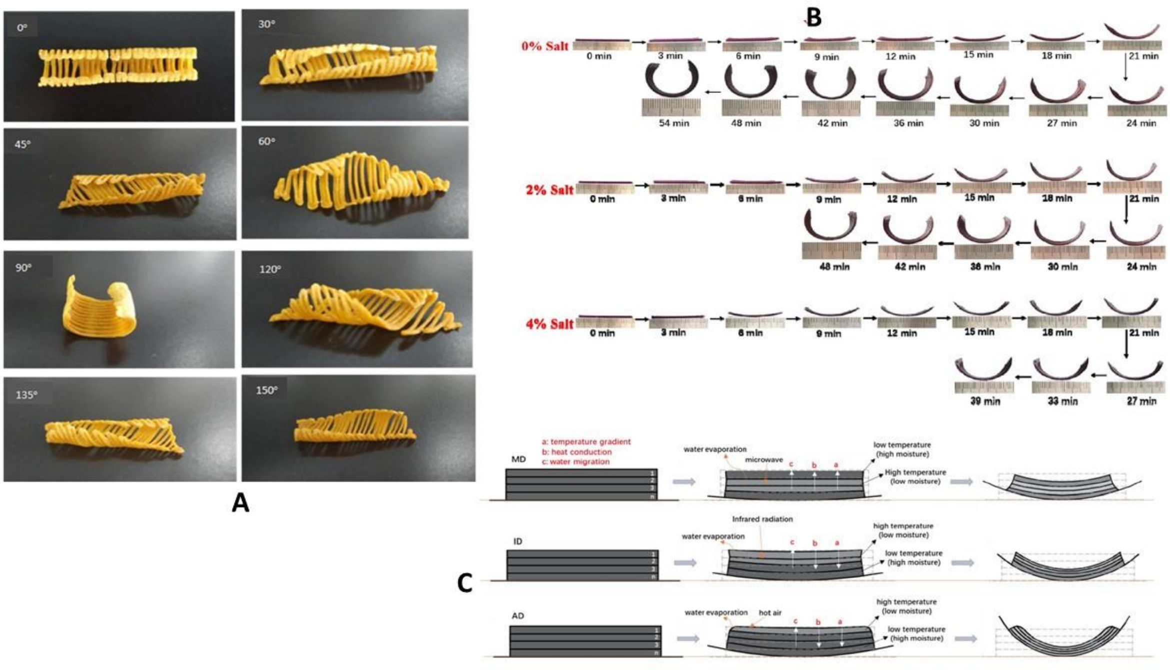


Fig. 3. 4D printed food examples with shape changes over time activated by different dehydration methods. (a) Complex deformation of the double-layer structure of pumpkin/paper (Chen et al., 2021b); (b) Bending deformation evolution of printed samples with different salt content (He et al., 2020b); (c) Shape change of samples triggered by different dehydration mechanisms (Liu et al., 2021).

2.2. *Activating aroma and nutritional changes over time of 3D printed food.*

As it is convenient for structure design and modelling, a controlled realization of chemical reactions that evolves in aroma compounds of 4D printed foods could be used to obtain a regular taste change or to prevent undesired changes. Phuhongsung et al. (2020) have studied the flavour changes of 3D printed dough containing soy protein isolate (SPI), k-carrageenan

(CAR) and vanilla flavour. After stimulating the changes by microwave heating the authors measured important changes of the aroma through electronic nose and GC-MS. They found that the physical changes in the sample were acceptable when the microwave heating power was increased to 80W, but the base of the sample was not smooth when the microwave heating power was increased to 110W. Therefore, they reported that the optimal heating conditions are 50-80 W. Foods were submitted to microwave heating for 20, 40 and 60 minutes to observe changes after microwave heating that are considered close to the common serving time for the consumers. The results concluded that the use of SPI gel with carrageenan and vanilla was the optimal condition for the 3D printer and, in addition, an innovative 4D printed product can be obtained by proper microwave heating (Figure 4). Another interesting application was reported by Guo et al. (2021) who incapsulated red pigment and cinnamaldehyde into a complex food formula subject to microwave heating. The authors observed the slow release of the cinnamaldehyde as a tool to get the aroma change of the 3D printed product. A further level of control could be obtained by affecting the heating coefficients as a result of modulated 3D printed structures such as infill level, the surface exposed to the heat, infill patterns, etc. So, although the published experiments have proved good opportunities for changing the aroma of 3D printed food as a function of time, further insight is needed to control the aroma changes over time and offering a great level of innovation and benefits to the food system. Finally, we want to shift our overview on the use of 4D food printing to activate the change of nutritional contents. Apart from a first example reported by Teng et al. (2020) in which seeds of edible plants were placed in a 3D printed edible nutrient matrix observing the seeds germinating and the plant growth as a function of time with modification of sensory and nutritional content, more recently Park et al. (2020) deposited an edible material consisting of carrot tissues embed in an alginate ink-gel. The authors printed a simple cube with an infill level of 60% and parallel filaments; after incubating the structure at 37°C for a maximum of 35 days they observed the cell growth making possible the nutritional changes over time. These data allow further considerations: by taking into account the respiration of the seeds or vegetable cells, this open for using 3D printing to generate programmable voids (i.e. porosity fraction) and interconnected channels controlling the rate of oxygen transfer as well as the diffusion of nutrients through the solid matrix (the edible ink) with an effect on the rate of cells growth. Nevertheless, we want to highlight the need for further experiments focused on the potential risks of the growth of pathogens for such interesting examples of 4D printed food for which the change of nutritional content occurs in a long period.

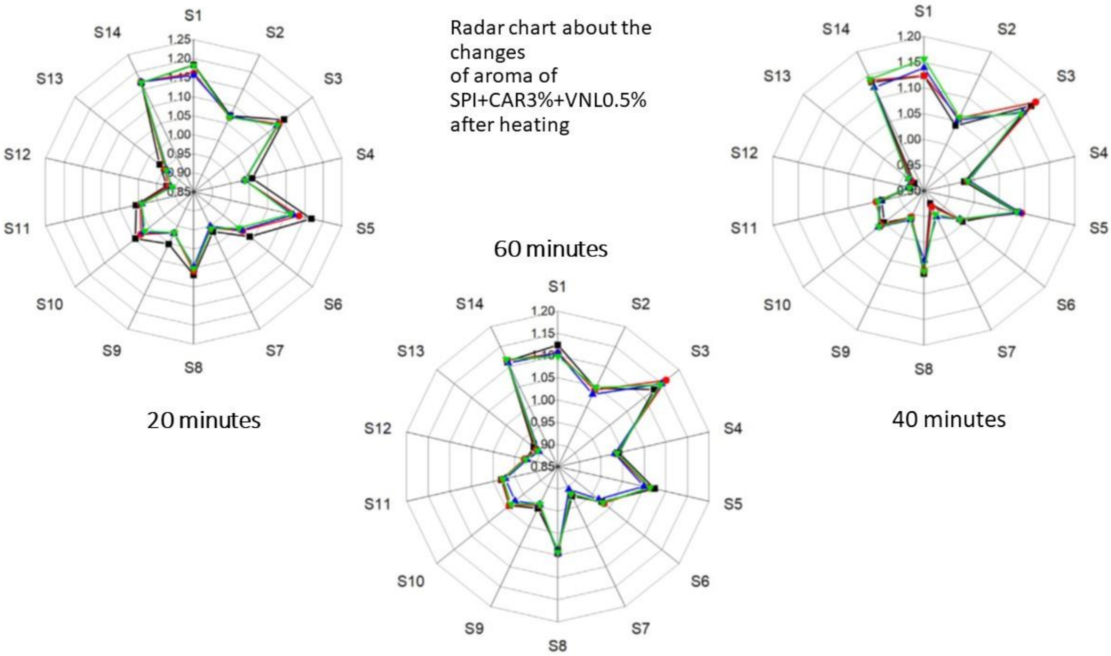


Fig. 4. Radar chart representing the changes of the aroma of 3D printed food as affected by the microwave heating time (from Phuhongsung et al., 2020). S1–S14: sensors of electronic nose.

3. *Programmable structure by 3D printing technology as an intervention to alter relevant sensorial and nutritional properties.*

The importance of the relationships among food structure and nutrients bioavailability (Yang et al., 2017), sensory perception (Aguayo-Mendozae et al., 2020; Patterson et al., 2021), eating rate, satiation and satiety (Bolhuis and Forde, 2020) as well as some physical properties (i.e. heating and cooling rate) have widely elucidated and proved. Currently, we have enough scientific evidence to prove that food texture, whether accurately designed, could create not only unparallel sensory perceptions but also to modulate the energy and nutritional intake of different consumers groups and to mitigate some specific issues such as mastication and swallowing troubles of the elderly. Shifting the aforementioned discoveries to the main aim of this contribution, 3D food printing could be used to model the shape, dimension and position in which voids are placed in food thus modifying the main morphological features of the solid elements. For instance, we know that by changing the nozzle diameter would be possible to reduce the size of the solid elements thus increasing the surface contact with enzymes responsible for the digestive phenomenon. All these possibilities are only attainable by 3D printing that control unique processing variables - i.e. nozzle diameter, infill percentage and the infill patterns, etc. - while this in unfeasible to modify with common manufacturing technologies. Some researchers published data on the effects of printing variables (Severini et al., 2016; Manthial et al., 2019) on the main texture properties of 3D food printed structure. Recently, Feng et al. (2020) used a mix of yam and potato to create innovative 3D printed food structures with a cylindrical external shell, infill level of 20%, 50% and 80% and by using six infill patterns. They highlighted that the air-fried products showed different hardness when using the same infill level but different infill patterns; interestingly, the order of the hardness was not constant through the infill levels with the hardest samples with parallel-structure and triangular-structure with low and medium infill level, respectively. However, the authors concluded showing robust and inverse linearity between the porosity fraction and the hardness of samples (Figure 5a). A significant impact of the infill level on the hardness of potato snacks has been observed also by Liu and Zhang (2021) (Figure 5b).

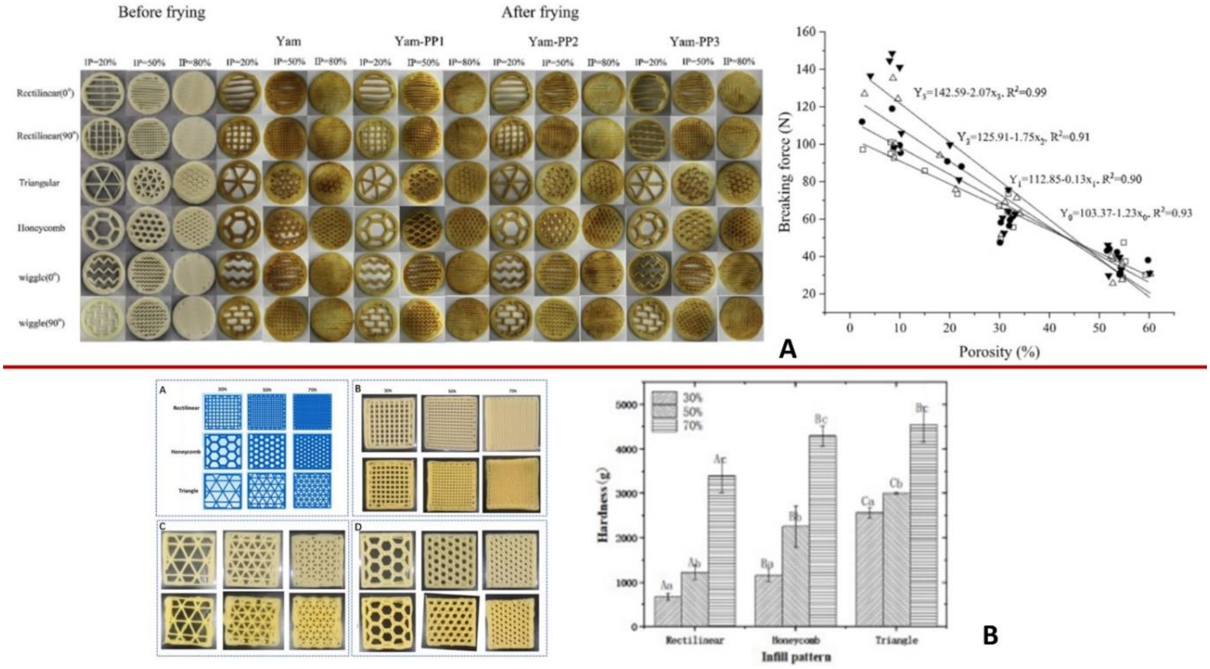


Fig. 5. (A) Changes in the breaking force of 3D printed structures obtained modulating the porosity fraction (20, 50 and 80%) (Feng et al., 2020); (B) Changes in Hardness of 3D printed structures with different infill pattern (rectilinear, honeycomb and triangle) (Liu and Zhang, 2021).

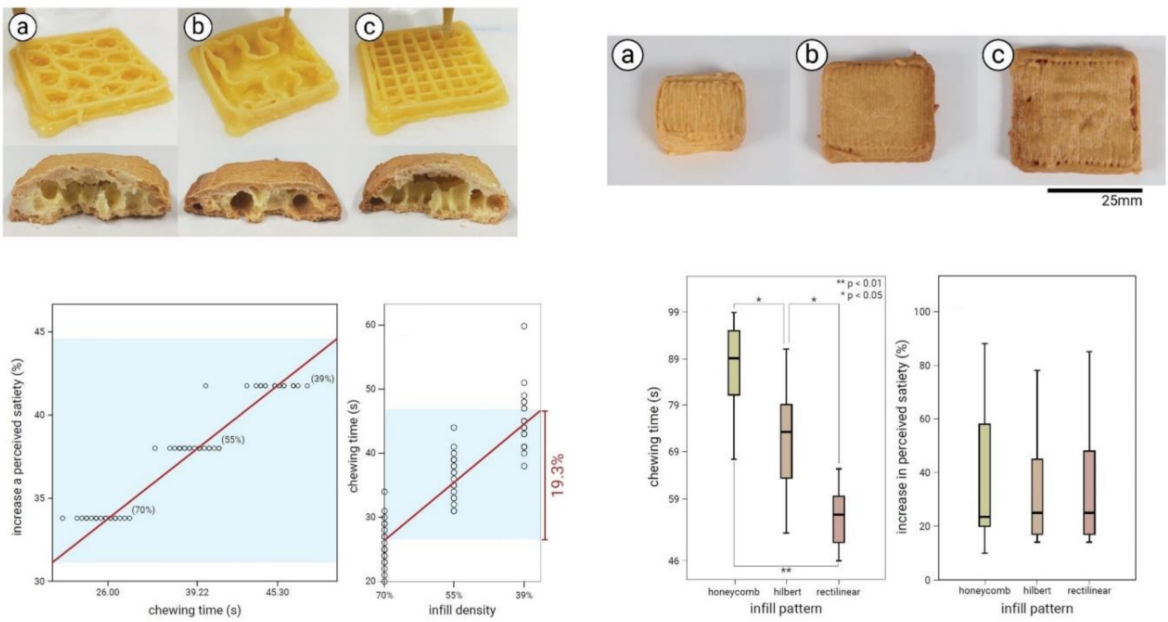


Fig. 6. Effect of internal morphology of 3D printed food on the chewing time and perceived satiety of different infill pattern (a, honeycomb; b, Hilbert; c, Rectilinear) (From Lin et al., 2020).

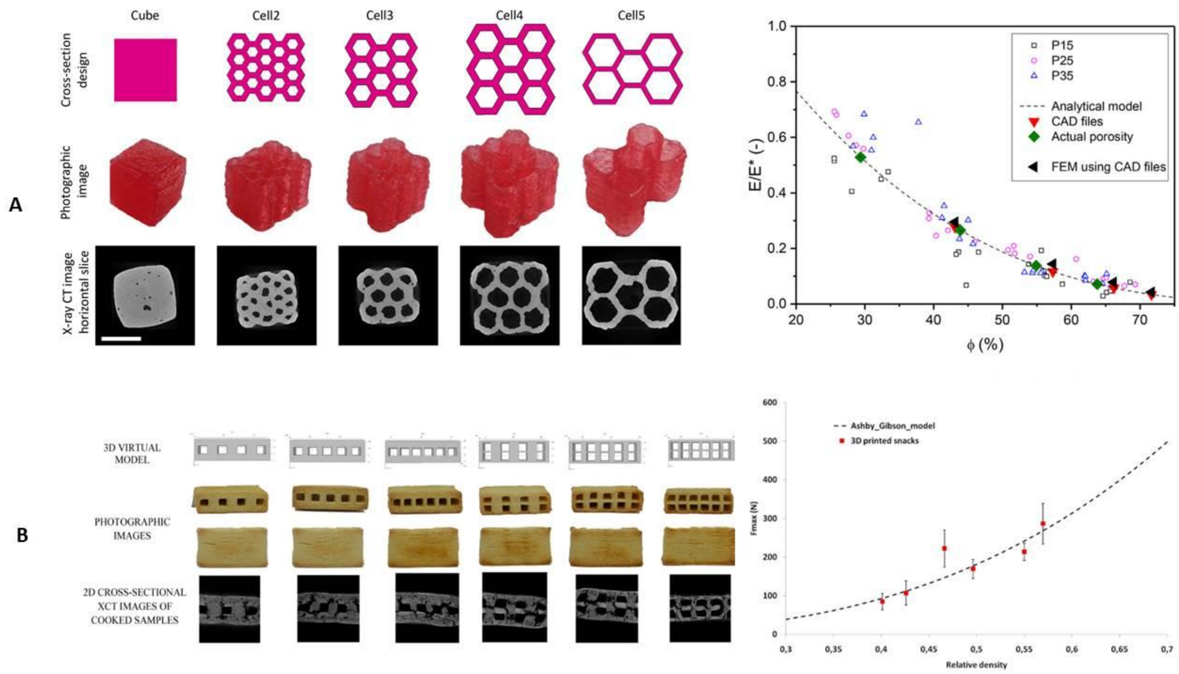


Fig. 7. Prediction of mechanical properties of 3D printed structures with different pores size, number and their location in 3D space. (a) edible-ink gel with honeycomb structure (Vancauwenberghe et al., 2018); (b) Cereal-based 3D printed food structures realized by changing the number the position of squared pores (Derossi et al., 2021c).

Finally, we want to highlight the intriguing idea of modulating the perception of satiety and satiation by complex 3D printed food with specific mechanical features. Lin et al. (2020) performed the first experiment devoted to satiety control by printing biscuits with different infill levels (39%, 55% and 70%) and infill patterns (honeycomb, Hilbert and rectilinear) keeping constant the number of calories. After measuring the chewing time and perceived satiety they proved that the higher was chewing time the greater was the perceived satiety and this occurred for the samples with increased infill levels while minor effects on the perceived satiety resulted by modifying the infill path although the chewing time decreased with the following order: honeycomb>Hilbert>rectilinear (Figure 6).

However, the reversing problem of obtaining a desired/customized texture based on the 3D CAD model shows a higher level of complexity, rarely faced in the food sector, but with great potential that will have to exploit. This goal is not completely new in the printing of non-food materials (Polyzos et al., 2020; Soufivand et al., 2020; Jindal, et al., 2020) but very few papers have been published on the food sector. In 2018, Vancauwenberghe and colleagues (2018), designed CAD models based on hexagonal honeycomb structures with structural diversities obtained by changing the number of cell size, number of cells and pore size and porosity fraction. They printed edible systems of low methoxylated pectin crosslinked with Ca2+ ions. Furthermore, by employing the finite element model (FEM) approach the authors were capable to estimate the relative young's modulus of 3D printed samples with excellent agreement with the analytical model. More precisely, by considering the structure reported in Figure 7a, the authors reported relative young's modulus of 0.28, 0.12, 0.06 and 0.03 when using analytical model and values of 0.29, 0.14, 0.09 and 0.04 by using FEM, respectively for the structures Cell2, Cell3, Cell4 and Cell5, corresponding to structures with increasing cell size dimension (from 2 to 5 mm) and/or porosity fraction. The same approach was used by Derossi et al. (2021c) who studied the capability to get a 3D food structure with a programmable texture. Also, they complicated the problem by using a cereal-based snack submitted to post-processing of cooking in an ordinary oven at 150°C. After cooking, a shifting of the samples toward higher fraction porosity was observed in comparison to the CAD model due to the generation of additional voids during material deposition as well as their expansion during baking. However, by using a generalized form of the Gibson and Ashby equation, the authors proved the capability of estimating the maximum force to break the 3D printed samples as a function of their relative density (Figure 7b).

Finally, we want to extend our discussion well beyond these first pioneering results and toward the futuristic challenges of converting the big body of data potentially at disposal (i.e. ingredients and their nutritional content, sensory perceptions, specific needs of macro-and micronutrients, lifestyle, habits, cultures, etc.) into a novel food structure. Two interesting examples come from the Japanese's company OPENMEALS. The CUBE is a project based on the idea of decomposing a general food formula into its own ingredients and using the obtained data to reshape it into a cube of 3 cm square by placing each ingredient in the desired location in the 3D space of the cube. This approach could open to obtain innovative 3D printed products grouped in three main classes: layered, wall and cell (Figure 8a). PIXEL FOOD printer is another pioneering approach based on the idea that when the generation of food with desired properties by a layer-by-layer method is too complicated, it would be possible to think similarly to a picture with high resolution. So, the 3D printer could generate small 3D cubes as voxels to be placed in a 3D space replicating with high fidelity the 3D CAD model. Figure 8b schematically represents this innovative idea for which by reducing the size of 3D printed food pixel it would be possible to obtain the best reproduction of the virtual model. Also, considering the use of pixels of edible gels with specific nutritional and sensory properties, the degree of freedom will exponentially improve.

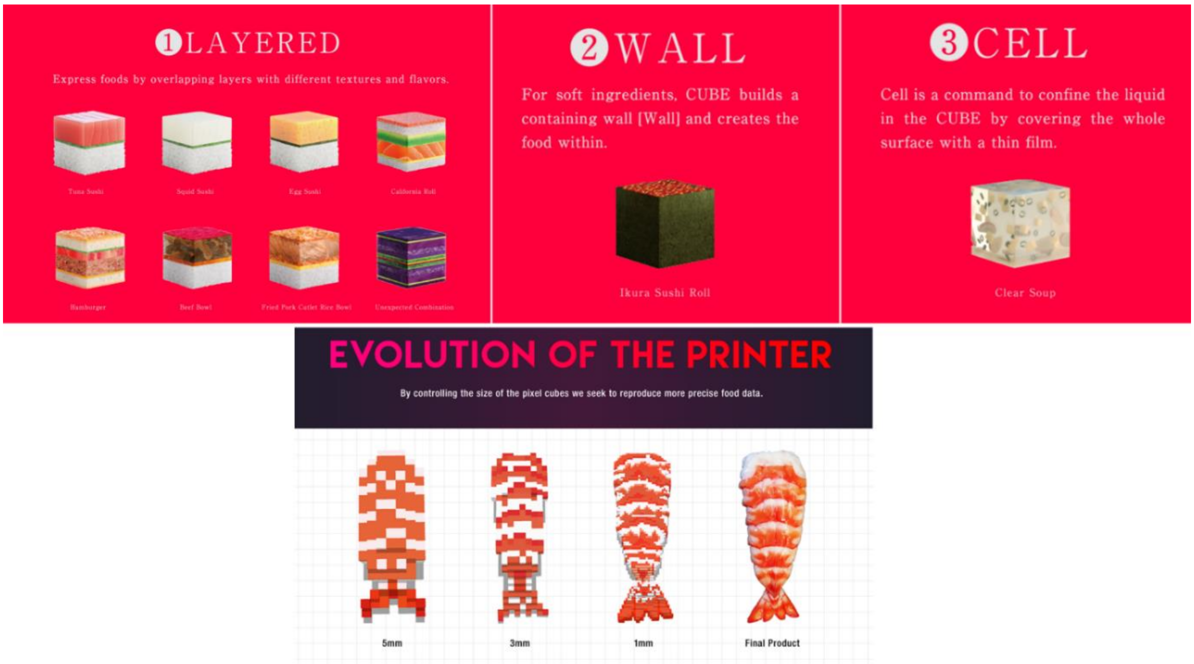


Fig. 8. Examples of innovative 3D food printing process (OpenMeals, 2021).

4. *Conclusion*

3D Food printing is currently under the spotlight of academia and the food industry. The capability to create complex geometries never realized before has paved the way for a great level of novelty in the food sector. However, the idea of using 3D printing is not limited to the creation of innovative and more fascinating shapes, but the researchers are working for the ambition of creating sensorial and nutritional customized food products, to reduce food waste and to bring 3D food printing at home, restaurants, and food industry. 3D printing is fast growing with the continuous evolution of the mode of application. 4D Food Printing is a recently born modality of application that - by exposing specific food materials to external stimuli - trigger changes of different properties as a function of time. Pioneering experiments shows that the changes of colour, aroma and shape of 3D food printed are feasible. This opens additional possibilities of creating food products with an extremely large range of quality parameters. This could be obtained by controlling the exposure time of the products to the external stimuli or by accurately designing some specific properties of the food formula – such as water mobility, shrinkage properties, mechanical features, etc.

A further recent innovation in 3D food printing is the creation of programmable food texture that means the process of designing a 3D CAD model and the printing of food formula that delivers desired mechanical properties. This interesting aspect not only opens for innovative food texture perception with expected high consumer's acceptance but also may help to mitigate relevant troubles of mastication and swallowing of elderly and modulate the perception of satiety and satiation through a controlled chewing time and mastication work. On these hot-points there is the need for multidisciplinary experiments and a higher level of collaboration between the few international research groups working on 3DFP but, at the moment, this seems one of the most promising technologies to activate the transition of the common method of thinking the relationship between the food industry and consumers in a more interrelated system of manufacturing and consumption driven by human-computer interactions.

References

1. Aguayo-Mendoza, M., Santagiuliana, M., Ong, X., Piqueras-Fiszman, B., Scholten, E., Stieger, M., 2020. How addition of peach gel particles to yoghurt affects oral behavior, sensory perception and liking of consumers differing in age. Food Res. Int., 134, 109213. https://doi.org/10.1016/j.foodres.2020.109213.
2. Bolhuis, D.P., Forde, C.G., 2020. Application of food texture to moderate oral processing behaviors and energy intake. Trends Foods Sci. Technol., 106, 445-456. https://doi.org/10.1016/j.tifs.2020.10.021.
3. Camarena, S., 2020. Artificial intelligence in the design of the transition to sustainable food systems. J. Clean. Prod., 271, 122574. DOI:10.1016/j.jclepro.2020.122574
4. Chen, C., Zhang, M., Guo, C., Chen, H., 2021a. 4D printing of lotus root powder gel: color change induced by microwave. Innov. Food Sci. Emerg. Technol., 68. https://doi.org/10.1016/j.ifset.2021.102605.
5. Chen, F., Zhang, M., Liu, Z., Bhandari, B., 2021b. 4D deformation based on double-layer structure of the pumpkin/paper. J. foosrt. 100168. https://doi.org/10.1016/j.foostr.2020.100168.
6. Cohen, D.L., Lipton, J.I., Cutler, M., Coulter, D., Vesco, A., Lipson, H. 2009. Hydrocolloid printing: a novel platform for customized food production. In: Solid Freeform Fabrication Symposium, Austin, TX, 807-818.
7. Derossi, A., Caporizzi, R., Paolillo, M., Oral, M.O., Severini, C. 2021a. Drawing the scientific landscape of 3D Food Printing. Maps and interpretation of the global information in the first 13 years of detailed experiments, from 2007 to 2020. Innov. Food Sci. Emerg. Technol., 70, 102689. https://doi.org/10.1016/j.ifset.2021.102689.
8. Derossi, A., Bhandari, B., van Bommel, K., Noort, M., Severini, C., 2021b. Could 3D food printing help to improve the food supply chain resilience against disruptions such as caused by pandemic crises? Int. J. Food Sci. Technol., 56, 9, 4338-4355. doi:10.1111/ijfs.15258.
9. Derossi, A., Caporizzi, R., Paolillo, M., Severini, C., 2021c. Programmable texture properties of cereal-based snack mediated by 3D printing technology. J. Food Eng., 289, 110160. https://doi.org/10.1016/j.jfoodeng.2020.110160.
10. Derossi, A., Azzollini, D., Albenzo, M., Severini, C., 2018a. On printability, quality and nutritional properties of 3D printed cereal based snacks enriched with edible insects. Food Res. Int., 106, 666-676. https://doi.org/10.1016/j.foodres.2018.01.034.
11. Derossi, A., Caporizzi, R., Azzollini, D., Severini, C., 2018b. Application of 3D printing for customized food. A case on the development of a fruit based snack for children. J. Food Engin., 220, 65-75. Doi: 0.1016/j.jfoodeng.2017.05.015
12. Eftimov, T., Korosec, P., Korousic Seljak, B., 2017. StandFood: standardization of foods using a semi-automatic system for classifying and describing foods according to Foodex2. Nutrients, 9 (6), 542. doi: 10.3390/nu9060542.
13. Feng, C., Zhang, M., Bhandari, B., Ye, Y., 2020. Use of potato processing by-product: effects on the 3D printing characteristics of the yam and the texture of air-fried yam snacks. LWT-Food Sci. Technol., 125, 109625. https://doi.org/10.1016/j.lwt.2020.109265.
14. Ghazal, A.F., Zhang, M., Bhandari, B., Chen, H., 2021. Investigation on spontaneous 4D changes in color and flavor of healthy 3D printed food materials over time in

response to external or internal pH stimulus. Food Res. Int., 142, 110215. DOI: 10.1016/j.foodres.2021.110215
15. Guo, C., Zhang, M., Bhandari, B., 2019. A comparative study between syringe-based and screw-based 3D food printers by computational simulation. Comput. Electron. Agric., 162, 397-404. https://doi.org/10.1016/j.compag.2019.04.032.
16. Guo, C., Zhang, M., Devahastin, S., 2021. Color/aroma changes of 3D-printed buckwheat dough with yellow flesh peach as triggered by microwave heating of gelatin-gum Arabic complex coacervates. Food hydrocoll., 112, 106358. https://doi.org/10.1016/j.foodhyd.2020.106358.
17. He, C., Zhang, M., Guo, 2020a. 4D printing of mashed potato/purple sweet potato puree with spontaneous color change. Innov. Food Sci. Emerg. Technol., 59. https://doi.org/10.1016/j.ifset.2019.102250.
18. He, C., Zhang, M., Devahastin, S., 2020b. Investigation on spontaneous shape change of 4D printed starch-based purees from purple sweet potatoes as induced by microwave dehydration. ACS Appl. Mater. Interfaces, 12, 37896-37905. https://dx.doi.org/10.1021/acsami.0c10899.
19. Jindal, P., Worcester, F., Siena, F.L., Forbes, C., Juneja, M., Breedon, P., 2020. Mechanical behaviour of 3d printed vs thermoformed clear dental aligner materials under non-linear compressive loading using FEM. J. Mech. Behav. Biomed. Mater., 112, 104045. https://doi.org/10.1016/j.jmbbm.2020.104045.
20. Lin, Y.J., Punpongsanon, P., Wen, X., Iwai, D., Sato, K., Obrist, M., Mueller, S., 2020. FoodFab: creating food perception illusions using food 3d printing. CHI'20, April 25-30, 2020, Honolulu, HI, USA. https://doi.org/10.1145/3313831.3376421.
21. Liu, Z., He, C., Guo, C., Chen, F., Bhandari, B., Zhang, M., 2021. Dehydration-triggered shape transformation of 4D printed edible gel structure affected by material property and heating mechanism. Food Hydrocoll., 115, 106608. https://doi.org/10.1016/j.foodhyd.2021.106608.
22. Liu, Z., Zhang, M. 2021. Texture properties of microwave post-processed 3D printed potato snack with different ingredients and infill structure, Future Foods. 3, 100017. https://doi.org/10.1016/j.fufo.2021.100017.
23. Liu, Z., Bhandari, B., Zhang, M., 2020. Incorporation of probiotics (*Bifidobacterium animalis* subsp. *Lactis*) into 3D printed mashed potatoes: Effects of variables on the viability. Food Res. Int., 128, 108795. https://doi.org/10.1016/j.foodres.2019.108795
24. Mantihal, S., Prakash, S., Bhandari, B., 2019. Texture-modified 3D printed dark chocolate: sensory evaluation and consumer perception study. J. Texture Stud., 50(5), 386–399. https://doi.org/10.1111/jtxs.12472
25. Nijdam, J.J., Agarwal, D., Schon, B.S., 2021. An experimental assessment of filament-extrusion models used in slicer software for 3D food-printing applications. J. Food Eng., 110711. https://doi.org/10.1016/j.jfoodeng.2021.110711.
26. Openmeals. https://www.open-meals.com/. Accessed 12/06/2021
27. Paolillo, M., Derossi, A., van Bommel, K., Noort, M., Severini, C., 2021. Rheological properties, dispensing force and printing fidelity of starchy-gels modulated by concentration, temperature and resting time. Food Hydrocoll., 117, 106703. https://doi.org/10.1016/j.foodhyd.2021.106703.

28. Park, S.M., Kim, H.W., Park, H.J., 2020. Callus-based 3D printing for food exemplified with carrot tissues and its potential for innovative food production. J. Food Eng., 271, 109781. https://doi.org/10.1016/j.jfoodeng.2019.109781.
29. Patterson, N.M., Zhong, Y., James, B.J., Gant, N., Hautus, M.J., 2021. Effect of basic structural variation, aimed at increasing perceivable textures in model foods, on the perception of textural complexity. Food Qual. Prefer., 91, 104196. https://doi.org/10.1016/j.foodqual.2021.104196.
30. Phuhongsung, P., Zhang, M., Bhandari, B., 2020a. 4D printing of products based on soy protein isolate via microwave heating for flavour development. Food Res. Int., 137, 109605. https://doi.org/10.1016/j.foodres.2020.109605.
31. Pini, A., Hayes, J., Upton, C., Corcoran, M., 2019. All inspired recipes: designing computationally creative food combos. CHI, Glasow, Scotland, UK.
32. Polyzos, E., Katalagarianakis, A., Polyzos, D., Van Hemelrijck, D., Pyl, L., 2020. A multi-scale analytical methodology for the prediction of mechanical properties of 3d-printed materials with continuous fibres. Addit. Manuf., 36, 101394. https://doi.org/10.1016/j.addma.2020.101394.
33. Pulatsu, E., Su, J., Lin, J., Lin, M., 2020. Factors affecting 3D printing and post-processing capacity of cookie dough. Innov. Food Sci. Emerg. Technol., 61, 102316. https://doi.org/10.1016/j.ifset.2020.102316
34. Pulatsu, E., Lin, M., 2021. A review on customizing edible food materials into 3D printable inks: Approaches and strategies. Trends Food Sci. Technol., 107, 68-77. https://doi.org/10.1016/j.tifs.2020.11.023.
35. Severini, C., Derossi, A., Azzolini, D., 2016. Variables affecting the printability of foods: preliminary tests on cereal-based products. Innov. Food Sci. Emerg. Technol., 38, 281-291. https://doi.org/10.1016/j.ifset.2016.10.001.
36. Soufivand A.A., Abolfathi, N., Hashemi, S.A., Lee, S.J., 2020. Prediction of mechanical behaviour of 3d bioprinted tissue-engineered scaffolds using finite element method (FEM) analysis. Addit. Manuf., 33, 101181. https://doi.org/10.1016/j.addma.2020.101181.
37. Spence, C., 2021. Explaining diurnal patterns of food consumption. Food Qual. Prefer. 104198. https://doi.org/10.1016/j.foodqual.2021.104198
38. Sun, J., Zhou, W., Huang, D., Fuh, J.Y., Hong, G.S., 2015. An overview of 3D printing technologies for food fabrication. Food Bioproc. Tech., 8 (8), 1605-1615. http://dx.doi.org/10.1016/j.jfoodeng.2017.02.028
39. Teng, X., Zhang, M., Mujumdar, A.S., 2021. 4D printing: recent advantages and proposals in the food sector. Trends Food Sci. Technol., 110, 349-363. https://doi.org/10.1016/j.tifs.2021.01.076.
40. Tian, H., Wang, K., Lan, H., Wang, Y., Hu, Z., Zhao, L., 2021. Effect of hybrid gelator systems of beeswax-carrageenan-xanthan on rheological properties and printability of litchi inks for 3D food printing. Food Hydrocoll., 113, 106482. https://doi.org/10.1016/j.foodhyd.2020.106482.
41. Tomasevic, I., Putnik, P., Valjak, F., Pavlic, B., Sojic, B., Markovinovic, A.B., Kovacevic, D.B., 2021. 3D printing as novel tool for fruit-based functional food production. Curr. Opin. Food Sci., 41, 138-145. https://doi.org/10.1016/j.cofs.2021.03.015.

42. Vancauwenberghe, V., Delele, M.A., Vanbiervliet, J., Aregawi, W., Verboven, P., Lammertyn, J., Nicolai, B., 2018. Model-based design and validation of food texture of 3D printed pectin-based food simulants. J. Food Eng., 231, 72-82. https://doi.org/10.1016/j.jfoodeng.2018.03.010.
43. Wang, L., Zhang, M., Bhandari, B., Yang, C., 2018. Investigation on fish surimi gel as promising food material for 3D printing. J. Food Eng., 220, 101-108. https://doi.org/10.1016/j.jfoodeng.2017.02.029
44. Xu, L., Gu, L., Su, Y., Chang, C., Wang, J., Dong, S., Liu, Y., Yang, Y., Li, J., 2020. Impact of thermal treatment on the rheological, microstructural, protein structures and extrusion 3D printing characteristics of egg yolk. Food Hydrocol., 100, 105399, https://doi.org/10.1016/j.foodhyd.2019.105399.
45. Yang, Y., Xiao, H., McClements, D.J., 2017. Impact of lipid phase on the bioavailability of vitamin e in emulsion-based delivery systems: relative importance of bioaccessibility, absorption, and transformation. J. Agric. Food Chem., 65, 19, 3946-3955. https://doi.org/10.1021/acs.jafc.7b00955.
46. Yoha, K.S., Anukiruthika, T., Anila, W., Moses, J.A., Anandharamakrishnan, C., 2021. 3D printing of encapsulated probiotics: Effect of different post-processing methods on the stability of *Lactiplantibacillus plantarum* (NCIM 2083) under static in vitro digestion conditions and during storage, LWT, 146, 111461. https://doi.org/10.1016/j.lwt.2021.111461.
47. Zhang, L., Lou, Y., Schutyser M.A.I., 2018. 3D printing of cereal-based food structures containing probiotics. Food Struct., 18,14-21. https://doi.org/10.1016/j.foostr.2018.10.002.
48. Zhao, Z., Wang, Q., Yan, B., Gao, W., Jiao, X., Huang, J., Zhao, J., Zhang, H., Chen, W., Fan, D., 2021. Synergistic effect of microwave 3D print and transglutaminase on the self-gelation of surimi during printing. Innov. Food Sci. Emerg. Technol., 67, 102546. https://doi.org/10.1016/j.ifset.2020.102546.

Chapter 3 Analyzing the effects of 3D printing process *per se* on the microstructure and mechanical properties of cereal food products

Authors: Derossi, A., Caporizzi*, R., Oral, M.O., Severini, C.

Department of Science of Agriculture, Food and Environment, University of Foggia, Italy

Corresponding author: rossella.caporizzi@unifg.it

Abstract

3D Food Printing has been gaining interest to create food with personalized properties. To customize the texture, it is necessary to explore whether and how 3D printing process *per se* has consequences on the mechanical features. As test case, a cubical cereal-based structure was manufactured by traditional processing and 3D printing. Microstructure properties and mechanical attributes were analyzed. Here we show that 3D printing clearly affects the microstructure generating bigger pores, less in number and like-round in shape. Also, we have observed that the positions of the pores are greatly driven by the printing movements. These features significantly affect the mechanical properties of 3D samples showing high hardness, chewiness, and cohesiveness. The obtained data have been linked and interpreted based on three main key points: 1. the printing path; 2. the imbalance between speed printing and extrusion rate; 3. the compression of the food formula in the extrusion system. These findings should be considered for creating food with innovative texture perceptions.

Industrial relevance: The creation of 3D food printing with programmed texture has the ambitions of getting personalized food improving industry competitiveness by novel texture perceptions and helping to mitigate swallowing or mastication problems vulnerable peoples. Here we show that 3D Printing process *per se* – intrinsically – modifies the morphology and the distribution of pores in 3D structure thereby modifying the texture of cereal-based snacks. All these because pores generation is not 'randomly' distributed as for traditional manufacturing methods but driven by printing movements previously planned during the slicing of digital model. With the aim to get personalized food texture the intimate relationship between the movements of printing and the food texture shall take into account by interested industry and producers.

Keywords: human-food-computer interactions; 3D printing pat; pores morphology; texture properties; food printing imbalance; programmable structure.

1. Introduction

3D Printing (3DP) is a technology belonging in the wider processes of Additive Manufacturing (AM) which, through a layer-by-layer deposition, create 3D objects from three-dimensional digital model. To date, 3D printing is everywhere and it is much more than the initial applications of fast prototyping (Kapetaniou et al., 2017). 3DP provides several benefits such as hyper flexibility, risk reduction for new products innovation, cost reduction, on-demands production, and reduction of waste and environmental impact. Further, 3DP involves end-users in the process of manufacturing with positive effects on the personalization of goods and decentralization of production. The users, works depositing layers of a wide range of materials from the most common thermoplastic materials (PLA, BAS), resins (Hosny et al., 2018), glass (Wu et al., 2018; MIT, 2020a), concretes (Vantyghem et al., 2020) and hybrid-living materials (MIT, 2020b). Also, 3D printing is becoming attractive for bioregenerative medicine (Gibney et al., 2017), pharmaceutical (Karavasili et al., 2020), surgical planning (Hosny et al., 2018), architecture industry (Chan et al., 2020), soft sensors (Emon et al., 2019), automotive (Nichols, 2019), etc. In addition, the application of 3DP in food sector is of great interest.

3D Food Printing (3DFP) has the potential of creating 3D edible structures with hundred degrees of freedom enabling the realization of food with shapes and dimensions; furthermore, 3DFP has the ambition of creating food designed to address specific nutritional or functional requirements or to satisfy specific consumers desires making reliable the idea of personalized food manufacturing (Lin et al., 2020). Lupton (2017), analyzing the online news reporting the potential interests in the application of 3D Food Printing, found five main promissory themes such as futuristic, creative, healthy, efficient and sustainable. Recently, some companies have introduced 3D printed products on the market such as pasta (blurhapsody, 2020), or the engagement of consumers for the creation of personalized biscuits (Maya, 2020). 3D Food printing (3DFP) is a relatively young field of research whit no more than 100 scientific papers from 2011 to now (Piyush et al., 2020). Such papers have been mainly focused on the study of several food inks to create 3D food printed structures such as chocolates (Lanaro et al., 2017; Mantihal et al., 2017), lemon gels (Yang et al., 2018), mashed potato (Liu et al., 2018a), vegetable and fruit blends (Severini et al., 2018a), cereal-based food formula (Severini et al., 2016; Severini et al., 2018b; Pulatsu et al., 2020), surimi paste (Wang et al., 2018), eggs yolk (Xu et al.,2020) mixed with rice flour (Anukiruthika et al., 2020), hydrocolloids (Chen et al., 2019; Gholamipour-Shirazi et al., 2019), cheeses (Le Tohic, 2018), yam powder (Feng et al., 2020), vegetables powder (Lee et al., 2019), ground meat (Dick et al., 2019). For their use, such edible materials should be printable. This term includes that food-inks should have shear thinning behaviour (Gholamipour-Shirazi et al., 2019) (the ability to be extruded from the nozzle) and capable to maintain the shape and dimension of the printed structure which include properties such as adhesiveness, viscosity and consistency. To help to mitigate these problems several researchers have studied the relationship between the rheological properties of food-inks, also by using different hydrocolloids or other structuring agents, and the stability of 3D printed food structures (Hamilton et al., 2017; Yang et al., 2018; Liu et al., 2018a; Dianez et al., 2019). However, instead of achieving food paste printability by a series of trial-and-error approach, Zhu et al., (2019) showed that it would be possible to predict the structural stability of printed food on the basis of aforementioned rheological measurements of such food pastes.

However, the capacity to replicate the 3D digital model with high accuracy – the printing fidelity - is also strongly affected by several printing variables such as print speed (Derossi et al., 2020a; Yang et al., 2018), nozzle size (Yang et al., 2018), layer height (Yang et al., 2018; Zhu et al., 2019), infill density (Liu et al., 2018b; Severini et al., 2018b; Dick et al., 2019), extrusion rate (Hamilton et al., 2017; Vanauwemberghe et al., 2017a), flow (Derossi et al., 2018) as well as by some non-printing variables such as travel speed, retraction speed and retraction distance (Derossi et al., 2020a). All such variables affect, with different weights, the balance between the extrusion rate and printing movements increasing/reducing the dimensional and shape discrepancies between printed food structure and the designed digital model. For instance, Wang et al. (2020) and Yang et al. (2018) proposed the computation of a critical layer height value (h_c) to keep a good equilibrium between extrusion rate and print speed. Derossi et al. (2018) who studied the effects of some variables on the morphological properties of cubic-shape banana-snack, highlighted the importance of the parameter flow that is directly related with the side length, the total weight and volume of the snacks.

Despite the broad numbers of published papers, current research lacks the understanding of the effects of 3D food printing process on structural and microstructure properties of end products. This could better unleash the global potential of 3D printing designing the internal structure for the creation of customized texture for consumer's groups with specific requirements, such as for elderly people with mastication or swallowing problems, or for novel 3D structures able offering innovative sensorial perceptions. By the way this is a theme not new in the field of 3D printing of non-food materials. For instance, the incorporation of 'life' in 3D printed structures, in other words 4D printing process, often is based on the creation of unique 3D microstructure which under different stimuli swell or bends in a desired modality creating an object with specific mechanical properties (Spiegel et al., 2020). Also, the capability to replicate by 3D printing the primary cell alignment in native tissue (Lee and Yeong, 2020) is of essential importance in engineering tissues because it is the only way to replicate mechanical and other functionalities of human organs (Ho, 2009).

However, in food sector the texture of 3D printed food has been studied on the basis of the food-inks composition (Lille et al., 2018; Vancauwenberghe et al. 2019; Chen et al., 2020) and as a function of infill level or the infill patter (Severini et al., 2016; Mantihal et al., 2017; Liu et al., 2018b; Feng et al., 2020; Vancauwenberghe et al., 2019). Feng et al. (2020), by working with air-fried products showed that, for sample at low infill of 20%, showed that the hardness follow the order parallel-structure > cross-structures > complex structures. More recently, Park et al. (2020), showed that by using cells lines of carrot callus tissue embedded in 3D printed alginate hydrogel it was possible to obtain different texture properties as a function of initial cell concentration and culture time. Also, Derossi et al. (2020b) studied the possibility to modulate the texture of cereal-based snack by a controlled generation of voids by using 3D printing technology.

However, whether the process of deposition of food ink is able to significantly affect the structure of food independently from the setting of printing variables, is a question that has not been elucidated yet. The 3D printing process is controlled by the G-code that finds the most efficient paths to replicate the 3D virtual model. This result in the cross-sectional deposition of food materials, sometime with very intricate movements, which is potentially able to affect the

microstructure and texture of 3D printed structures *per se,* independently from the choice of the infill parameters or of any other printing condition.
This has been widely experimented in no-food sector proving the effect of 3D printing on material microstructure's properties. As reported by Carlton et al (2016) undesired porosity are very common in additive manufacturing with a negative impact on mechanical properties on 3D printed objects (Carlton et al., 2016; DebRoy et al., 2018)
Li et al., (2018) by working with selective laser melting (SLM) of a equiatomic CoCrFeMnNi high-entropy alloy powder clearly showed the creation of irregular shaped and randomly distributed pores generation in a simple cylindrical sample. Also the authors showed the changes of some microstructure feature in line with the increase of the applied volumetric density energy which, in turn affected the mechanical properties of samples (yield stress, tensile strength, elongation to fracture).

For our knowledge, this topic has been undervalued with very few published data. For instance, Le Tohic et al. (2018) reported that printed cheese was less hard in comparison with traditional samples. Liu et al., (2018b) by studying the texture of 3D printed objects from mashed potatoes reported hardness, gumminess and firmness of 3D printed structure at 100% infill significantly lower than the cast samples. However, further experiments would of great importance to acquire new structural information toward a programmable food texture.

In this paper we addressed this topic by analyzing the effects of 3D printing process *per se* on the microstructure and mechanical properties of cereal-based snacks. To do this, we compared four food formulas obtained by modifying the type of fat and the blend of wheat/rice flours which were used to build a cubical-shaped snack by 3D printing and traditional process. On these we analyzed the fidelity of printing, microstructure properties and the mechanical features.

2. Material and Methods

2.1 Materials and food formula preparation

Wheat flour type 00 (Barilla, Italy) having chemical properties reported in the Italian legislation (2001) for wheat flour and pasta, Rice flour (Le Farine Magiche, Lo Conte, Italy), olive oil (Dentamaro, Italy), milk butter (Granarolo, Italy), leavening powder (Paneangeli, Italy) and water (Lilia, Italy) were purchased locally. A total number of 4 food formulas were prepared by modulating the flours and the fat at two different weight fractions (%) as reported in Table 1. The amount of water and fats (olive oil and butter) was defined based on preliminary experiments (data not shown) aiming to obtain batters with appropriated features of printability, the capability to be extruded through the nozzle and the potential ability to keep the shape of 3D printed structure (Gholamipour, Shirazi, Norton, & Mills, 2019). Also, the weight fraction of wheat and rice flour employed in the food formulas was deliberately chosen with the aim to tests the potential effect of different food formula on the microstructure and mechanical properties of the 3D printed snacks compared against the hand-made snacks.

Table 1 – Composition of the printable dough used during experiments

Label	Wheat flour (%)	Rice flour (%)	Olive oil (%)	Milk butter (%)	Water (%)	Leavening powder (%)
WO	*56*	-	*10*	-	*32*	*2*
RO	*28*	*28*	*10*	-	*32*	*2*
WB	*56*	-	-	*10*	*32*	*2*
RB	*28*	*28*	-	*10*	*32*	*2*

The ingredients were mixed for 3 min at low speed (22 rpm) in a planetary kneader (mod. Cooking chef, Kenwood Ltd, UK). Then the food formulas were covered with a plastic wrap and left to rest for 60 minutes before experimental tests.

2.2 3D Printing experiments and traditional process

A 3D printer mod. Delta 2040 (Wasp project, Italy) equipped with a screw-extrusion system for clay was used for printing experiments. The food formula was loaded in the piston chamber connected by a plastic tube to the extruder. Marlin firmware was used to control printer movements (Marlin, 2020). A pressure of 1 bar was applied to assure the continuous filling of the batter to the extruder. A simple digital model of cubical shape with a dimension 20*20*15 mm was designed by using the software Tinkercad (Autodesk, Inc) while the slicing was carried out by CURA mod. 3.6.0 (Ultimaker, B.V., The Netherlands). Three layers of skirt have been designed to trigger the filaments avoiding problems during 3D construction (Figure 1).

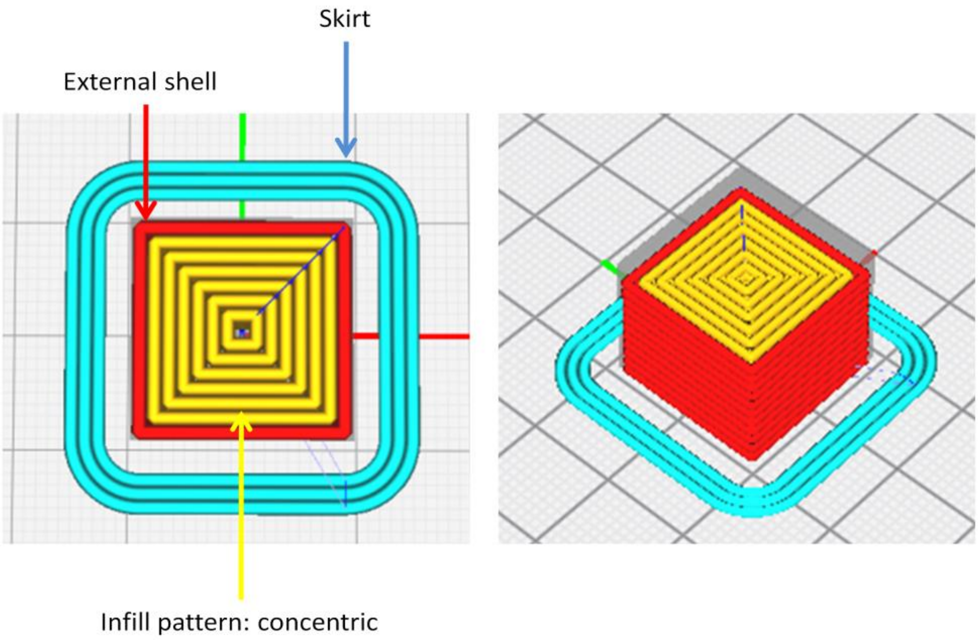


Fig. 1. 3D digital model used for 3D printing experiments.

The obtained G-codes were used to control the movements of the printer during layer-by-layer process. The main printing conditions are reported in Table 2. These conditions were defined on the basis of the results of previous papers (Severini et al., 2018b; Derossi et al., 2020a; Derossi et al., 2020b) and other additional further preliminary experiments aimed to find the best balance between extrusion rate and printing movements. This digital model allowed creating raw structures with a weight of 5±0.2 g for all food formulas. The 3D printed structures were left to equilibrate at room temperature for 10 minutes before baking reaching an average temperature of 25±3 °C. The cooking was performed by placing the 3D printed structure in a squared metal mold as reported below.

Table 2 – Main printing conditions used for 3D printing experiments

Basic settings	**Value**
Layer height	1.0 mm
Initial Layer height	0.3 mm
External shell	1
Nozzle size	1.2 mm
Infill density	100%
Infill pattern	Concentric
Print speed	30 mm/s
Travel speed	100 mm/s
Retraction distance	0.5 mm
Retraction speed	50 mm/s
Piston pressure	1 bar
Total time for printing	≈ 12 min

To compare 3D printing against traditional process, a second series of samples was prepared by employing a traditional hand-made process. In this case, after resting, the batters were cut into small pieces of 5±0.2 g then modeled by hand in like-spherical shape and finally placed in a square metal mold with length of 30*30 mm and 30 mm of height. Here the batters were left to rest for 10 minutes of relaxation enabling it to adhere to mold. Then the samples were cooked as reported below. For simplicity, in the next sections of the paper, 3D printed samples will be labeled as '3D' while the hand-made samples will be labeled as 'H'.

2.3 Cooking process of cereal-based snack

The cooking was performed in an ordinary oven at 150°C by varying the baking time for each food formula within a range of 16 min and 18 min, as defined by preliminary experiments (not shown) aiming to get baked samples at the same moisture content. The baking was performed by placing the samples on a metal tray at half of the internal height of the oven and 10 min after reaching 150°C assuring a constant temperature inside the oven.

After baking the sample were cooled at room temperature till 2 h before used for analyses.

2.4 Dimensional discrepancies of printed samples

The main dimensions of 3D printed samples were measured by using ImageJ ver. 1.53 (NIH Image) on photographic images acquired by using a cellular photo camera (iPhone, Apple brand) with a resolution of 12 Mega Pixels, f/1.8, 1/3", PDAF, OIS. For each samples two images were acquired on top view and lateral view. From the top view the length of the samples was measured in both x and y directions. From the lateral view, it was measured the height. At least 10 replicates were performed for each dimension.

2.5 Moisture content of baked samples.

To measure moisture content at different layers of the samples, the upper, the lower and the two laterals' crusts were removed from the samples for thickness of approximately 0.2 mm. Then the moisture content was measured by as described in AACC method 44-15.02 (AACC, 2010). The moisture content of both the crust and internal crumb was performed by analyzing at least 10 repetitions for sample.

2.6 X-ray images scanning and microstructure characterization

2D X-ray cross-sectional images of the traditional and 3D printed samples were obtained by using a Skyscan 1174 desktop CT system (Bruker, Kontich, Belgium). The following conditions were used for the microtomography images acquisition: resolution of 28.5 and 11.7 um; step angle of 0.3°; exposure time of 1200 ms; 50 kV; averaging frame of 3; total scanning time of 36 min. Image reconstruction was performed by using Nrecon 1.6.2.0 software (Bruker, Kontich, Belgium). First, the entire stack of the images was cropped to get a volume of interest (VOI) of 420^3 voxel by using the CTAn 1.12.0.0 (Bruker microCT, Belgium) presenting more than the 70% of the total volume of the samples. Then, segmentation process was performed with the aim to separate void phase (black pixel) with solid phase (white pixel). First, the gray-scale histogram was studied to preliminary delineate general lower and upper threshold limits. Next, these values were used as input values for binarization performed by using a global approach as tool ImageJ ver 1.52v (National Institute of Health, USA). Finally, the main microstructure descriptors such as aspect ratio (AR), circularity (CIRC), roundness (ROUND), the area fraction of each pores, porosity fraction (%) were evaluated on a total of 2D images of N=420 for each sample. More specifically, the following equation have been used to compute morphological properties of internal voids: $AR = \frac{Major\ Axis}{Minor\ Axis}$; $CIRC = 4\pi * \frac{[Area]}{[Perimeter]^2}$; $ROUND = 4 * \frac{[Area]}{\pi*[Major\ Axis]^2}$. All computations have been performed by using ImageJ employing the tools 'analyze particles'.

2.7 Mechanical properties determination

To determine the main mechanical properties of the samples the top, bottom and lateral crusts were removed by using a mini electric grinder rotary tool hand (Dremer, Multipro) till a maximum thickness of 2 mm. Then the internal cubical-shape of crumb, with average side dimension of 14.809± 0.859 mm for both traditional and 3D printed samples, was submitted to texture profile analysis (TPA) by performing a double compression test as reported by Derossi et al. (2020b) with minor modification. A TA-XT plus texture analyzer (Stable Microsystems, Surrey, UK) equipped with a 50 N load cell was used for measures. Two compression cycles were performed for all samples by using a plate of 75 mm diameter (P/75, Stable

MicroSystems, Surrey, UK), pre-test speed of 1.5 mm/s, test speed of 1 mm/s, post-speed of 10 mm/s, compression of 50% of the initial height of the samples. Time between two compressions was set at 5 s. The stress-strain curves were detected and the obtained data were used to compute the following mechanical properties: Hardness (N) as the peak of the first compression; Cohesiveness obtained from area of work during of the second compression cycle divided by the area of work during first compression; Springiness as ratio of the deformation samples of the second compression to that at the first compression, at peak force; Chewiness was obtained by multiplying Hardness, Cohesiveness and Springiness (Kiumarsi et al., 2018). All results were analyzed adopting the software EXPONENT version 2.0.6.0 (Stable Micro System, Surrey, UK). At least 10 replicates were performed for each sample.

3. Results and Discussion

3.1 Balance between printing movement and material deposition

Preliminary, we want to show and discuss on the balance between the printing movements and the deposition of the food formula which is reported to be of primary importance for printing quality (Yang et al., 2018; Guo et al., 2019; Derossi et al., 2020a). With this aim, we have acquired a video during printing process for sample 3D_RO (as representative of the material deposition process). Only 90 s of such video may be observed in video, V1, during which the first 3 layers of batter were deposited. By using the initial layer height of 0.3 mm, the filament of food formula is squeezed on the bed of the printer creating a rectangular cross-sectional shape with semicircular ends (Zhu et al, 2019; Slicr3, 2020) rather than a filament with cylindrical shape. This is commonly considered a good practice to improve the adhesiveness of the first layer on the print bed that, in turn, helps to improve the stability of the 3D printed structures. Also, in this condition, adjacent filaments overlap each other's without space between two parallel depositions. Next, during printing of the second and third layers, it is possible to appreciate some voids between the adjacent filaments. These are the results of the increased layer height at 1.0 mm that reduced the squeezing of the food formula thereby leaving space between parallel depositions (Slic3, 2020). These first considerations allow to state that, although we designed a sample with an infill level of 100%, the printing process may induce the creation of some unexpected and undesired voids that will be better discussed later in a specific section of the paper.

3.2 Visual comparison between traditional and 3D printed sample and printing performance

To compare the overall visual aspect and the main morphological features, some photographic images of the traditional, *H*, and printed, 3D, samples are shown in Figure 2.

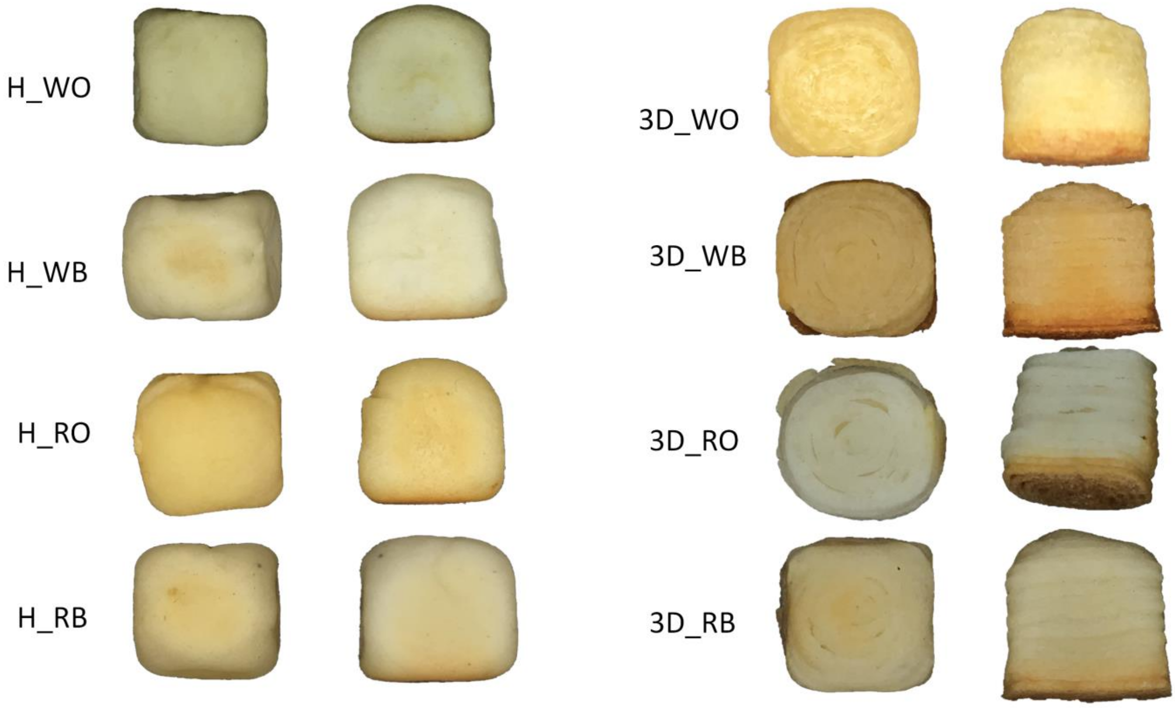


Fig. 2. Photographic images of samples obtained by traditional method and by 3D printing. First and third column show the top view of the samples; second and fourth column show the lateral view.

3D printed samples satisfactorily replicated the overall shape of the digital model. The lateral view allows appreciating the layers of food formula deposited during printing process. Contrarily, the samples prepared by the traditional methods are smooth on the surface. However, from the top view of 3D samples some defects occurring during food formula deposition may be observed, with a like-round shape rather than the expected squared concentric infill pattern of Figure 1. These defects mainly occurred on the top layers while at the bottom of the structure the food formula was correctly deposited according to the squared-shape path. To better illustrate this point Figure 3 shows the changes of the maximum length for 3D_WB sample along its height. According to previous consideration the first 5 mm – from the bottom of the sample – show a highest length of ≈ 32 mm indicating the enlargement of the structure caused by the crushing of the first layers under the total weight of the sample. Then the length of the sample remains approximately constant at 30 mm till the height of 23 mm. After that, the dimension of the samples suddenly decreases till a minimum of 5.98 mm at height of 31 mm. Furthermore, the inner images illustrate the contour of some cross-sectional X-ray images taken at different heights. These allow appreciating the modification of the shape of samples from a like-squared structure (as designed in 3D virtual model of Figure 1) on the bottom of the sample to a clear round-shape at the top of the structure. More precisely, we used the parameter roundness, *Round*, to evaluate the morphological differences of the slices. Results showed values of 0.353, 0.504, 0.809 and 0.811 respectively from the bottom to the top of the sample proving that the roundness of the printed layers significantly increase toward the top of the 3D printed structure.

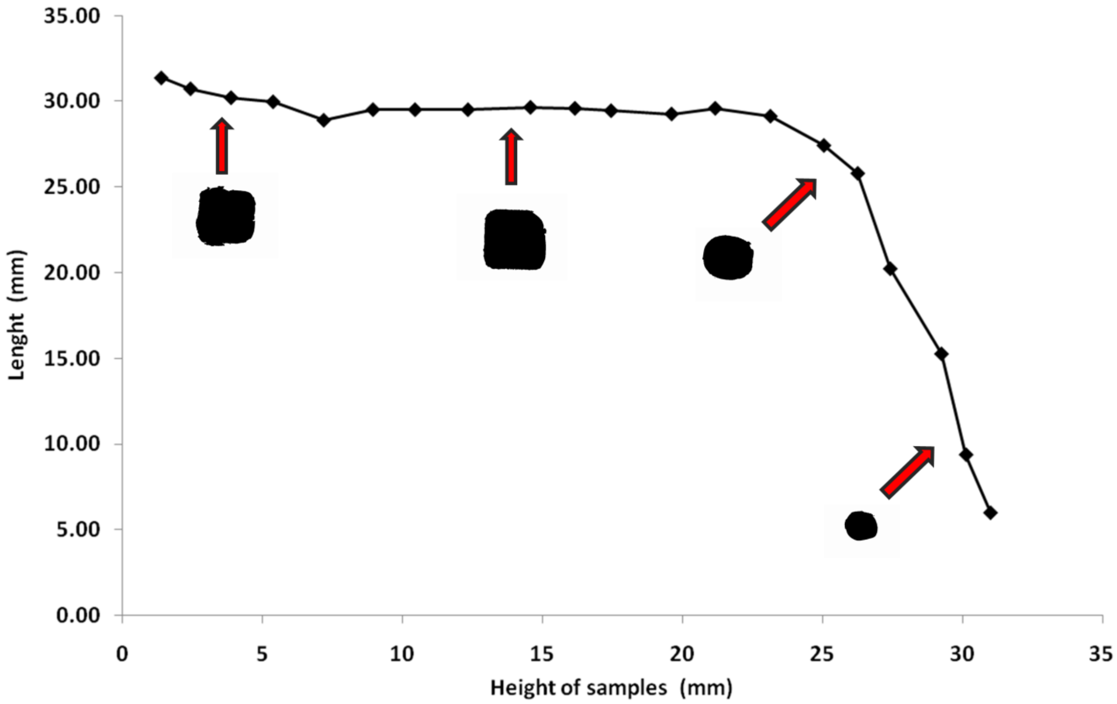


Fig. 3. Dimensional changes of 3D printed samples as a function of its height. Inner figures are the binarized cross sectional X-ray images of 3D_WB samples.

The rising overlaying of layers during the printing process increased the pressure on the bottom layers due to the weight of the sample, thereby generating the partial crushing of such layers (Mantihal et al., 2017). This problem could be related to a low yield stress of the materials that is recognized as an index of the ability of the materials to keep its shape under gravity or the increased stress created by the overlap layers (Lille et al., 2018). As widely reported, an high printability of food inks should admit not only an easy flow through the nozzle but also, it should include the capability of self-supporting the materials which are tightly related with rheological properties of the food-ink such as G', G'' and yield stress (Lille et al., 2018; Liu et al., 2019a; Liu et al. 2019b). The aforementioned crushing of the bottom layers provokes a critical imbalance between the actual layer height (against the defined values of 1.0 mm) and the extrusion rate resulting in a dragging effect of filaments during the movement of the printer. Such a situation significantly reduced printing precision (Khalil and Sun, 2007) of our samples with the result of the observed round-shaped effect as well as the decrease of length of the layer (Guo et al., 2019). Severini et al. (2016) explained how the changes of layer height may affect the dimensional properties of 3D printed cylinder. They reported that for baked samples the layer height exhibited a great effect on the diameter of hollow cylinder printed by using a cereal-based food formula. Accordingly, other authors reported the great importance of the layer height on the shape and dimension of 3D printed food structure (Yang et al., 2018; Wang et al., 2018). Further data aiding this hypothesis are reported in Figure 4 in which some X-ray images of the 3D_WB sample from different heights of the sample are showed. The slice N. 80 (from the first 0.6 mm of the sample) not only shows a well squared external perimeter but also the internal infill exhibits a squared path as effect of the high adhesion of the first layers to the bed of the printer and the crushing of the filaments. Also, we can appreciate the angles of the structure during the printing of infill with approximately 90 degrees. Diversely, the slices

N.340 and N.560 display an internal path with a like-circular movements rather than the designed squared ‘concentric’ infill pathway that is the result of the above discussed imbalance between the actual layer height and the extrusion leading to the dragging effect during printing.

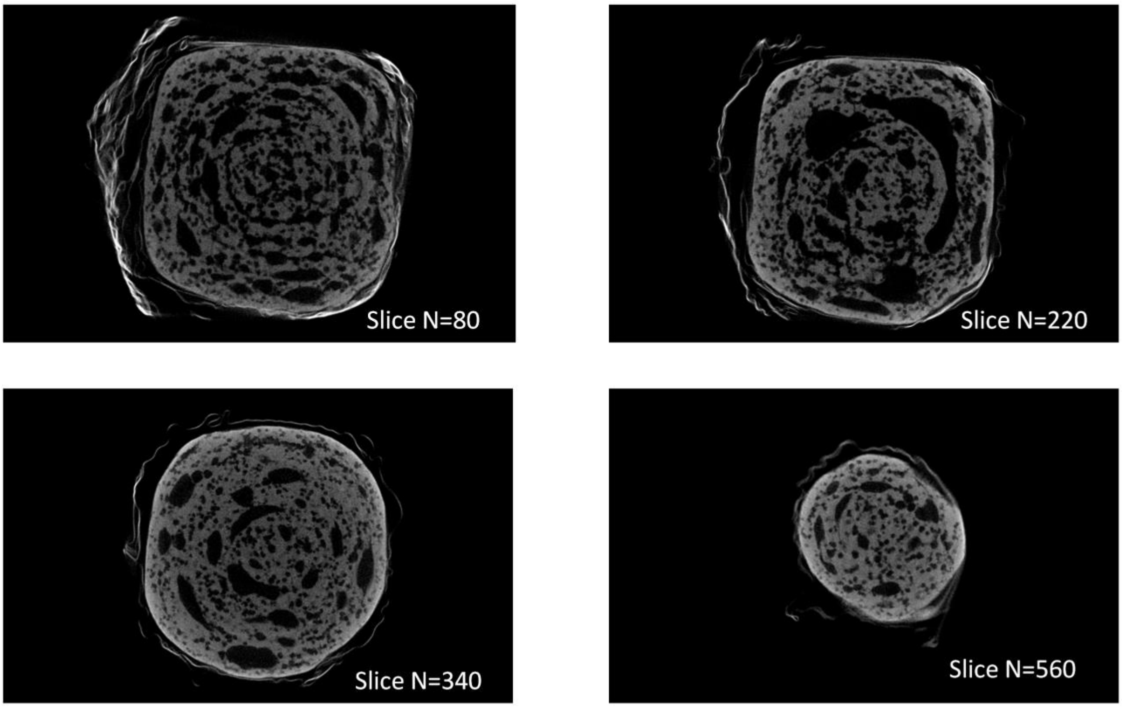


Fig. 4. Cross-sectional X-ray images of 3D printed samples. Different slices represent images taken at different height of the sample.

The overall printing fidelity may be evaluated by the main dimensional properties of H and 3D samples after baking (Table 3).

Table 3 – Main dimensional properties of samples

Type of samples	**L1 (mm)**	**L2 (mm)**	**H (mm)**	**Layer height (mm)**
H_WO	29.36 ± 0.349^{a}	25.78 ± 1.251^{a}	32.53 ± 2.465^{a}	-
H_WB	26.54 ± 2.96^{b}	$28.85\pm0.836^{b,c}$	$32.91\pm2.495^{c,d}$	-
H_RO	$27.74\pm1.31^{a,b}$	$26.98\pm1.334^{a,b}$	$30.25\pm3.413^{b,d}$	-
H_RB	29.14 ± 0.287^{a}	29.01 ± 1.736^{c}	$32.29\pm2.218^{a,b}$	-
3D_WO	29.02 ± 2.52^{a}	$28.47\pm3.0^{b,c}$	29.00 ± 2.810^{d}	1.52 ± 0.27
3D_WB	$28.17\pm3.32^{a,b}$	$29.03\pm2.8^{b,c}$	26.82 ± 2.020^{c}	1.25 ± 0.47
3D_RO	$27.42\pm4.60^{a,b}$	25.37 ± 5.65^{a}	30.03 ± 1.743^{d}	1.87 ± 0.27
3D_RB	$26.44\pm2.11^{a,b}$	$28.66\pm2.201^{b,c}$	29.89 ± 2.591^{d}	2.27 ± 0.48

3D printed samples are significantly larger and taller than the digital model. Instead of 20 mm of length for the 3D virtual model, baked samples exhibited a length in the ranges of 26.44 mm and 29.02 mm. Again, the greater length of the samples is the result of crushing of the layers during printing which was also exacerbated during the first steps of baking in which the yield stress reduced in line with the increase of temperature (Calderon-Dominguez et al., 2007). Contrarily, the greater height of 3D samples, with values between 26.82 and 30.03 mm is caused by the vapor produced during baking which increased the size of the existing pores as well as creating new one (Severini et al., 2016). This phenomenon also caused the increase of the real layer height of the samples as measured from the photographic images of baked samples. Values between 1.25 and 2.27 mm are significantly greater than the layer height of 1 mm defined during slicing. For regard the H samples the lengths are comparable with the 3D printed structure due to the use of the metal mold during baking.

3.3 Microstructure characterization

A first visual comparison of the microstructure of H and 3D samples suggests a broad difference for all food formulas (Figure 5). The pores of 3D printed structures appear larger and situated with a like-roundness structure in line with the above discussed imbalance between actual layer height and extrusion rate (Derossi et al., 2020a; Derossi et al., 2020b). Contrarily, in traditional samples prevails pores randomly distributed with some fissure-like voids as a result of connecting pores. This is also noticeable from the 3D images of H_WO and 3D_WO samples reported in Figure 5 representing a volume of 420^3 pixels.

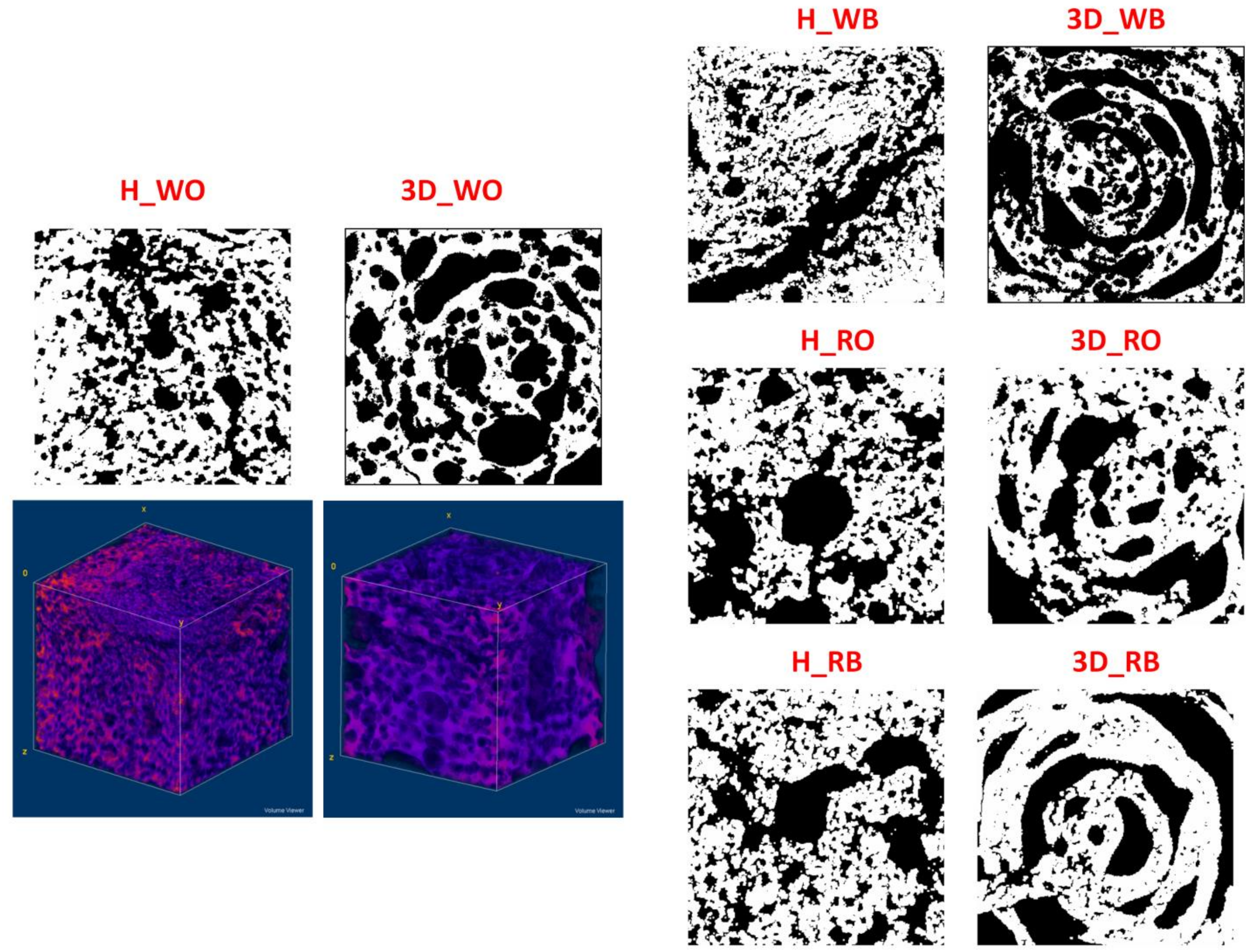


Fig. 5. Cross sectional XCT images of samples obtained by 3D printing and traditional manufacturing process. All the images are taken at the half height of each sample.

In order to get a better characterization of the morphological diversities of these samples a more meaningful microstructure data was considered. Two of the most basic indexes for microstructure characterization are porosity fraction and the total number of pores (Figure 6). Apart from the sample 3D_WO which shows the higher average porosity fraction of 47.40±10.74, the others sample did not show any substantial variations exhibiting values between 33.26±7.80 (H_WB) and 40.20±7.00 (3D_WB). When the rice was added in food formula the porosity fraction reduced for 3D printed samples ($p<0.05$) while remaining approximately constant for H sample. The addition of rice flour and the related reduction of gluten has been reported to reduce the main mechanical properties of food formula (i.e. hardness, chewiness) (Yilmaz et al., 2015) lowering its capacity to resist to the compression in the piston chamber as well as in the extrusion system. Reasonably, for 3D printed samples the higher compressed food formula contains fewer pores in the filament deposited during printing leading to the observed porosity fraction. Contrarily, the reduced hardness of the food formula may be responsible of a slight improvement of the dimension of pores during baking for H samples formulated with olive oil. For the same reasons the 3D printed sampled formulated with rice showed a significant reduced porosity fraction in comparison to the food formula formulated only with wheat flour (47.40% and 40.20 % for 3D_WO and 3D_WB against 35.64% and 37.21% for 3DR_RO and 3D_RB). These data generally recall the great effect of food-ink design on the quality of 3D printed structure through its effect on printability (Zhu et al., 2019; Guo et al., 2019) but also, for the first time, suggest the effects on porosity fraction of deposited food-ink.

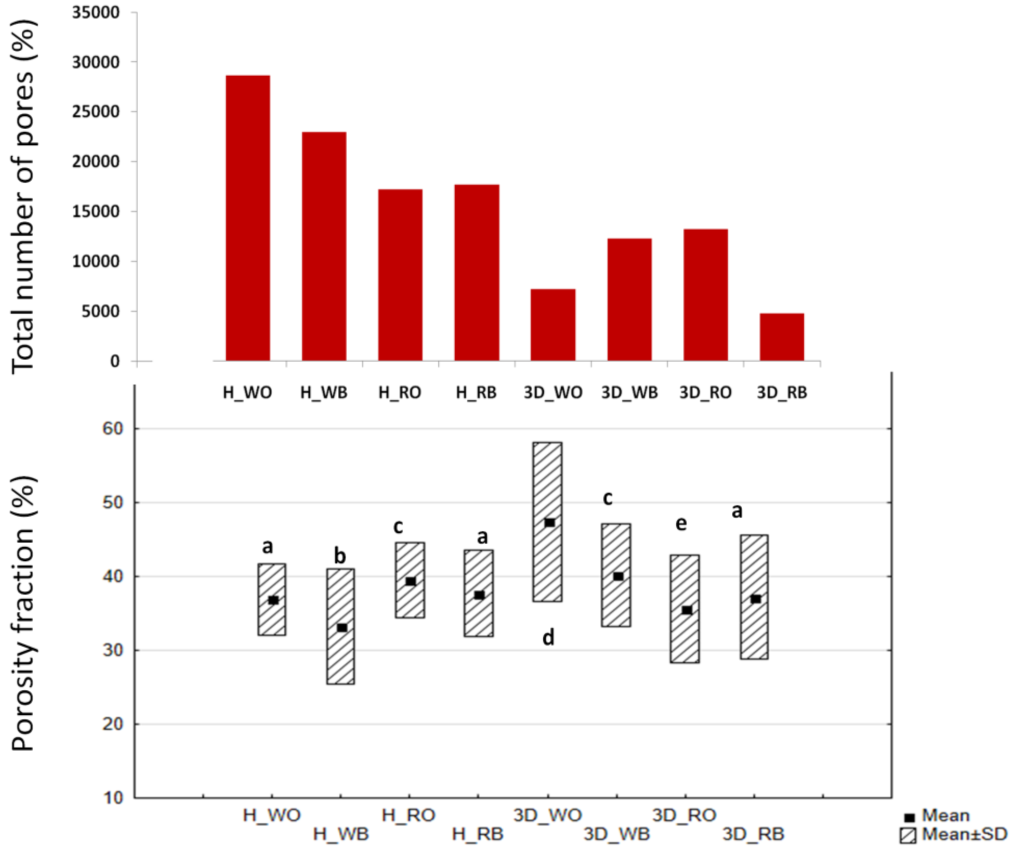


Fig. 6. Porosity fraction and total number of pores of samples obtained by traditional preparation method and 3D printing technologies.

The total number of pores furnishes further information clearly showing that traditional samples contain a significantly higher amount with a peak of 28,654 pores (H_W0) within the all 420 cross-sectional images analyzed. Contrarily, the 3D printed samples showed a peak count of 13,245 pores for 3D_RO. Again, the mechanical compression of the food formula that occurs – for the 3D printer used in these experiments – both in the piston chamber and in the screw-extrusion system could have reduced the number of pores. Furthermore, we want to recall that whereas the compression of food formula in the piston may directly reduce the number of internal pores of the food formula, the screw-extrusion system is equipped with degasifying systems that reinforce the pores reduction during food formula deposition. Once again, we want to recall that the commercial 3D printers used for scientific experiments are still far from optimizing for the customized properties of food products (Derossi et al., 2020). This is challenging for 3D food printing application that could receive several benefits from systems able to modulate the compression force in line with the mechanical properties of food ink (Zhu et al., 2019).
A higher level of microstructure characterization may be obtained by the assessment of pore's morphology by studying the relation between circularity and aspect ratio. Figure 7 reports these morphological properties for sample H_WO for which the following information may be

obtained: 1) the majority of pores exhibited an aspect ratio up to 5; 2) The lack of circularity with the majority of the pores with CIRC < 0.5; 3) the pores with CIRC > 0.7 are few while those with CIRC > 0.9 are rare. To better elucidate the morphological differences among the pores Figure 7 also shows the representing shape of five classes of pores defined on the basis of CIRC and AR values: 1) Fissure-like, 0<CIRC<0.3 and 0<AR<4.5, (Blue); 2) elongated-dissected, 0.3<CIRC<0.55 and 4.5<AR<1.6, (red); 3) Isometric dissected, 0.55<CIRC<0.75 and 1.6<AR<1.2, (green); 4) isometric slightly-dissected, 0.75<CIRC<0.9 and 1.2<AR<1 (yellow); 5) Round, 0.9<CIRC<1 and 1.2<AR<1 (purple). A similar comparison was very useful to classify the morphometric types of the pores space for different soil microstructure (Karsanina et al., 2015; Skvortsova and Sanzharova, 2007) but, for our knowledge, never it has been used for food microstructure.

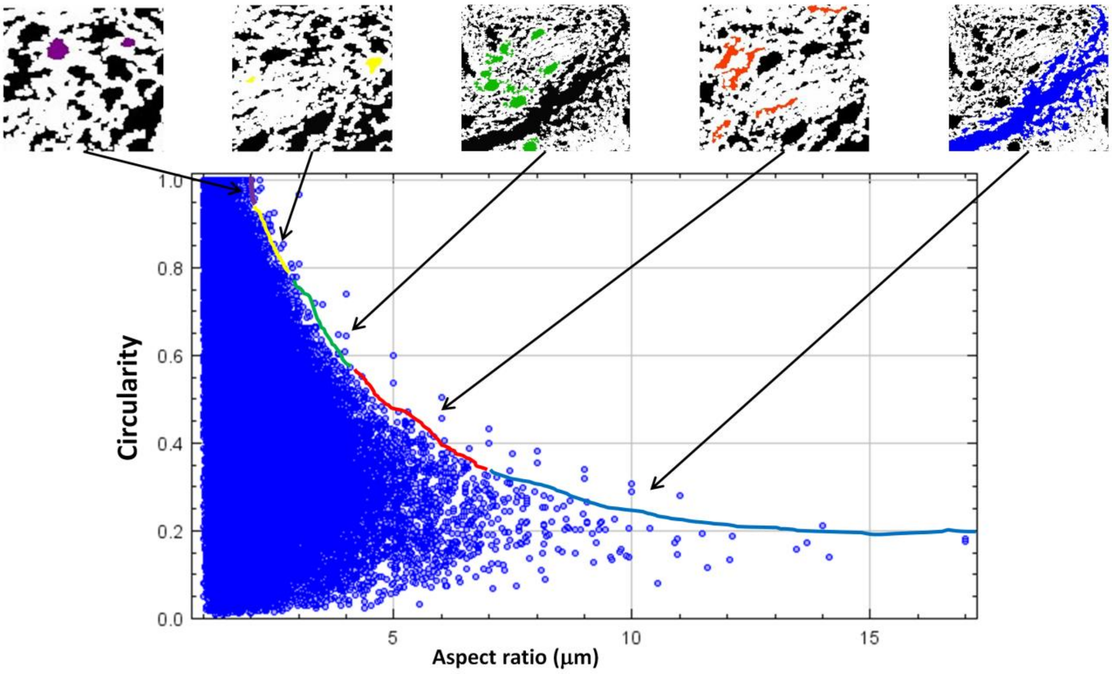


Fig. 7. Morphological properties of pores and main results for sample H_WO.

Furthermore, to better compare the pore morphology of all samples, the fraction of the pores having different circularity and aspect ratio values have been reported in Figure 8. More specifically we used the above reported ranges of CIRC and AR values to classify the pores morphology in 5 different classes. Each figure is obtained by analyzing all pores encountered along the 420 cross sectional X-ray images of each food formula that means a number of data not less than 30,000. First, the analysis of circularity shows that samples obtained by traditional processing contains more pores with CIRC < 0.3 (i.e., fissure like morphology) for all food formula analyzed while the 3D printed samples are better represented by pores with 0.75<CIRC<0.9 that means pores belonging in the class of isometric slightly dissected. Secondly, pores with AR < 1.2 highly represented the 3D printed structures suggesting the existence of pores with a shape more roundness in comparison to the traditional samples which, on the other hand, showed a higher fraction for pores with 1.60<AR<2.5. This is also sustained by the cross-sectional X-ray images in which the perimeter of the pores is highlighted. Another important analysis of pores is the cumulative area distribution (Figure 9). The trend of data clearly show a slower increase for 3D printed samples proving as such samples contain a significant fraction of pores - between 7% and 15% - with an area greater than 5 mm^2 while only a fraction between 2 % and 4.5% of the pores exhibited an area > 5 mm^2 for samples prepared by traditional process. Simply this proves the creation of bigger pores in 3D printed samples probably as consequences of the imbalance between speed print and extrusion rate reported and discussed in video, V1.

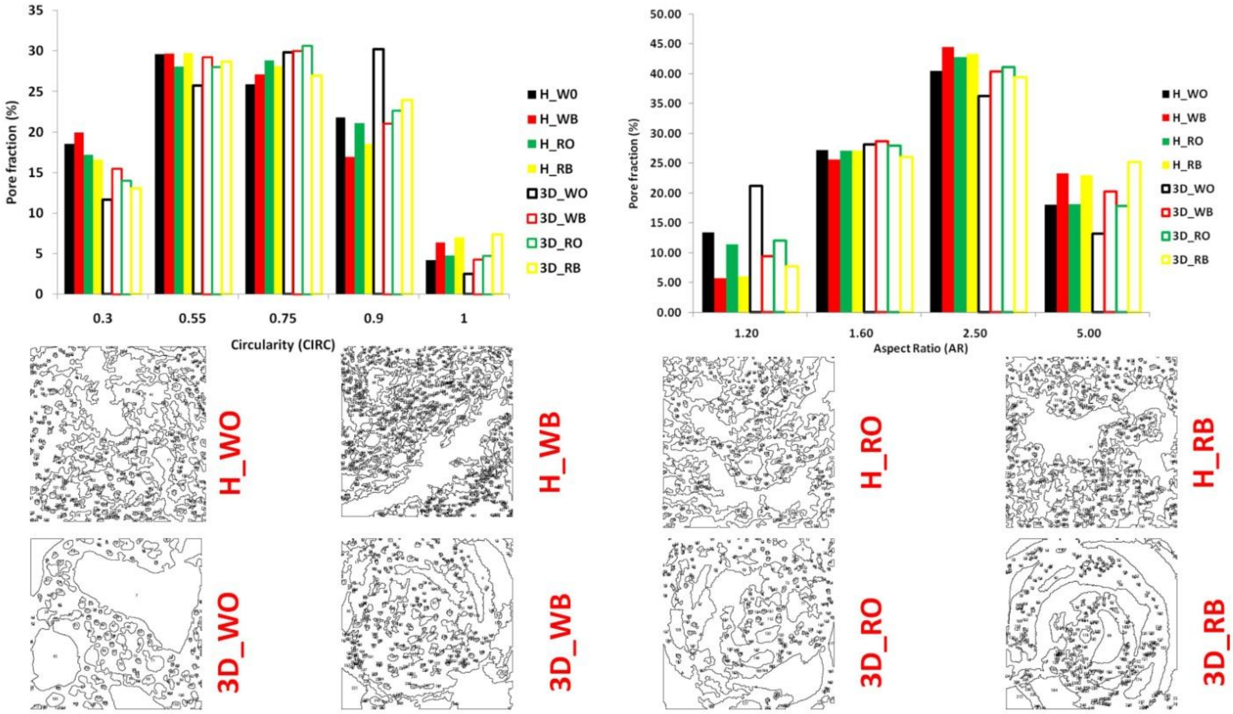


Fig. 8. Relationship between pore's circularity and aspect ratio for samples obtained by traditional processing and by 3D printing techniques. Inner contour images represent the contour of the pores for images taken at the same height, *i.e.* cross-sectional image n. 304 in a total of 420.

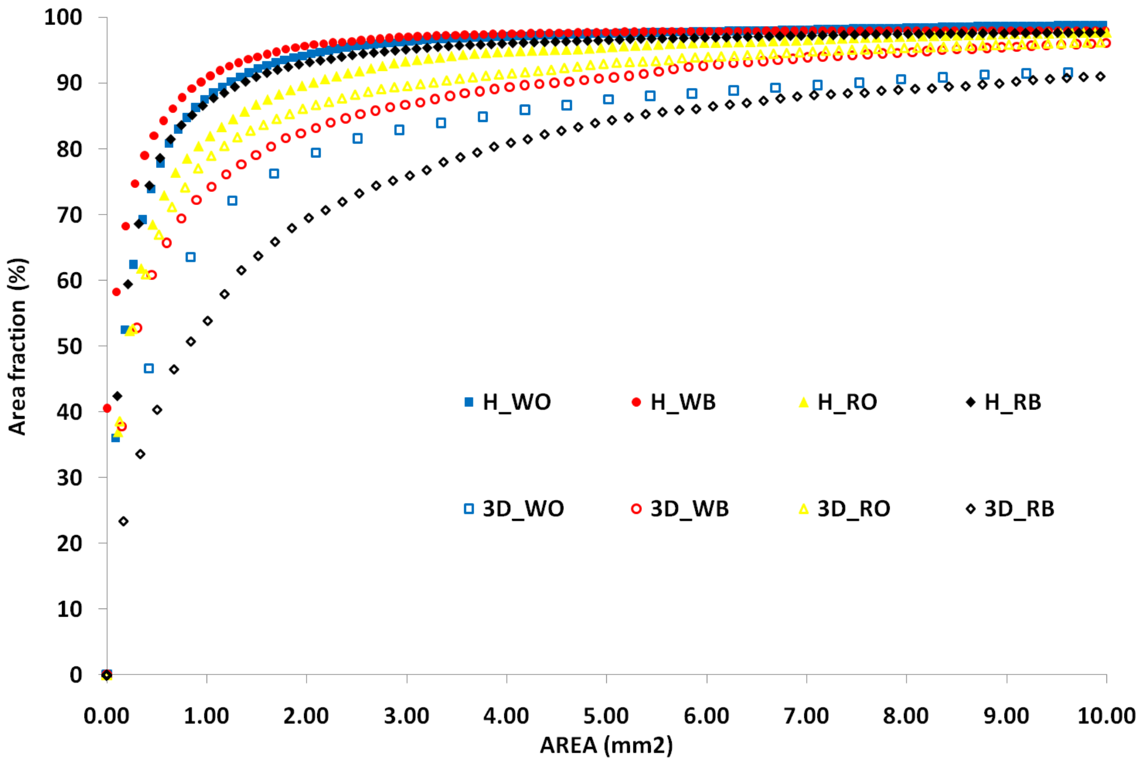


Fig. 9. Area distribution of pores for samples obtained by traditional processing and 3D printed samples.

3.4 Mechanical properties of samples

First, with the aim to analyze possible effect of moisture content on the mechanical properties of samples, Figure 10 shows the moisture content for H and 3D samples. Although the samples were obtained with different techniques, by modulating the baking time (as reported in Material and Methods) the samples exhibited moisture content not statistically different ($p>0.05$) with values of ≈ 30 g H_20/ 100 g w.b. for the crumb and ≈ 15 gH_20/100 g w.b. for the crust.

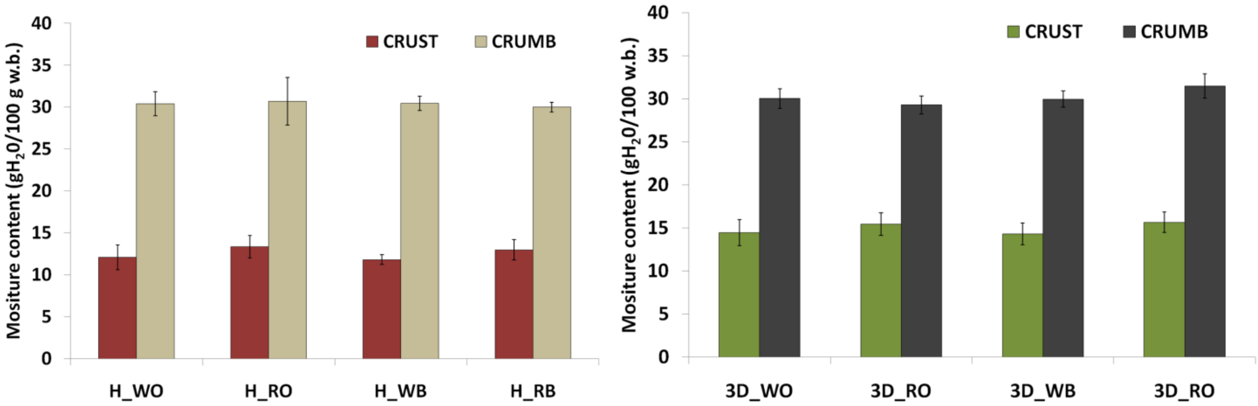


Fig. 10. Moisture content of samples obtained by 3D printing and by traditional processing.

A representative difference of compression-deformation curves for H_WO and 3D_WO samples is reported in Figure 11. The first compression – for both the samples - shows a typical trend with an initial linear-elastic behavior representing the elastic compression of pores inside structure. For sample H_WO this region seems to occur till 2 mm of compression while for 3D_WO this region is prolonged till 4 mm of strain. Then, a plateau region is observed as consequence of the initial formation of some internal damages of the solid phase. Finally, a steeply rising portion of the curves, also called densification (Gibson et al., 1982a; Gibson et al., 1982b), is observed for both samples corresponding to progressive increase of structure density with the samples that behave like a compacted and cohesive mass that opposes high resistance to further compression. On the other hand, the second compression-deformation curve is characterized by the absence of the plateau region – or very slight and difficult to reveal - but only a clear first linear-elastic compression and a sudden rising is observed. The first peak is considered the hardness of the sample and shows a significantly higher value for 3D printed samples than the traditional processing with values respectively of 17.21±3.25 N and 8.37±1.587 N. Also, the second peaks were significantly higher than the first ones suggesting a good adhesion of the crumb particularly for 3D printed sample which, after the first compression, remained more densely packaged inducing a high resistance during the second load. Another interesting mechanical difference is for cohesiveness that was lower for H_WO (0.925±0.089) than for 3D_WO (1.208±0.209).

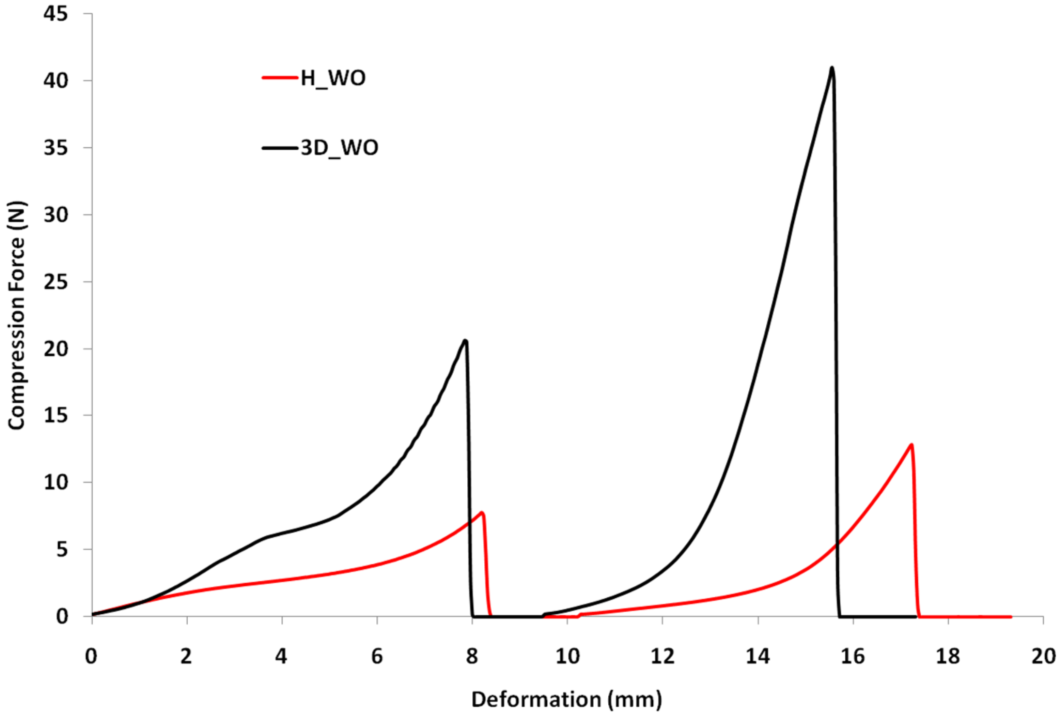


Fig. 11. Stress-strain curve of two representative samples obtained by traditional processing and 3D printing.

In Figure 12 the main values of mechanical properties of samples are reported while a color map indicates the statistical differences among all samples. For hardness, cohesiveness and chewiness the color map clearly indicates the existence of two statistically different groups: the H samples prepared by traditional processing and the 3D printed samples.

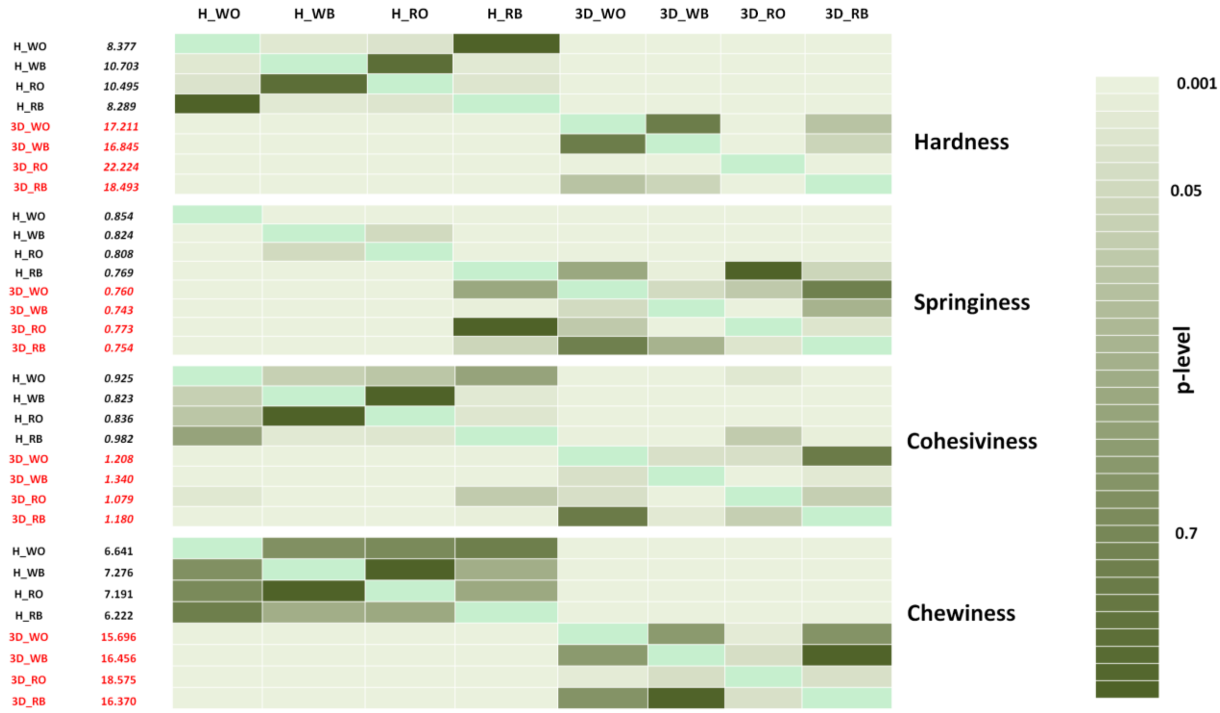


Fig. 12. Main mechanical properties of samples and color map of the significant differences.

By grouping the samples in H and 3D, the hardness of printed structures resulted approximately double that of traditional hand-made sample with values ranging between 16.845 N and 22.224 N. This, in addition, is also observed when we separately compared H and 3D samples for each food formula proving that the layer-by-layer deposition definitely increased the hardness of the samples. These results are nicely complemented with the 3D microstructure information. A possible reason why 3D printed samples showed high hardness is the creation, during layer-by-layer deposition, of few pores bigger in size which were inhomogeneously distributed inside 3D structure of sample (as previously discussed in the section of microstructure analysis). Contrarily, the samples prepared by traditional hand-made process were characterized by a larger number of pores (Figure 6) but small in dimension (Figure 7) which showed a higher homogeneous distribution generating a structure with thinner solid elements that resulted in a lower resistance during the compression test. Another possible rational cause is related to the compression of the food formula occurring inside the piston chamber as well as in the extrusion system – consisting in a screw embedded in extrusion chamber equipped with a degasifying system. These two steps could have reduced the number of closed pores (i.e. isolated pores in the solid phase) inside food formula thereby depositing a more dense food formula filament. So, the majority of the pores detected in 3D samples would be created by printing movements under the control of the G-codes created by slicing software used during slicing of the 3D virtual model as previously observed from the like-roundness distribution of pores (Figure 3). To sustain these hypothesis, we can observe that the higher hardness of 3D printed samples was measured also for samples having high porosity fraction as in the instance of 3D_WO (47.40%) and H_WO (36.89%) or, in addition, for samples 3D_WB (40.20%) compared with H_WB (33.26) ($p<0.05$) (Figure 6). From these first results it can be affirmed that 3D printed samples seem to deviate from the popular and highly useful approach of study that idealize bakery products as repeated cellular solids (Gibson and Ashby 1988; Liu and Scanlon, 2003) – i.e. voids surrounded by solid material in a certain spatial organized structure – that follow a tight relationship between mechanical properties and relative density. This is because the majority of the voids are not randomly distributed in the food formula, as commonly occur during traditional processing, but the majority of them were created during the material deposition in a way that is difficult to manage because resulting from the printing pathway and from the aforementioned imbalance between printing speed and extrusion rate which need further experiments and a better understanding. Furthermore, the experimental data of Figure 12 shows a high chewiness for 3D printed samples, for any food formula considered, with values in the range of 15.696 and 18.575 against the values of traditional samples between 6.222 and 7.272. This is a clear indicator of the higher energy necessary to prepare food before swallowing for 3D printed samples (Peng et al., 2017). Also, there are not statistical differences among the four food formulas when analyzing them within the groups H or 3D ($p>0.05$). This, again, sustains the great effect of 3D printing technology on the mechanical properties of the samples. Further mechanical information is given by the comparison of springiness and chewiness data. Springiness was statistically lower for 3D printed than for traditional samples, independently from the food formula analyzed, stating a lower tendency to be broken during mastication ($p<0.05$) expect for samples H_RB and 3D_RB ($p>0.05$). This is in agreement with the data of cohesiveness that were higher for 3D samples than for H samples. We want to recall that a good printability of ink-food means, among many other aspects, a good adhesion of the filaments deposited during printing. With the purpose of increasing the adhesion between layers a common approach is to employ a layer height of ≈ 80% of the nozzle size (Table 2) thereby depositing filament with rectangular cross-sectional shape with semicircular ends rather than a cylindrical filament (Zhu et al., 2019). Recalling Figure 2 the presence of filaments stuck each other are easy to detect. Reasonably, the good adhesion of the filaments of food formula was the main reasons for the higher cohesiveness and the lower springiness.

4. Conclusions

In this contribution we explored whether 3D printing process *per se* may induce modification on the morphological, microstructure and mechanical properties of food products. Here we show that 3D printed snacks well matched the virtual model but some discrepancies were observed mainly in the top layers – with an undesired like-round deposition path - as a result of the increased actual layer height, against the used values, that was given, in large part, from the crushing of the layers at the bottom of the structure under the weight of overlying layers. 3D printed structures showed a limited number of pores in comparison to the common products and such pores were significantly bigger in size. The inherent compression of the food formula in the piston chamber as well as in the screw-extrusion system was responsible of the reduction of the number of pores thereby depositing a denser food formula filament. However, the aforementioned imbalance between the layer height and extrusion rate was responsible of the creation of the majority of pores structure. Furthermore, the distribution of such pores in 3D printed samples resulted from the defined infill pathway but also it was greatly affected by the above imbalance of printing conditions. This, of course, greatly deviates from the commonly observed random distribution of pores in the samples produced by traditional process. Finally, these diversities in microstructures of samples greatly affected the mechanical properties with 3D printed samples showing a significantly higher hardness, chewiness and cohesiveness than the traditional samples.

5. References

1. AACC (2010). AACC, Approved Methods of Analysis. Method 44-15.02 'Moisture-Air Oven Method'. AACC International. 11th Edition

2. Anukiruthuka, T., Moses, J.A., & Anandharamakrishnan, C. (2020). 3D Printing egg yolk and white with rice flour blends. *Journal of Food Engineering*, 265, 109691. https://doi.org/10.1016/j.jfoodeng.2019.109691

3. Blurhapsody (2020). Unlesh your creativity in the kitchen. https://blurhapsody.com/en/ Accessed 6 July 2020-07-06

4. Calderon-Dominguez, G., Vera-Dominguez, M., Farrera-Rebollo, R., Arana-Errasquin, R., & Mora-Escobedo, R. (2017). Rheological changes of dough and bread quality prepared from a sweet dough: effect of temperature and mixing time. *International Journal of Food Properties*, 7:2, 165-174. Doi: 10.1081/jfp-120025393.

5. Carlton, H.D., Haboub, A., Gallegos, G.F., Parkinson, D.Y., & MacDowell, A.A. (2016). Damage evolution and failure mechanisms in additively manufactured stainless steel. Material Science and Engineering, 651, 406–14. doi: 10.1016/j.msea.2015.10.073

6. Chan, S.S.L., Pennings, R.M., Edwards, L., & Franks, G.V. (2020). 3D printing of clay for decorative architectural applications: Effect of solids volume fraction on rheology and printability. *Additive Manufacturing*, 35, 101335. https://doi.org/10.1016/j.addma.2020.101335

7. Chen, J., Mu, T., Goffin, D., Blecker, C., Richard, G., Richel, A., & Haubruge, E. (2019). Application of soy protein isolate and hydrocolloids based mixtures as promising food material in 3D food printing. *Journal of Food Engineering*. doi:10.1016/j.jfoodeng.2019.03.016

8. Chen, J., Mu, T., Goffin, D., Blecker, C., Richard, G., Richel, A., & Haubruge, E. (2020). Application of soy protein isolate and hydrocolloids based mixtures as promising food material in 3D food printing. *Journal of Food Engineering*, 261, 76-86. Doi: 10.1016/j.jfoodeng.2019.03.016.

9. DebRoy, T., Wei, H.L., Zuback, J.S., Mukherjee, T., Elmer, J.W., Milewski, J.O., Beese, A.M., Wilson-Heid, A., De, A., & Zhang, W. (2018). Additive manufacturing of

metallic components – Process, structure and properties. *Progress in Material Science*, 2, 112-224. doi: 10.1016/j.pmatsci.2017.10.001

10. Derossi, A, Caporizzi, R., Azzollini, D., & Severini, C. (2018). Application of 3D printing for customized food. A case on the development of a fruit-based snack for children. *Journal of Food Engineering*, 220, 65-75.

11. Derossi, A., Paolillo, M., Caporizzi, R., & Severini, C. (2020a). Extending the 3D food printing test at high speed. Material deposition and effect of non-printing movements on the final quality of printed structures. *Journal of Food Engineering*, 275, 109865. https://doi.org/10.1016/j.jfoodeng.2019.109865

12. Derossi, A., Caporizzi, R., Paolillo, M., & Severini, C. (2020b). Programmable texture properties of cereal-based snack mediated by 3D printing technology. *Journal of Food Engineering*, in press. https://doi.org/10.1016/j.jfoodeng.2020.110160

13. Dianez, I., Gallegos, C., Brito-de la Fuente, E., Martinez, I., Valencia, C., & Sanchez, M.C. (2019). 3D printing in situ gelification of k-carrageenan solutions: effect of printing variables on the rheological response. *Food Hydrocolloids*, 87, 321-330.

14. Dick, A., Chandari, B., & Prakash, S. (2019). Post-processing feasibility of composite-layer 3D Printed beef. *Meat Science*, 153, 9-18. https://doi.org/10.1016/j.meatsci.2019.02.024

15. Emon, Md.O.F., Alkaidi, F., Philip, D.G., Kim, D.H., Lee, K.C., & Choi, J.W. (2019). Multi-material 3D printing of a soft pressure sensor. *Additive Manufacturing*, 28, 629-638. https://doi.org/10.1016/j.addma.2019.06.001

16. Feng, C., Zhang, M., Bhandari, B., & Ye, Y. (2020). Use of potato processing by-product: effects on the 3D printing characteristics of the yam and the texture of air-fried yam snacks. *LWT-Food Science and Technology*, in press. doi: 10.1016/j.lwt.2020.109265.

17. Gholamipour-Shirazi, A., Norton, I.T., & MIlls, T. (2019). Designing hydrocolloids based food-ink formulations for extrusion 3D printing. *Food Hydrocolloids*, 95, 161-167

18. Gibney, R., Matthyssen, S., Ferraris, J.P.E., & Zakaria, N. (2017). The human cornea as a model tissue for additive biomanufacturing: a review. *Procedia CIRP*, 65, 56-63. https://doi.org/10.1016/j.procir.2017.04.040

19. Gibson L.J., &, Ashby, M.F. (1988). Cellular solids. Structure and properties. Cambridge Solids State Science Series

20. Gibson, L.J., Ashby, M.F., Schajer, G.S., & Robertson, C.I. (1982a). The mechanics of two-dimensional cellular materials. *Proc. R. Soc. Lond. A*, 382, 25-42.

21. Guo C., Zhang, M., & Bhandari, B. (2019). Model building and slicing in Food 3D printing processes: a review. *Comprehensive reviews in food science and food safety*. https://doi.org/10.1111/1541-4337.12443

22. Hamilton, A.C., Alici, G., & in het Panhuis, M. (2017). 3D printing Vegemite and Marmite: Redefining 'breadboards'. *Journal of Food Engineering*, 220, 83-88. doi: 10.1016/j.jfoodeng.2017.01.008.

23. Ho, S.Y. (2009). Anatomy and myoarchitecture of the left ventricular wall in normal and in disease. *European Journal of Echocardiography*, 10, iii3–iii7. doi:10.1093/ejechocard/jen243

24. Hosny, A., Keating, S.J., Dilley, J., Ripley, B., Kelil, T., Pieper, S., Kolb, D., Bader, C., Pobloth, A.M., Grifin, M., Nezafat, R., Duda, G., Chiocca, E.A., Stone, J.R., Michaelson, J.S., Dean, M.N., Oxman, N., & Weaver, J. (2018). Planning: high-resolution functionally graded multimaterial 3D printing of biomedical tomographic data sets. *3D Printing and Additive Manufacturing*, 5, 2, 103-113. Doi: 10.1089/3dp.2017.0140.

25. Kapetaniou, C., Rieple, A., Pilkington, A., Frandsen, T., & Pisano, P. (2017). Building the layers of a new manufacturing taxonomy: how 3D printing is creating a new landscape of production eco-systems and competitive dynamics. *Technological Forecasting & Social Change*, 128, 22-35. https://doi.org/10.1016/j.techfore.2017.10.011

26. Karavasili, C., Gkaragkounis, A., Moschakis, T., Ritzoulis, C., & Fatouros, D. G. (2020). Paediatric-friendly chocolate-based dosage forms for the oral administration of both hydrophilic and lipophilic drugs fabricated with extrusion-based 3D printing.

European Journal of Pharmaceutical Sciences, 30, 147, 105291. doi:10.1016/j.ejps.2020.105291

27. Karsanina, M.V., Gerke, K.M., Skvortsova, E.B., & Mallants, D. (2015). Universal spatial correlation functions for describing and reconstructing soil microstructure, *PLOSone*, 10 (5), e0126515. Doi:10.1371/journal.pone.0126515.

28. Khalil, S., & Sun, W. (2007). Biopolymer deposition for freeform fabrication of hydrogel tissue constructs. *Materials Science and Engineering*: *C*, 27(3), 469–478. https://doi.org/10.1016/j.msec.2006.05.023

29. Kiumarsi, M., Shahbazi, M., Yeganehzad, S., Majchrzak, D., Lieleg, O., & Winkwljann, B. (2018). Relation between structural, mechanical and sensory properties of gluten-free bread as affected by modified dietary fibers. *Food Chemistry*, 277, 30-664-673. https://doi.org/10.1016/j.foodchem.2018.11.015

30. Italian Legislation for wheat flour and pasta (2001). Decreto del Presidente della Republica del 9 Febbraio 2001, n. 187. https://www.politicheagricole.it/flex/files/2/f/6/D.34ca305e98ded6c87bfc/DPR_187_2001.pdf

31. Lanaro, M., Forrestal, D.P., Scheurer, S., Slinger, D.J., Liao, S., Powell, S.K., & Woodruff, M.A. (2017). 3D printing complex chocolate objects: platform design, optimization and evaluation. *Journal of Food Engineering*, 215, 13-22. https://doi.org/10.1016/j.jfoodeng.2017.06.029

32. Lee, J.M., & Yeong, W.Y. (2020). Engineering macroscale cell alignment through coordinate toolpath design using support-assisted 3D bioprinting. Journal of the Royal Society Interface, 17(168), 20200294, https://doi.org/10.1098/rsif.2020.0294

33. Le Tohic, C., O'Sullivan, J.J., Drapala, K.P., Chartrin, V., Chan, T., Morrison, A.P., Kerry, J.P., & Kelly, A.L. (2018). Effect of 3D printing on the structure and textural properties of processed cheese. *Journal of Food Engineering*, 56-64. https://doi.org/10.1016/j.jfoodeng.2017.02.003

34. Lee, J. H., Won, D. J., Kim, H. W., & Park, H. J. (2019). Effect of particle size on 3D printing performance of the food-ink system with cellular food materials. *Journal of Food Engineering*, 256, 1-8. doi:10.1016/j.jfoodeng.2019.03.014

35. Li, R., Niu, P., Yuan, T., Cao, P., Chen, C., & Zhou, K. (2018). Selective laser melting of an equiatomic CoCrFeMnNi high-entropy alloy: Processability, non-equilibrium microstructure and mechanical property. *Journal of Alloys and Compounds*, 746, 125-134. https://doi.org/10.1016/j.jallcom.2018.02.298

36. Lille, M., Nurmela, A., Nordlund, E., Metsa-Kortelainen, S.M., & Sozer, N. (2018). Applicability of protein and fiber-rich food materials in extrusion-based 3D printing. *Journal of Food Engineering*, 220, 20-27. https://doi.org/10.1016/j.jfoodeng.2017.04.034

37. Lin, Y.J., Punpongsanon, P., Wen, X., Iwai, D., Sato, K., Obrist, M., & Mueller, S. (2020). FoodFab: Creating Food perception illusion using Food 3D Printing. In Proceeding of CHI 2020.

38. Liu, Z., Bhandari, B., Prakash, S., Manthial, S., & Zhang, M. (2019a). Linking rheology and printability of a multicomponent gel system of carrageenan-xanthan-starch in extrusion based additive manufacturing. *Food Hydrocolloids*, 87, 413-424. https://doi.org/10.1016/j.foodhyd.2018.08.026

39. Liu, Y., Tu, Y., Liu, C., Regenstein, J.M., Liu, X., & Xhou, P. (2019b). Rhelogical and mechanical behavior of milk protein composite gel for extrusion-based 3D food printing. *LWT-Food Science and Technology*. doi. 10.1016/j.lwt.2018.12.053.

40. Liu, Z., Zhang, M., Bhandari, B., & Yang, C., (2018a). Impact of rheological properties of mashed potatoes on 3D printing. *Journal of Food Engineering*, 220, 76–82. https://doi.org/10.1016/j.jfoodeng.2017.04.017.

41. Liu, Z., Bhandari, B., Prakash, S., & Zhang, M. (2018b). Creation of internal structure of mashed potato constructed by 3D printing and its textural properties. *Food Research International*, 111, 2018, 534-543. https://doi.org/10.1016/j.foodres.2018.05.075.

42. Liu, Z., & Scanlon, M.G. (2003). Predicting mechanical properties of bread crumb. Trans IChemE, 81, 224-238.

43. Lupton, D. (2017). 'Dowload to delicious': promissory themes and sociotechnical imaginaries in coverage of 3D printed food in online news sources. *Futures*, 93, 44-53. https://doi.org/10.1016/j.futures.2017.08.001

44. Manthial, S., Prakash, S., Condi Godoi, F., & Bhandari, B. (2017). Optimization of chocolate 3D printing by correlating thermal and flow properties with 3D structure modeling. *Innovative Food Science and Emerging Technologies*, 44, 21-29. https://doi.org/10.1016/j.ifset.2017.09.012

45. Marlin (2020). Configuring Marlin. https://marlinfw.org/docs/configuration/configuration.html Accessed 13 May 2017.

46. Maya (2020). On-demand manufacturing transforms the relationship between consumer and brand. https://Maya.com/work/oreo Accessed 6 July 2020

47. MIT (2020a). Printing transparent glass in 3D. http://news.mit.edu/2015/3-d-printing-transparent-glass-0914. Accessed 6 July 2020

48. MIT (2020b). Printing objects that can incorpporate living organisms. http://news.mit.edu/2020/3-d-bioprinting-living-materials-0123. Accessed 17 April 2020

49. Nichols, M.R. (2019). How does the automotive industry benefit from 3D metal printing. *Metal Powder Report*, 74, 5, 257-258. https://doi.org/10.1016/j.mprp.2019.07.002

50. Park, S.M., Kim, H.W., & Park, H.J. (2020). Callus-based 3D printing for food exemplified with carrot tissues and its potential for innovative food production. *Journal of Food Engineering*, 271, 109781. DOI: 10.1016/j.jfoodeng.2019.109781.

51. Peng, B., Li, Y., Ding, S., & Yang, J. (2017). Characterization of textural, rheological, thermal, microstructural, and water mobility in wheat flour dough and bread affected by trehalose. *Food Chemistry*, 233, 369-377. 10.1016/j.foodchem.2017.04.108

52. Piyush, Kumar, R., & Kumar, R. (2020). 3D printing of food materials: A state of art review and future. *Material Today: Prooceedings*, in press. https://doi.org/10.1016/j.matpr.2020.02.005

53. Pulatsu, E. T., Su, J.-W., Lin, J., & Lin, M. (2020). Factors affecting 3D printing and post-processing capacity of cookie dough. *Innovative Food Science & Emerging Technologies*, 102316. doi:10.1016/j.ifset.2020.102316

54. Rodriguez, M.J., Brown, J., Giordano, J., Lin, S.J., Omenetto, F.G., & Kaplan, D.L. (2017). Silk based bioinks for soft tissue reconstruction using 3-dimensional (3D)

printing with in vitro and in vivo assessments. *Biomaterials*, 117, 105-115. https://doi.org/10.1016/j.biomaterials.2016.11.046

55. Severini, C., Derossi, A., & Azzollini, D. (2016). Variables affecting the printability of foods: Preliminary tests on cereal-based products. *Innovative Food Science & Emerging Technologies*, 38, 281–291. doi:10.1016/j.ifset.2016.10.001

56. Severini, C., Derossi, A., Ricci, I., Caporizzi, R., & Fiore, A. (2018a). Printing a blend of fruit and vegetables. New advances on critical variables and shelf life of 3D edible objects. *Journal of Food Engineering*, 220, 89-100. https://doi.org/10.1016/j.jfoodeng.2017.08.025

57. Severini, C., Azzollini, D., Albenzio, M., & Derossi, A. (2018b). On printability, quality and nutritional properties of 3D printed cereal based snacks enriched with edible insects. *Food Research International*, 106, 666–676. doi:10.1016/j.foodres.2018.01.034.

58. Skvortsova, E.B., & Sanzharova, S.I. (2007). Micromorphometric features of pore space in the plow horizons of loamy soils. *Mineralogy and Micromorphology of Soils*, 40 (4), 445-455. doi:10.1134/S1064229307040114.

59. Slic3r (2020). Slic3r Manual – Flow math. https://manual.slic3r.org/advanced/flow-math Accessed 1 July 2020.

60. Spiegel, C.A., Hippler, M., Munchinger, A., Bastmeyer, M., Barner-Kowollik, C., Wegener, M., & Blasco, E. (2020). 4D Printing at the microscale. *Advanced Functional Material*, 30, 1907615. doi: 10.1002/adfm.201907615

61. Vancauwenberghe, V., Katalagarianakis, L., Wang, Z., Meerts, M., Hertog, M., Verboven, P., Moldenaers, P., Hendrickx, M.E., Lammertyn, J., & Nicolai, B. (2017a). Pectin based food-ink formulations for 3D printing of customizable porous food stimulants. *Innovative Food Science and Emerging Technologies*, 42, 138-150. doi:10.1016./j.ifset.2107.06.011.

62. Vancauwenberghe, V., Mbong, V.B.M., Vanstreels, E., Verboven, P., Lammertyn, J., & Nicolai, B. (2019). 3D printing of plant tissue for innovative food manufacturing: encapsulation of alive plant cells into pectin based bio-ink. *Journal of Food Engineering*, 263, 454-464.

63. Vantyghem, G., De Corte, W., Shakour E., & Amir, O. (2020). 3D Printing of a post-tensioned concrete girder designed by topology optimization. *Automation in Construction*, 112, 103084. https://doi.org/10.1016/j.autcon.2020.103084

64. Wang, l, Zhang, M., Bhandari, B., & Yang, C (2018). Investigation of fish surimi gel as promising food material for 3D printing. *Journal of Food Engineering*, 220, 101-108. https://doi.org/10.1016/j.jfoodeng.2017.02.029

65. Wang, L., Zhang, M., Bhandari, B., Yang, C. (2020). Investigation of fish surimi gel as promising food material for 3D printing. *Journal of Food Engineering*, 220, 101-108.

66. Wu, W., Haidong, D., Hang, S., Sun, B., Wang, B., YU, Z., Ni, H., Li, G., & Zhao, J. (2018). Study of printing parameters of pneumatic-injection 3D printing of Fe-based metallic glass. *Journal of Non-Crystalline Solids*, 489, 50-56. https://doi.org/10.1016/j.jnoncrysol.2018.03.027

67. Xu, L., Gu, L., Su., Y., Chang, C., Wang, J., Dong, S., Liu, Y., Yang, Y., & Li, J. (2020). Impact of thermal treatment on the rheological, microstructural, protein structures and extrusion 3D printing characteristics of egg yolk. *Food Hydrocolloids*, 100, 105399. https://doi.org/10.1016/j.foodhyd.2019.105399

68. Yang, F., Zhang, M., Bhandari, B., & Liu, Y. (2018). Investigation on lemon juice gel as food material for 3D printing and optimization of printing parameters. *LWT-Food Science and Technology*, 87, 67-76. https://doi.org/10.1016/j.lwt.2017.08.054

69. Yilmaz, M.T., Yildiz, O, Yurt, B., Toker, O.S., Karam, S., & Basturk, A. (2015). A mixture design study to determine interaction effects of wheat buckwheat, and rice flours in an aqueous model system. *LWT – Food Science and Technology*, 61, 583-389. https://doi.org/10.1016/j.lwt.2014.11.045

70. Zhu, S., Stieger, M.A., van der Goot, A.J., & Schutyser, M.A.I. (2019). Extrusion-based 3D printing of food pasts: Correlating rheological properties with printing behavior. *Innovative Food Science and Emerging Technologies*, 58, 102214. doi: 10.1016/j.ifset.2019.102214

Chapter 4 Accelerating the process development of innovative food products by prototyping through 3D printing technology

Authors: Derossi, A.[Φ], Corradini, M.G.[Ƨ], Caporizzi, R.[Φ], Oral, M.O.[Φ], Severini, C.[Φ]

[Ƨ]*Department of Agriculture, Food, Natural Resources and Engineering, University of Foggia, Italy*
[Φ]*Department of Food Science & Arrell Food Institute, University of Guelph, Guelph, ON, Canada*

Corresponding author: rossella.caporizzi@unifg.it

Abstract

Discoveries and applications of 3D food printing (3DFP) keep growing with a large set of outcomes, such as the development and optimization of novel edible inks and post-printing processes. However, the main principle at the core of 3D printing, i.e., Rapid Prototyping hasn't received the deserved attention. Thus, beyond generating innovative food products, this study emphasizes the opportunities and benefits of 3DFP for food prototyping. Several cereal-based snacks were generated, and their corresponding baking kinetics were monitored considering changes in the most important characteristics of the product, its impact on productivity and potential safety concerns. The prototypes had two shapes ` (cylindrical and cubical), three infill levels (30, 60, and 100%) and two infill paths (grid and gyroid). Moisture content was satisfactorily described by using a modified version of a logistic model (r >0.98), enabling to estimate and then, successfully validating the baking time to get a moisture content lower than 0.1 g H_2O/g dry basis (d.b.). 3D printed structures with 30% infill exhibited a significant reduction in baking time (from 16.5 to 19.2 min) against not only those with 60% infill but also the control, which needed 38.6 min of baking; this emphasizes the benefits of controlled pattern designs on energy consumption and increasing hourly production. Furthermore, acrylamide formation was estimated to be between 39 and 91% lower than in the control samples after receiving comparative thermal treatments. 3DFP may be used as an important prototyping tool, extending and accelerating the manufacturing of food products with economic, nutritional/toxicological, societal, and environmental benefits.

Keywords: Food prototyping, 3D printing, process development, baking time, cost, sustainability, food quality

1. Introduction

The advent of additive manufacturing has changed the mindset of food science and technology professionals, with an exponential increase in multidisciplinary experiments involving digital, engineering, rheological, chemical, and physical aspects of foods (Attaran et al., 2020). As reported in SCOPUS, 170 manuscripts authored by 582 professionals have been published on 3D food printing (3DFP) from 2007 to 2020 (Derossi et al., 2020). The interest in 3DFP has been fueled by 1) the opportunity of converting a digital CAD model into a physical edible structure and 2) the substantial degree of freedom that facilitates the creation of complex structures unattainable by traditional manufacturing technologies. This enables market innovation while also materializing new visionary concepts such as the creation of nutrition and sensory personalized food products, the advancement of sustainable and healthy diets (SHD), food waste reduction, and on-demand food production (Wong et al., 2022). Furthermore, all these opportunities are strictly relevant to the Sustainable Development Goals of the United Nations (SDGs), such as SDG 2 (Zero hunger), SDG3 (Good health and well-being), SDG 9 (Industry, innovation and infrastructure), SDG12 (Responsible consumption and production), as well as the principles of better healthy and sustainable diets (WHO, 2019,) and food systems (Mattas et al., 2022).

Initially, 3DFP experiments have focused on the potential application of different printing technologies to food products (Zhang et al., 2022; Ling et al., 2022; Pulatsu et al., 2022a; Pulatsu et al., 2022b), the effect of printing variables (Derossi et al., 2019), and the possibility of diversifying the food materials such as ink-gels, cereal-based dough, chocolate, fruit and vegetables, etc. (Chen et al., 2021; Kim et al., 2021; Rando and Ramaioli, 2021; Tomasevic et al., 2021; Pulatsu et al., 2021). Later, 3DFP research concentrated on the description, optimization and selection of the best properties (e.g., rheological) to make food materials highly printable. Food printability is a complex term, encompassing many aspects such as easy and homogeneous flow through a nozzle, optimal adhesion of the food layers, and the capacity of the printed structure to withstand the weight of overlying layers (Zhang et al., 2022). Overall, the successive stages during the material deposition of an edible link can be divided into the initiation of flow, continuous extrusion, recovery stage and final deposition. The most relevant characteristics of the edible ink to optimize during these phases are its yield stress, viscosity, shear-thinning and recovery behaviour (Siacor et al., 2021; Kim et al., 2019; Qiu et al., 2022; Rahman et al., 2020). An ideal food-ink should be highly homogeneous so that its rheological properties remain constant during its continuous deposition. Also, its yield stress and viscosity should not be too high, which could hamper the deposition process or too low, which would result in insufficient mechanical strength to bear the weight of successive layers (Zhang et al., 2022; Cheng et al., 2022; Paolillo et al., 2021). In addition, high shear-thinning and fast recovery behaviour ensure an easy flow during deposition and optimal mechanical properties of the food filament after leaving the nozzle (Dick et al., 2019). Dimensional stability, e.g., lack of spreading after printing, is essential to ensure the retention of the shape of the printed food during manufacturing, post-printing processing, storage and transportation (Nijdam et al., 2021). To facilitate flow and maintain structural stability several structuring agents such as hydrocolloids, other carbohydrates and lipids have been used (Chen et al., 2022; Liu et al., 2021; Guo et al., 2021). More recently highly concentrated emulsions (Zhang et al., 2022; Feng et al., 2022) and cellulose nanocrystals (Armstrong et al., 2022) have been proposed as novel structuring agents in 3DFP.

Lately, this field has been enriched by intriguing approaches and applications mainly focused on the expansion of food printing from 3D to 4D, 5D and 6D (Teng et al., 2021; Cheng et al., 2021; Zhagal et al., 2022) and the possibility to get programmable texture properties (Fahmy et al., 2021; Oral et al., 2021). Regarding 4DFP the research has centered on changing the color, aroma, morphology (shape or specific dimensions), and even nutritional accessibility of the printed structures in response to external stimuli such as high temperature (Guo et al., 2021; Guo et al., 2021; Chen et al., 2021) or pH (Phuhongsung et al., 2020; Ghazal et al. 2021; Chen et al., 2021; Liu et al., 2021Oral et al., 2021). For instance, Chen et al. (2021) printed edible inks consisting of lotus root powder, water and a curcumin emulsion that elicit different colors, from yellow to red, depending on curcumin concentration and the extent of microwave heating that it was exposed to. Also, He et al. (2020) modulated the color throughout a printed food product composed of sweet potato puree rich in anthocyanins (SPS) and mashed potatoes (MP) which exhibited layers at different pHs. After depositing layers of SPS on MP a color change was observed as a function of time due to the diffusion of curcumin through the layers and their progressive contact with layers at different pHs. Another external stimulus often used in 4D printing applications is ionic strength (Champeau et al., 2020). Future applications will involve additional dimensions and will imply the development of complex structures with unique and currently unattainable properties by adding movement to the ‘traditional’ 3D matrix. This could be achieved by printing while moving the printing head and the printing beds at different angles. Using the numbers of peaks recorded during the compression was reported as the Crispness of the products this set up, the curved deposition of the food ink will allow for new benefits not only in terms of visual appeal but also structural strength and stability, reduced number of layers of the deposited materials, etc.

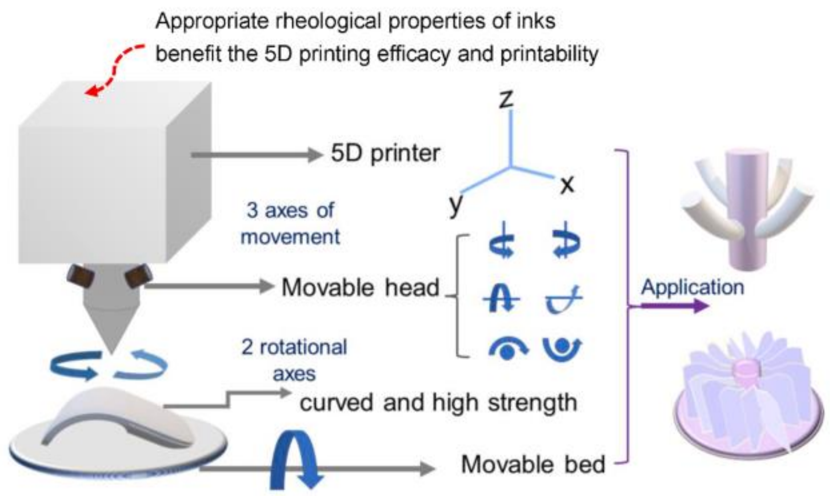


Figure 1 shows the additional dimension of 5D printing as schematically reported by Cheng et al. (2022).

Despite the impressive advances in 3DFP, it is surprising that researchers have significantly undervalued the original aim of additive manufacturing technology; Rapid prototyping (RP) and/or Rapid Tooling (RT). RP has been extensively employed by the plastic industry to identify design defects, shortening prototype development time (from weeks to days/hours) and cost for process building. RT, which involves the production of customized tools such as jings, hardware and molds, has become essential in increasing the efficiency of the manufacturing process in many industries. The cost and time benefits of RP/RT have been exemplified by Zonder and Sella (2013) that lowered production times from 30 days to seven hours and costs from 1400$ to 785$ when using 3D printing instead of building traditional aluminium molds. Also, RP and RT can facilitate product modification and customization for market segments with specific requirements. Even a very limited number of items, e.g., less than 10 pieces, might be produced by using 3D-printed molds without incurring prohibited costs, thus satisfying several heterogeneous needs. Also, RP allows the exploration of the design space, learning about design problems, the discovery of unexpected phenomena, and solving mechanical and thermal problems, among others (Rayna and Striukova, 2016; Camburn et al., 2017; Menold et al., 2017). For instance, Sun et al. (2021) used 3D printing to prototype and study the thermal performance of concrete buildings. Interestingly, they observed that the printing path and cross-section design of the internal structure determined the (in) homogeneous temperature distribution in the buildings. Similarly, Prasittisopin et al. (2019) have focused on the creation of building prototypes to optimize thermal insulation by changing surface texture and mortar mixture composition, while Lowke et al. (2018) introduced cavities in concrete building prototypes in regions where low thermal conductivity was needed to improve insulating properties. From a business perspective, RP and RT positively affect value creation, value proposition, and value delivery, contributing to the economic sustainability of a company (Rayna and Striukova, 2016). Other interesting implementations of RP and RT have been applied in tissue engineering (Liu et al., 2017), bone tissue prototypes (Farzadi et al., 2015; Belloti et al., 2021), chitosan hydrogels (Gang et al., 2022), orthodontic models (Venezia et al., 2022), and 3D micromixer-based microfluidic devices (Vasilescu et al., 2020). However, based on our knowledge no systematic analysis dedicated to the rapid prototyping of food products has been reported yet despite the many evident benefits of this approach, including but not limited to market innovation, increased food acceptance, customization of food properties, reduced production cost per piece, reduced processing times and energy requirements.

This study explores food prototyping by 3D printing technology. More specifically, prototypes of biscuits were produced by modulating their shape and internal morphology to assess their effects on the baking kinetics and identify the optimal structural layout capable of reducing baking time and formation of a potential process-induced toxicant, i.e., acrylamide.

2. Material and methods

2.1. Cereal-based printable food ink

A cereal-based dough consisting of wheat flour type 00 (Casillo, Italy) (120 g), semolina (Casillo, Italy) (120 g), extra virgin olive oil (Farchioni, Italy) (20g), sunflower oil (Coop, Italy) (24g), salt (2g), water (85g), baking powder (Paneangeli, Italy) (4g) and white wine (Tavernello, Italy) (30g) was prepared. Detailed information regarding the chemical composition of each ingredient may be found on the following links: 1. Wheat flour (https://shop.molinocasillo.com/it/farine-e-semole/le-essenziali/tipo-00); 2. Semolina (https://shop.molinocasillo.com/it/farine-e-semole/le-essenziali/semola-rimacinata); 3. Sunflower oil (https://www.e-coop.it/il-prodotto-coop/coop/olio-e-aceto/alimentari-confezionati/olio-di-semi/olio-di-semi-di-girasole-1); 4. White wine (www.tavernello.it); 5. Extra virgin olive oil (https://farchioni1780.com/olio-extra-vergine-oliva-classico/); 6. Baking powder (https://paneangeli.it/prodotti/lieviti-e-ingredienti-base/lieviti-dolci/lievito-pane-degli-angeli).

The ingredients in powder form were first combined and mixed for 2 min in a planetary kneader (Cooking Chef, Kenwood, UK) at a speed level of 2; then, the vegetable oils were added while continuously mixing for 2 min at a speed level of 2 and, finally, the water and wine were added and mixed for 1 min at a speed level of 1 allowing to obtain a dough, which was rested at room temperature for 10 min before 3D printing it.

2.2. Digital models, printing variables and G-codes

A total of 8 digital biscuits were designed by modulating their external and internal morphologies using the software Tinkercad (Autodesk, https://www.tinkercad.com/) and CURA ver 3.6.0 (Ultimaker, https://ultimaker.com/it/software/ultimaker-cura). The digital models were first designed in Tinkercad that allows generating STL files readable by CURA, which proceeded with the slicing of the digital models, the optimization of the printing variables and the generation of the G-codes containing the information for the printer head/bed movements and material depositions in the 3D space. More specifically, two external shapes - square and cylinder -, two infill levels - 30 and 60% - and two infill paths - grid and gyroid - were combined to create the eight digital models as reported in Figure 2. The choice of these printing conditions was based on our previous experience on the effects of infill level (from 0 to 100%) on several quality parameters of printed biscuits. Additionally, the selected infill paths, i.e., grid and gyroid, were chosen based on preliminary experiments aimed to test the printability and the structural differences obtained using different paths. Considering the different infill levels of the digital models, a preliminary series of printing experiments were performed, aiming to define the sample's dimensions to ensure comparable weights. All experiments were performed using a Delta printer (Mod. 2040, Wasproject, Italy). **Table 1** shows the main dimensions of each printed sample and their corresponding weights. These preliminary experiments allowed identifying the best printing conditions (e.g., layer height, printing speed, travel speed, retraction distance, retraction speed, etc.) capable of replicating the digital models with high accuracy. However, since assessing the effect of the printing variables selection on printing fidelity is not the main purpose of this study, this section only contains the values of the printing conditions relevant to each final sample.

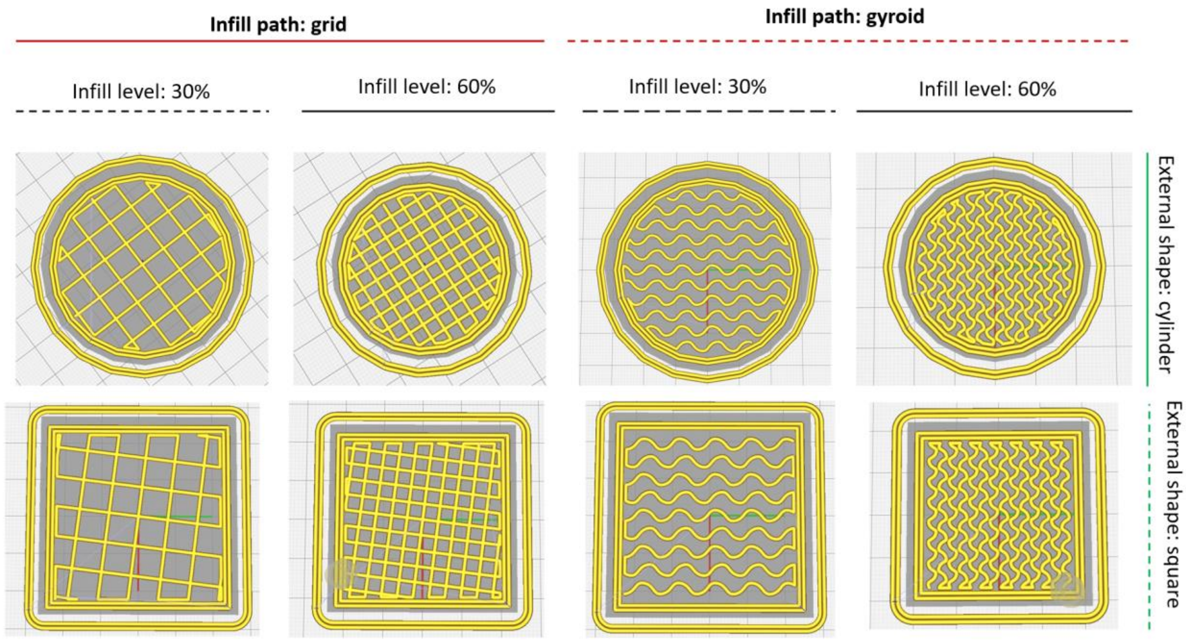


Figure 2: 3D digital models generated based on the combinations of the external shapes (cylinder and square), the infill levels (30% and 60%) and the infill paths (grid and gyroid) proposed.

In addition, we want to note that the control sample (i.e. no-printed) was manually shaped by using the same food formula as reported in section 2.1 obtaining samples cylinder of dimensions reported in Table 1.

Table 1 - Main morphological properties of 3D printed samples and corresponding weights

Sample's morphologies (shape_infill-path_infill-level)	Main dimension of the raw 3D printed biscuits (width * length * height, mm)	Weight (g)
Cylider_grid_30%	56 * 56 * 12	16.08 ± 1.13
Cylinder_grid_60%	45 * 45 * 12	16.97 ± 0.54
Square_grid_60%	45 * 45 * 12	16.32 ± 0.28
Square_grid_30%	50 * 50 * 12	17.17 ± 0.51
Cylinder_gyroid_30%	60 * 60 * 12	16.91 ± 0.36
Cylinder_gyroid_60	45 * 45 * 12	17.23 ± 0.55
Square_gyroid_60%	40 * 40 * 12	17.90 ± 0.75
Square_gyroid_30%	51 * 51 * 12	17.14 ± 0.44
Control (no Printed)	45 * 45 * 12	17.37 ± 0.33

2.3. Baking process and temperature data-acquisition

The 3D-printed biscuits were baked in a commercial oven (G10075, Ferrari, Italy) at 190°C for a total of 50 min. The biscuits were sampled at selected time intervals, namely after 1, 2, 3, 4, 5, 7, 10, 13, 16, 20, 25, 30, 35, 40, and 50 min of baking. The temperature of the biscuits was monitored, at least, at three different locations on the sample's surface using an infrared thermometer (Mod. 800C, Mestek, China). Also, at least three replicates were performed for each experiment.

2.4. Physico-chemical analyses

Moisture content was determined in triplicate according to the gravimetric method described in AOAC–925.10 (AOAC, 2005). The weight loss of the samples' during baking was determined using an analytical scale (Mod. Crystal 200, Gibertini, Italy). The water activity of the samples was estimated using the model reported by Arepally et al. (2022) who demonstrated the ability to accurately estimate water activity based on the moisture content of the biscuits.

2.5. Assessment of printing fidelity and color using image analysis

The printing fidelity was analyzed by comparing the main morphological properties of the 3D digital models and the printed samples. To this end, the images of the raw and baked 3D printed samples were acquired using a camera Canon, EOS 1200D and saved as jpeg files. After calibrating the image's dimension, the main external (length and width) and internal morphological characteristics (pore's dimension, distance between the crests of the filaments and their size) were calculated using ImageJ (https://imagej.nih.gov/ij/). Similar procedures have been used to measure such features for the 3D digital models in this study, which were directly captured on CURA and saved as jpeg files also. The color parameters, on the CIEL*a*b* scale, were extracted from the jpeg images using the embedded protocols for color determination in ImageJ. The color parameters were measured, at least, on 15 points of the sample after dividing the image into three main sections; external, medium and internal ring. Furthermore, the Browning index (BI) was computed by using the equation reported by Isleroglu et al., (2012):

$$BI = [100*((a_t+1.79L_t)/(5.645*L_t+at-3.102b_t))-0.31]/0.17 \quad (1)$$

Where a, b and L are red index, yellow index, and the lightness of the samples, respectively, at definite time (*t*) of baking.

2.6. Dehydration kinetics of the samples during baking

The dehydration kinetics during baking, expressed as moisture content as a function of time, was described using a logistic model (Corradini and Peleg (2006)):

$$Y(t) = Y0 - [A/1+\exp(k*(t_c-t))] \quad (2)$$

where Y0 (gH20/g d.b.) is the initial moisture content; A corresponds to the range of moisture content observed (Y0 - Yend); k (1/min) is the dehydration rate; Yend (gH20/d.b.) is the moisture content at equilibrium; and t_c (min) is the time at which dehydration intensifies.

In addition, the temperature profile during baking was described using a power law equation after slight modifications (Corradini and Peleg, 2004a; 2004b; 2005; 2007):

$$T(t) = T_0+(T_{end}-T_0)*(1-\exp(-k*t^n)) \quad (3)$$

where T_0 and T_{end} refer to the beginning of baking and at equilibrium, *k* is the heating rate (1/min), *n* a dimensionless shape parameter.

The experimental temperature profiles were fitted using the nonlinear regression routine of the software STATISTICA ver. 10 (Statsoft, USA). The correlation coefficient (r) and the confidence interval (95%) associated with each estimation were used to evaluate the goodness of fit. In terms of the dehydration kinetics, the model parameters of Equation 1 were initially obtained using a global estimation procedure (including all data in a single batch), and then, the data were also adjusted after grouping them based on a single morphological feature (i.e., external shape, infill level, infill path).

2.7. Estimations of acrylamide formation during baking of the 3D printed samples

Data on acrylamide generation kinetics, previously obtained for a similar baked good (Chen, et al. 2022), were used to extract the kinetic parameters assuming that the process could be described by the following empirical model (Equation. 4):

$$AA(t)= (AAf[T(t)] /(1 + Exp[k[T(t)]*(tf[T(t)]-t)) - (1/(1 + Exp[kf[T(t)* tf[T(t)])) \quad (4)$$

where AAf is the maximum level of acrylamide attainable (ng of acrylamide/g of biscuit), kf is the formation rate (1/min), and tf is the time required for effective formation (min). The temperature dependence of the respective kinetic parameters was described as:

$$AAf[T]= 684+Log[1+Exp[2.4*(T-207)] \quad (5)$$

$$k[T]= log[1+Exp[0.05*(T-184)] \quad (6)$$

$$tf[T]=8.6-0.024*T \quad (7)$$

The rate model of Equation 4 was solved in Mathematica 12 (Wolfram Corporation, Champaign, IL, USA) for the temperature profiles reported for all samples, i.e., control and 3D printed biscuits, after characterizing them with Equation 3. The rate equation was evaluated from t=0 min until the time corresponding to the target endpoint of baking, when the product's final moisture content of 0.1 g/100 g d.b. was reached. Although other models have been previously used to describe the formation and degradation of acrylamide in several products (Corradini and Peleg, 2006, Knol et al., 2006), Equation 4 provided a good characterization of the acrylamide content over time for this product, while also requiring fewer parameters. Hence, Equation 4 was preferred to obtain the estimations over more complex models.

3. Results and Discussion

3.1. Replicability of the 3D digital models and color changes during baking

The images of the 3D printed samples allowed evaluating the quality of printing in the context of overall visual appearance and the absence of major defects (Figure 3). The results of the image analysis are summarized in **Table 2**. The close correspondence between the actual and the digital designs is observed, even before and after baking.

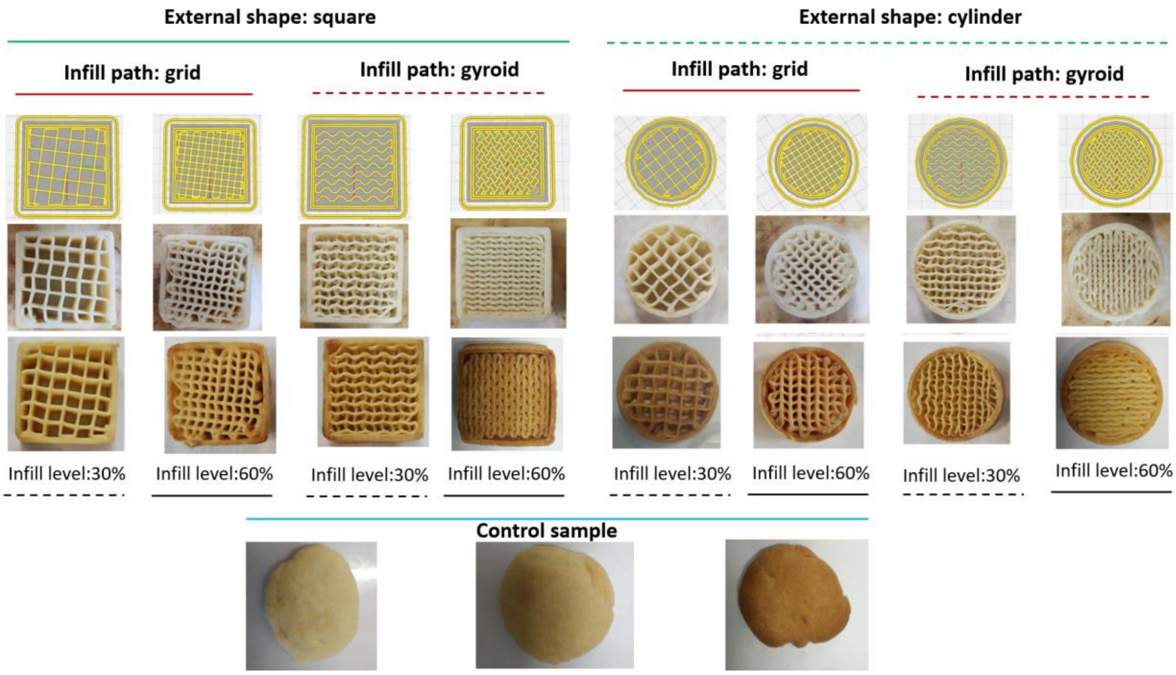


Figure 3 - Digital designs (top) and photographic images of the raw (middle), and baked (bottom) 3D printed biscuits. The images of the control samples are also reported.

Table 2 - Main morphological features of the 3D digital models and raw 3D printed samples

	Sample	3D digital model		3D printed samples	
		average	**Dev.st**	**average**	**Dev.st**
	Cylinder_grid_30%				
External length (mm)		56.07	0.16	53.95	0.28
Pore dimension (mm)		9.542	0.249	8.469	0.630
Filament size (mm)		1.200	0.001	1.190	0.076
	Cylinder_grid_60%				
External length (mm)		44.85	0.26	53.95	0.28
Pore dimension (mm)		3.992	0.035	8.469	0.630
Filament size (mm)		1.200	0.001	1.190	0.076
	Square_grid_30%				
External length (mm)		50.01	0.14	50.05	0.61
Pore dimension (mm)		2.905	0.063	6.082	0.354
Filament size (mm)		1.200	0.001	1.311	0.093
	Square_grid_60%				
External length (mm)		45.07	0.30	44.42	0.31
Pore dimension (mm)		2.905	0.061	2.719	0.130
Filament size (mm)		1.200	0.001	1.214	0.120
	Cylinder_gyroid_30%				
External length (mm)		60.36	0.08	57.89	0.56
Distance between throughs of two sinusoidal waves (mm)		10.267	0.282	8.743	0.399

Distance between crests of two sinusoidal waves (mm)	3.489	0.805	3.280	0.190
Filament size (mm)	1.200	0.001	1.704	0.108
Cylinder_gyroid_60%				
External length (mm)	44.76	0.10	43.53	0.47
Distance between throughs of two sinusoidal waves (mm)	5.222	0.068	4.835	0.163
Distance between crests of two sinusoidal waves (mm)	1.74	0.12	1.298	0.130
Filament size (mm)	1.200	0.001	1.345	0.160
Square_gyroid_30%				
External length (mm)	51.06	0.05	51.38	0.16
Distance between throughs of two sinusoidal waves (mm)	9.964	0.180	10.145	0.271
Distance between crests of two sinusoidal waves (mm)	3.625	0.901	3.208	0.255
Filament size (mm)	1.200	0.001	1.600	0.150
Square_gyroid_60%				
External length (mm)	38.90	0.10	38.63	0.38
Distance between throughs of two sinusoidal waves (mm)	5.102	0.231	4.571	0.152
Distance between crests of two sinusoidal waves (mm)	1.878	0.034	1.043	0.084
Filament size (mm)	1.200	0.001	1.219	0.145

The results confirm the high printing fidelity obtained in this study, with the external length of the biscuits showing less than 2.5 mm difference from their digital counterparts (total length = 40-60 mm depending on the selected layout). Similarly, the internal pores were precisely replicated with differences lower than 1 mm (largest pore size =9 mm for the cylinder_grid_30% samples). Furthermore, for the gyroid path, the distance between the troughs of two sinusoidal waves differed from the digital models by 0.181 mm and 1.524 mm (‘square_gyroid_30%’ and ‘cylinder_gyroid_30%’), while the distance between the crests of two waves was between 0.209 mm and 0.835 mm (‘cylinder_gyroid_30%’ and ‘square_gyroid_60%’). Finally, the thickness of the deposited filaments spread beyond the opening of the nozzle (size = 1.2 mm); this is in accordance with the observations of several authors (Derossi et al., 2020; Paolillo et al., 2021). Although this spread was not surprising, the values observed exceeded those expected. A layer height of approximately 80% of the nozzle size (from 0.9 to 0.96 mm depending on the digital models) was selected to deposit food filaments with a rectangular cross-sectional shape instead of a cylinder so that the overall structural stability of the overlying layers significantly increased. Under these circumstances, it was reasonable to observe a filament size greater than 1.2 mm, as reported in **Table 1**.

The overall color of the biscuits during baking was analyzed as the evolution of BI as a function of baking time in three different regions of the samples, i.e. external, medium and internal ring. The results agree with previous experiments (Isleroglu, 2012) which showed a slight increase in the biscuit’s color during the first minutes of baking (Shibukawa, 1989) after which the samples exhibited a sudden increase of BI due to a significant reduction of the lightness (Shibikawa, 1989) in line with the increase of surface temperature (Arepally, 2020). Also, once a maximum was reached, approximately at 30 min, BI started to decrease.

3.2. Dehydration kinetics - description and validation experiments

The moisture content of the biscuits during baking was effectively described by Equation. 1. The non-linear regression was performed first using a global estimation procedure, i.e., all data in a single batch. Then, the data was grouped based on the external shape, infill level and infill path, which required applying the fitting routine 8 times and comparing the parameters of the model to determine which morphological feature had the largest impact on the dehydration kinetics (Figure 4). The baking time required to achieve a final moisture content equal to or below 0.1 g/g d.b. varied from 14 to 40 min, revealing a large difference in the behavior of the samples depending on their main morphological features (Figure 4a). Indeed, when the fitting was performed on all the data as a single batch, a lower correlation coefficient ($r = 0.950$) was obtained than when the model was applied to all the 3DP samples ($r = 0.963$). This discrepancy provides evidence of the significant differences in the dehydration kinetics between 3DP and traditional (non-3D printed) biscuits. Furthermore, the goodness of fit measures of the nonlinear regression indicates that infill level was a better criterion to group the samples than the other selected characteristics (correlation coefficients of 0.987 and 0.986 for infill levels of 60% and 30%, respectively). It should also be noted that, when the data were grouped by infill path, a higher correlation coefficient was obtained for the gyroid path ($r = 0.972$) while when considering the external shape, the cylindrical samples showed a higher correlation coefficient than the square samples (0.969 vs. 0.957). These results emphasize a large divergence in the baking behavior of the traditional and 3DP samples and, the importance of the infill level followed by the infill path, in particular, for the gyroid set-up in obtaining consistent results.

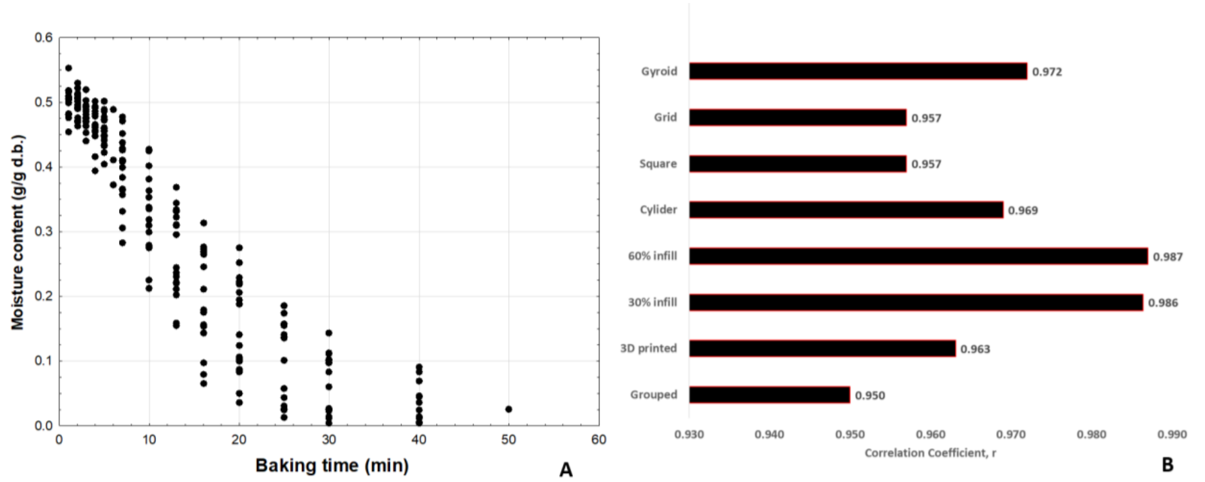


Figure 4: Dehydration kinetics of biscuits during baking. A) Moisture content of all samples as a function of time. B) Correlation coefficients, *r*, of the fits for the experimental data grouped by shape, infill level and infill path.

A second iteration of the fitting procedure was performed by grouping the data individually by morphology, as reported in Figure 4b. In this case, the kinetic parameters - the dehydration rate, k, and the critical time, t_c - were estimated and compared to determine the contributions of the external and internal structure to the dehydration kinetics of the samples. The experimental data and the fits of the cylindrical 3D-printed biscuits with different infill levels are reported in Figure 5 to exemplify the results obtained. Based on the visual verification and the correlation coefficients which were consistently between 0.987 and 0.994, it can be concluded that

Equation. 1 effectively described the dehydration of such samples during baking at 190°C (data not shown).

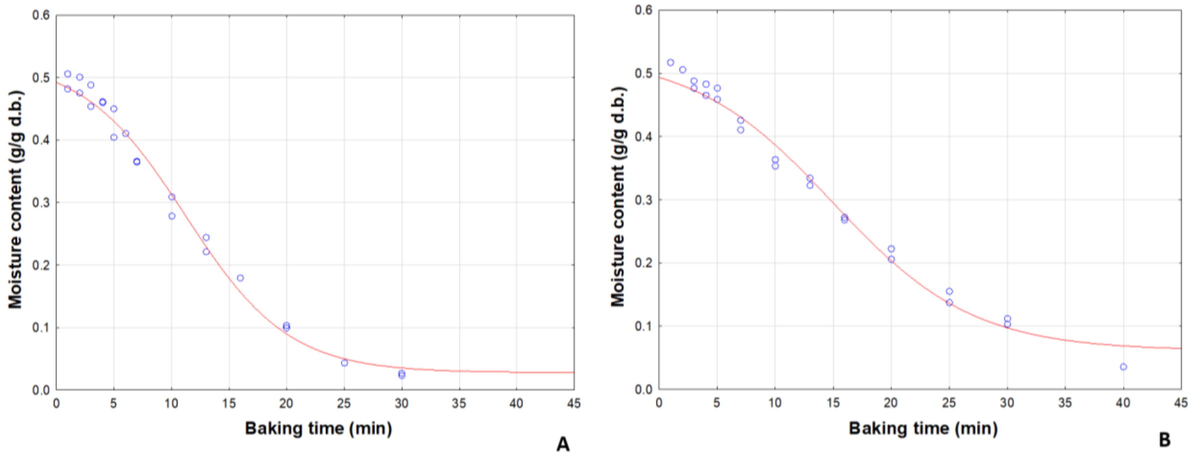


Figure 5: Moisture content of cylindrical 3D printed biscuits as a function of time. A) infill level = 30%, infill path: grid; B) infill level = 60%, infill path: grid.

An additional analysis was performed to compare the dehydration process in samples with different morphology based on the parameters of Equation. 1. Figure 6 shows the estimated dehydration, *k*, and the critical time, tc, for each sample.

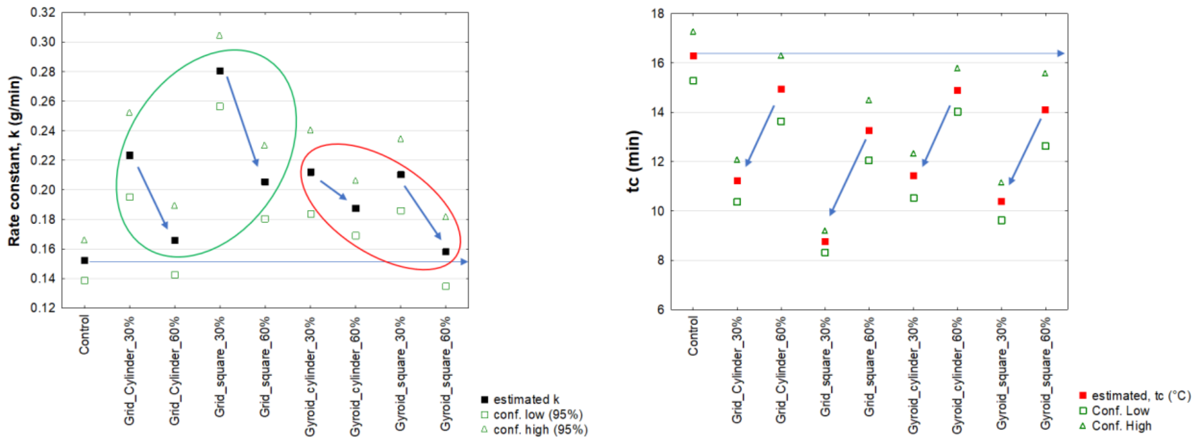


Figure 6: Estimated dehydration kinetics parameters, *k* and *tc*, including the CI (0.95) of the 3D printed samples with different morphological characteristics baked at 190 °C.

The control samples baked slower than any other of the 3D printed samples (i.e., it exhibited the lowest dehydration rate constant, 0.152 g/g d.b. * min, and required the longest time to achieve the target final moisture content, 16.3 min). Also, for both parameters, the limited or lack of overlapping of confidence intervals (95%) confirms that the control behaved significantly different than the 3D printed samples. From Figure 7, it can also be inferred the substantial effect of the infill level on baking time, based on the significantly higher dehydration rates for biscuits with 30% of infill, regardless of their external shape or internal path. Indeed, estimated k values for the 'cylinder_grid_30%', 'square_grid_30%', 'cylinder_gyroid_30%' and 'square_gyroid_30%' samples were 0.224, 0.280, 0.212 and 0.210 g/g*min, respectively. On the other hand, the samples with higher infill levels (i.e., 60%)

exhibited lower k values such as 0.165, 0.205, 0.187, and 0.158 g/g*min that corresponded to the ‘cylinder_grid_60%’, ‘square_grid_60%’, ‘cylinder_gyroid_60%’ and ‘square_gyroid_60%’ samples. The opposite behavior was observed for the other parameter of Equation. 2; *tc*, with lower values (8.74 to 11.4 min) for samples with an infill level of 30% than those with 60% infill (13.3 to 15.0 min). This suggests that, for 3D printed samples with an infill of 30%, the baking became more effective than samples with an infill of 60%.

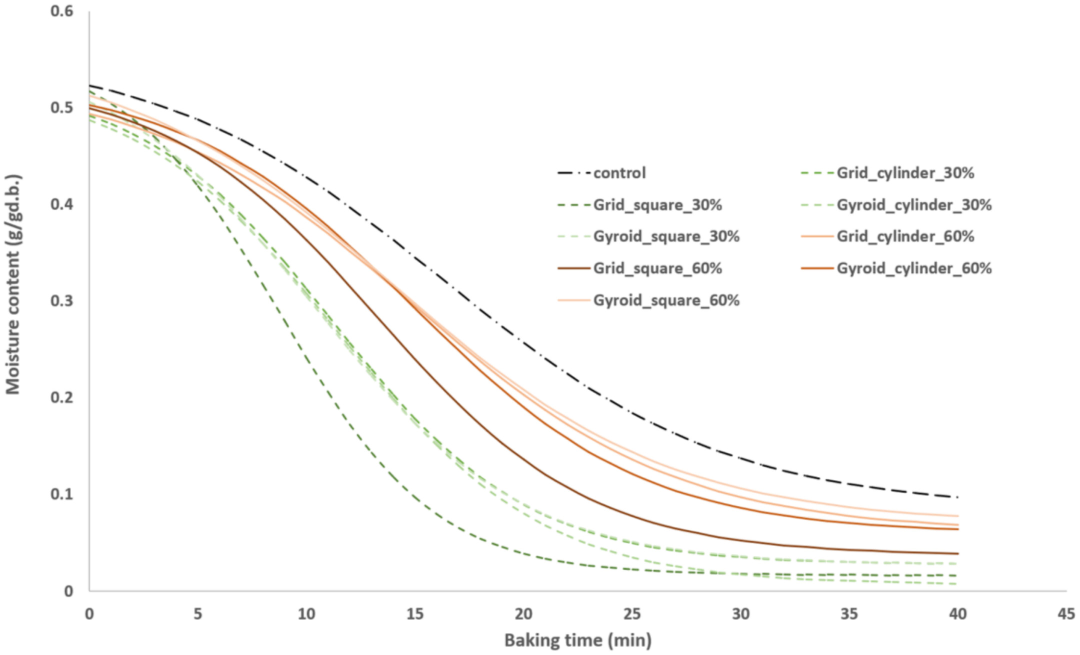
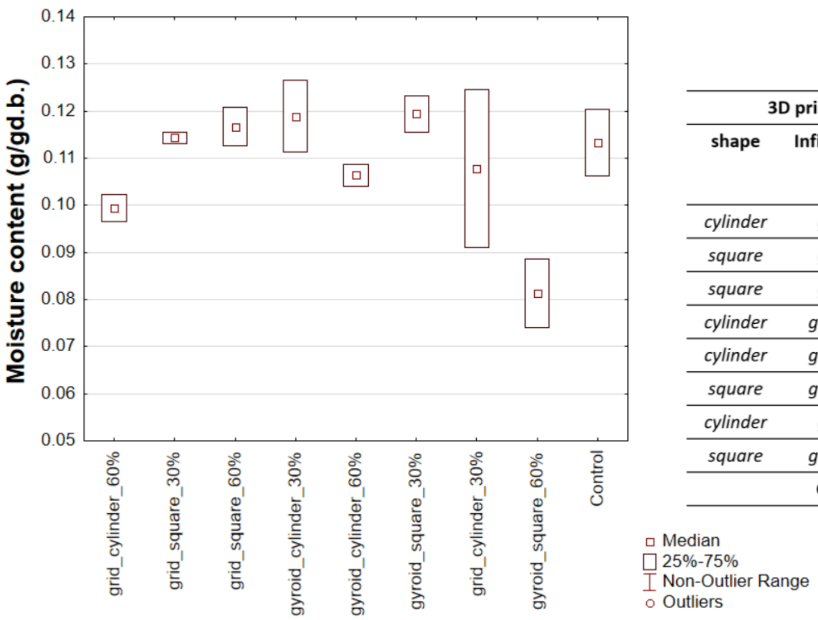


Figure 7: Moisture content vs time relationships generated using the estimated kinetic parameters from Equation. 1.

Further valuable information was obtained by grouping the samples based on the infill paths; indeed, ‘grid’ samples (green circles) showed large variability in their dehydration rates (between 0.165 and 0.280 g/g*min). This observation suggests that the rational design of the infill level (and the morphological features of a food matrix) provides an opportunity to better modulate the baking process depending on what the user would like to obtain, e.g. reduced baking time, controlled formation of undesirable chemical compounds, preservation of heat-sensitive nutrients, etc. Conversely, in the high-filled samples when using the path ‘gyroid’ (red circles), the dehydration rates concentrated around a narrower range (*k* values from 0.158 g/g*min to 0.212 g/g*min). In addition, considering the confidence intervals (95%) associated with these estimates only statistical differences were observed in the case of ‘gyroid_square_30%’ and ‘gyroid_square_60%’ samples.

Also, it is worth noting that among the printed samples having 60% infill, the shape that better enabled speeding up baking was the square with a grid infill path. To facilitate comparing how the morphological features can modify the baking process, all the moisture content vs time relationships were estimated using Equation. 1 is reported in Figure 7. According to these results, the control achieved a moisture content of 0.1 g/g d.b. after about 40 min of baking, much slower than any of the 3D printed samples. Conversely, the 3D printed samples, and especially those with an infill level of 30% (dotted lines) attained the targeted moisture content much faster, i.e., between 15- and 32-min. Intermediate velocities were observed for samples printed with 60% infill. Interestingly, the samples with the fastest baking for both infill levels (30 and 60%) corresponded to the samples with a squared geometry and a grid path.

Finally, considering that this study focused on using 3DFP to create food prototypes with different geometrical features and evaluating their effects and potential benefits, the time needed to reach a final moisture content of 0.1 g/ d.b. was estimated. This target value was chosen considering an estimated water activity lower than 0.4 and the range of moisture content observed in other oven-baked commercial biscuits (Mundt and Wedzicha, 2007). Figure 8 shows the estimated baking time of all external and internal geometry combinations of the 3D printed samples and the control. The measured moisture content of the samples in the validation experiments is also shown in Figure 8.



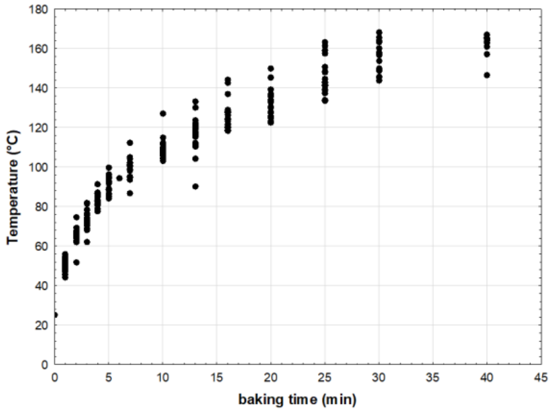
3D printed samples			
shape	**Infill path**	**Infill level (%)**	**Estimated baking time (min)**
cylinder	*grid*	*60*	29.60
square	*grid*	*30*	16.50
square	*grid*	*60*	22.63
cylinder	*gyroid*	*30*	18.60
cylinder	*gyroid*	*60*	27.40
square	*gyroid*	*30*	19.00
cylinder	*grid*	*30*	19.19
square	*gyroid*	*60*	30.05
	Control		38.60

Figure 8: Estimated baking time to obtain a moisture content of 0.1 g/g d.b (right) and experimental data (left) measured on biscuits obtained from the validation experiments.

The data confirm the large differences between the samples, especially the longer baking time of the control (i.e., 38.60 min). All the 3D printed samples showed a shorter baking time with a minimum of 16.50 min for ‘square_grid_30%’ and a maximum of 30.05 min for the ‘square_gyroid_60%’. Again, according to the estimated *k* and *tc* values, for all samples with 30% infill, shorter baking times were estimated - 19.19, 16.50, 18.60 and 19.00 min - while a 60% infill resulted in an increase in baking time between 22.63 and 30.05 min. A good correlation between estimated and experimental moisture content was verified through validation (Figure 8). For example, for the shortest baking time of 16.50 min for samples ‘square_grid_60%’, the experimental moisture content values were between 0.1130 g/g d.b. (25%) and 0.1150 g/g d.b. (75%). The highest variability was obtained for the sample ‘cyliinder_grid_30%’ for which experimental moisture content ranged from 0.091 g/g d.b. (25%) to 0.1245 g/g d.b. (75%). In all cases, the measured moisture content was very close to the desired value of 0.1 g/g d.b. These results largely support the importance and opportunity of using 3D food printing to generate food prototypes that could be used and tested to modulate the dehydration kinetics of a baked product, for instance, to reduce the baking time, energy consumption, the overall cost of production while potentially increasing the amount of baked product per unit of time. Besides increasing the economic sustainability of the production process, the improvement of the baking process by modulating the structural characteristics of a product can also achieve other benefits such as limiting or favoring chemical reactions that can lead to nutrient degradation, toxicants formation, color changes, etc., to mention only a few. This opportunity will be further explored in the next section.

3.3. Estimating acrylamide formation during baking

Acrylamide is a processing-induced contaminant found in a wide selection of foods. Consumption of cereal-based food products, such as biscuits, can play a significant role in dietary acrylamide exposure (Rifai and Saleh, 2020). Due to acrylamide's classification as a potential carcinogen, acrylamide reduction in baked goods has become a growing interest in the food industry. Acrylamide is a by-product of the Maillard reaction, and it is formed when food products are processed at temperatures above 120°C. Mitigation strategies to reduce acrylamide content in cereal-based food products include preheating treatments of the raw materials, incorporation of additives, enzymatic removal of reagents (e.g., asparagine by asparaginase). Optimizing processing parameters has also been proposed as an effective strategy to reduce acrylamide content in a product. Within this context, 3DFP can play an effective role in identifying food product's characteristics (shape, infill level, infill path) that minimize acrylamide formation. To this end, the rate model of Equation. 3 was solved for the temperature profiles of all the samples. Figure 9 shows the temperature profiles evaluated after characterizing them with Equation. 3. Figure 10 provides a comparison of the temperature profiles and their respective acrylamide formation curves for the control and two examples of the 3D-printed biscuits. As shown in the figure, selecting different infills reduced the baking requirements (time and temperature) to reach the target moisture content, which in turn had significant effects on the potential safety of the biscuit. **Table 3** summarizes the overall acrylamide formation in ng/g and the percentage reduction of all the samples towards the control. All the 3D-printed samples showed a significant reduction in acrylamide content when compared to the control. It should be noted that all the estimated acrylamide values but the control, are below the benchmark for biscuits established by the EU Commision, Regulation 2017/2158, i.e., 350 ng/g. Among the 3D printed samples, the square with 30% infill, regardless of the infill path, exhibited the lowest acrylamide content. Hence, prototyping by using 3DFP could enhance the chemical safety of a food product by elucidating ways to mitigate the formation of potentially toxic substances without the need of additives or changes in composition. It should be noted that the reported values are estimations, not predictions since the purpose of this section was to advance the understanding of 3DP prototyping for food safety applications. Thus, the results have not been independently validated, which will be the subject of future work. Also, the acrylamide content estimations were conducted based on a model that takes into consideration the most predominant contributions to its formation and degradation process, i.e., availability of main reagent and temperature. Although the contribution of the structural components of a food to acrylamide kinetics might play a role, it has not been systematically studied or expressed into a validated model. Limited information is available on this contribution (e.g., Pulatsu, et al. 2022). This is an area of active research and future studies will potentially elucidate the extent of this contribution to the overall acrylamide content in a baked good.



Sample	k (1/min)	std.error	p-value	Conf. low	Conf. high	n	std.err	p-value	Conf. low	Conf. high	r
Control	0.187	0.011	0.0001	0.164	0.209	0.641	0.023	0.0001	0.593	0.689	0.989
Cylinder_grid_30%	0.198	0.016	0.0001	0.165	0.231	0.671	0.034	0.0001	0.600	0.742	0.991
Cylinder_grid_60%	0.219	0.011	0.0001	0.197	0.242	0.644	0.022	0.0001	0.599	0.689	0.995
Cylinder_gyroid_30%	0.197	0.015	0.0001	0.167	0.227	0.645	0.031	0.0001	0.581	0.709	0.987
Cylinder_gyroid_60%	0.198	0.010	0.0001	0.178	0.218	0.603	0.019	0.0001	0.564	0.642	0.989
Square_grid_30%	0.176	0.017	0.0001	0.141	0.210	0.645	0.038	0.0001	0.565	0.724	0.994
Square_grid_60%	0.182	0.008	0.0001	0.166	0.198	0.663	0.017	0.0001	0.628	0.698	0.986
Square_gyroid_30%	0.180	0.013	0.0001	0.152	0.208	0.659	0.031	0.0001	0.595	0.723	0.993
Square_gyroid_60%	0.176	0.011	0.0001	0.154	0.198	0.632	0.023	0.0001	0.584	0.680	0.990

B

Figure 9: Temperature profiles of all samples during baking and estimated parameters of the Equation 3.

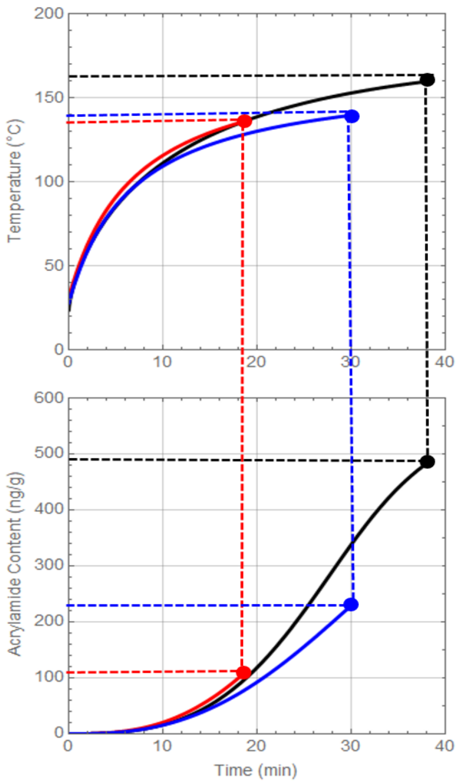


Figure 10: **-** Top- Temperature profiles for the differently shaped biscuits, Bottom – Acrylamide content in ng/g in selected biscuits. Black –control, red – Square_gyroid_30%, blue – Square_gyroid_60%. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 3 - Estimated acrylamide content and its reduction towards the control of all the biscuit prototypes

Sample	Acrylamide content (ng/g)	Reduction in acrylamide content (%)
Control	495 ± 6	-
Cylinder_grid_30	142 ± 65	71 ± 13
Cylinder_grid_60	301 ± 74	39 ± 15
Square_grid_30	47 ± 2	91 ± 1
Square_grid_60	168 ± 52	66 ± 11
Cylinder_gyroid_30	91 ± 9	82 ± 2
Cylinder_gyroid_60	210 ± 12	58 ± 3
Square_gyroid_30	85 ± 11	83 ± 2
Square_gyroid_60	220 ± 24	56 ± 5

4. Conclusions

Despite the recent interest and developments in 3D food printing applications, its adoption as a prototyping tool, capable of identifying the optimal shapes and internal structures to attain desired functionalities has been limited. This study has shown that 3D printing technology can be successfully used to experiment and define optimal 3D food architectures capable of minimizing the baking time of biscuits. All 3D printed samples baked significantly faster (from 16.50 to 30.05 min) than the control (38.60 min). Infill level influenced the baking kinetics the most, with faster dehydration rates and shorter critical times for samples with lower fills (30%). When the printing movements drew a 'grid' path, the samples exhibited wider variability in terms of their dehydration kinetics, depending on infill levels and external shapes; this could be used as an extended experimental space to modulate baking conditions and related effects, e.g. baking time, formation of chemical compounds, color/aroma development, etc. The logistic model adequately described the baking kinetics for all 3D printed samples and the control, allowing to accurately estimate the baking time to attain a moisture content of 0.1 g/g d.b. In addition, a second series of experiments accurately validated these estimates, corroborating the possibility of designing biscuits with different desired properties. For the first time, the feasibility of modulating chemical reactions in foods by exploiting the unique properties of 3D food printing technology was explored. For the 3D-printed biscuits, a significant reduction (39-91%) in the acrylamide content was obtained, corresponding to values lower than the recognized EU Commission limit of 350 ng/g. Conversely, control samples exhibited an estimated value of 495 ng/g. This could open the applicability of 3D printing to the improvement of chemical safety of food products by designing the shape and internal structure - thanks to the rapid food prototyping obtained by 3D printing technology. It is worth noting the possibility of extending this concept by prototyping food structures to design desired color, aroma compounds, texture, mass/heat transfer rates during dehydration/rehydration, etc. All these aspects will be the core of novel experiments in the future.

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CRediT authorship contribution statement

AD: Conceptualization, Methodology, Investigation, Formal analysis; Writing, Review & Editing. RC: Methodology, Investigation, Formal analysis, Review & Editing, MGC: Methodology, Writing, Review & Editing; OMO: Methodology, Formal analysis, Investigation; C.S.: Resources, Review & Editing.

Literature

1. Arepally, D., Reddy, R.S., Goswami, T.K., & Datta, A.K. (2020). Biscuit baking: A review. LWT – Food Science and Technology, 131, 109726.

2. Armstrong, C.D., Yue, L., Deng, Y., & Qi, H.J. (2022). Enabling direct ink write edible 3D printing of food purees with cellulose nanocrystals. Journal of Food Engineering, 330, 111086, https://doi.org/10.1016/j.jfoodeng.2022.111086.

3. Attaran, S., & Attaran, M. (2020). Food printing: Evolving technologies, challenges, opportunities, and best adoption strategies. Journal of International Technology and Information Management, 29(1), 25–55.

4. Belloti, J.C., Alves, B.V.P., Faloppa, F., & Balbachevsky, D., Archetti Netto, N., Jun Tamaoki, M. (2021). The malunion of distal radius fracture: Corrective osteotomy through planning with prototyping in 3D printing. Injury, 52, 3, S44-S48, https://doi.org/10.1016/j.injury.2021.05.048.

5. Camburn, B., Viswanathan, V., Linsey, J., Anderson, D., Jensen, D., Crawford, R., Otto, K., & Wood, K. (2017). Design prototyping methods: State of the art in strategies, techniques, and guidelines. Design Science, 3 (13), https://doi.org/10.1017/dsj.2017.10

6. Champeau, M., Heinze, D.A., Viana, T.N., De Souza, E.R., Chinellato, A.C., & Titotto, S. (2020). 4D Printing of Hydrogels: A Review. Advanced Functional Material, 30, 1910606, 10.1002/adfm.201910606

7. Chen, F., Zhang, M., Liu, Z., Bhandari, B., 2021b. 4D deformation based on double-layer structure of the pumpkin/paper. J. foosrt. 100168. 10.1016/j.foostr.2020.100168.

8. Chen, Y., Zhang, M., Sun, Y., & Phuhongsung, P. (2022). Improving 3D/4D printing characteristics of natural food gels by novel additives: A review. Food Hydrocolloids, 123 107160, https://doi.org/10.1016/j.foodhyd.2021.107160.

9. Chen, Y., Zhang, M., Sun, Y., & Phuhongsung, P. (2022). Improving 3D/4D printing characteristics of natural food gels by novel additives: A review. Food Hydrocolloids, 123, 107160, https://doi.org/10.1016/j.foodhyd.2021.107160.

10. Chen, C., Zhang, M., Guo, C., & Chen, H. (2021). 4D printing of lotus root powder gel: Color change induced by microwave. Innovative Food Science and Emerging Technologies, 68, 102605, 10.1016/j.ifset.2021.10260.

11. Chen, C., Espinal-Ruiz, M., Joye, I.J., Corradini, & M.G. (2022). Modeling temporal-spatial distribution of AA and color during the cookie baking process – a numerical simulation. Proceedings of Conference of Food Engineers (COFE) 2022, Rayleigh, NC, USA.

12. Chen, C., Zhang, M., Mujumdar, A.S., & Phuhongsung, P. (2021). Investigation of 4D printing of lotus root-compound pigment gel: Effect of pH on rapid colour change. Food Research International, 148, 110630, 10.1016/j.foodres.2021.110630

13. Cheng, Y., Fu, Y., Ma, L., Yap, P.L., Losic, D., Wang, H., & Zhang, Y. (2022). Rheology of edible food inks from 2D/3D/4D printing, and its role in future 5D/6D printing. Food Hydrocolloids, 132, 107855, https://doi.org/10.1016/j.foodhyd.2022.107855.

14. Cheng, Y., Fu, Y., Ma, L., Yap., P.L., Losic, D., Wang, H., & Zhang, Y. (2022). Rheology of edible food inks from 2D/3D/4D printing, and its role in future 5D/6D printing. Food Hydrocolloids, 132, 107855, https://doi.org/10.1016/j.foodhyd.2022.107855.

15. Corradini, M. G., & Peleg, M. (2004a). A model of non-isothermal degradation of nutrients, pigments and enzymes. Journal of the Science of Food and Agriculture, 84(3), 217e226.

16. Corradini, M. G., & Peleg, M. (2004b). A model of non-isothermal degradation of nutrients, pigments and enzymes.Journal of the Science of Food and Agriculture, 84(3), 217–226.

17. Corradini, M. G., & Peleg, M. (2004b). Demonstration of the Weibull-Log logistic survival model's applicability to nonisothermal inactivation of E. coli K12 MG1655. Journal of Food Protection, 67(11), 2617–2621.

18. Corradini, M. G., & Peleg, M. (2005). Estimating non-isothermal bacterial growth in foods from isothermal experimental data. Journal of Applied Microbiology, 99(1), 187e200.

19. Corradini, M. G., & Peleg, M. (2006). Prediction of vitamins loss during non-isothermal heat processes and storage with non-linear kinetic models. Trends in Food Science & Technology, 17(1), 24e34.

20. Corradini, M.G., & Peleg, M. (2007). Shelf-life estimation from accelerated storage data. Trends in Food Science & Technology, 18, 37-47.

21. Derossi, A., Caporizzi, R., Oral, M.O., & Severini, C. (2020). Analyzing the effects of 3D printing process per se on the microstructure and mechanical properties of cereal food products, Innovative Food Science & Emerging Technologies. 66, 102531, https://doi.org/10.1016/j.ifset.2020.102531.

22. Derossi, A., Caporizzi, R., Paolillo, M., Oral, M.O., Severini, C. (2021). Drawing the scientific landscape of 3D Food Printing. Maps and interpretation of the global information in the first 13 years of detailed experiments, from 2007 to 2020. Innovative Food Science & Emerging Technology, 70.

23. Derossi, A., Caporizzi, R., Ricci, I., Severini, C. Critical variables in 3D Food Printing, Editors: Godoi, F.C., Bhandari, B., Prakash, S., Zhang, M. Fundamentals of 3D Food Printing and Applications, Academic press, 2019, 41-91, https://doi.org/10.1016/B978-0-12-814564-7.00003-1.

24. Dick, A., Bhandari, B., & Prakash, S. (2019). 3D printing of meat. Meat Science, 153, 35-44, https://doi.org/10.1016/j.meatsci.2019.03.005.

25. Fahmy, A.R., Amann, L.S., Dunkel, A., Frank, O., Dawid, C., Hofmann, T., Becker, T., & Jekle, M. (2021). Sensory design in food 3D printing ng – Structuring, texture modulation, taste localization, and thermal stabilization, Innovative Food Science & Emerging Technologies, 72, 102743, https://doi.org/10.1016/j.ifset.2021.102743

26. Farzadi, A., Waran, V., Solati-Hashjin, M., Rahman, Z.A.A., Asadi, M., & Osman, N.A.A. (2015). Effect of layer printing delay on mechanical properties and dimensional accuracy of 3D printed porous prototypes in bone tissue engineering. Ceramics International, 41, 7, 8320-8330, https://doi.org/10.1016/j.ceramint.2015.03.004.

27. Feng, T., Fan, c., Wang, X., Wang, X., Xia, S., & Huang, Q. (2022). Food-grade Pickering emulsions and high internal phase Pickering emulsions encapsulating cinnamaldehyde based on pea protein-pectin-EGCG complexes for extrusion 3D printing. Food Hydrocolloids, 24 (A), 107265, https://doi.org/10.1016/j.foodhyd.2021.107265.

28. Gang, F., Ma, C., Guo, C., Shi, R., & Xiao, Y. (2022). Integrated rapid-prototyping and strength-increasing all-in-one 3D printing of chitosan hydrogels. Materials Letters, 323, 132542, https://doi.org/10.1016/j.matlet.2022.132542.

29. Ghazal, A.F., Zhang, M., Bhandari, B., & Chen, H. (2021). Investigation on spontaneous 4D changes in color and flavor of healthy 3D printed food materials over time in response to external or internal pH stimulus. Food Research International, 142, 110215, 10.1016/j.foodres.2021.110215

30. Ghazal., A.F., Zhang, M.M., Mujumdar, A.S., & Ghamry, M. (2022). Progress in 4D/5D/6D printing of foods: applications and R&D opportunities. Critical Reviews in Food Science and Nutrition, https://doi.org/10.1080/10408398.2022.2045896

31. Guo, C. Zhang, M., & Devahastin, S. (2021). Improvement of 3D printability of buckwheat starch-pectin system via synergistic Ca2+-microwave pretreatment. Food Hydrocolloids, 113, 106483, https://doi.org/10.1016/j.foodhyd.2020.106483

32. Guo, C., Zhang, M., & Devahastin, S. (2021). Color/aroma changes of 3D-Printed buckwheat dough with yellow flesh peach as triggered by microwave heating of gelatin-gum Arabic complex coacervates. Food Hydrocolloids, 112, 106358, https://doi.org/10.1016/j.foodhyd.2020.106358

33. He, C., Zhang, M., & Guo, C. (2020). 4D printing of mashed potato/purple sweet potato puree with spontaneous color change. Innovative Food Science & Emerging Technology, 59, 10.1016/j.ifset.2019.102250

34. Isleroglu, H., Kemerli, T., Yilmazer-Sakin, M., Guven, G., Ozdestan, O., Uren, A., & Kaymak-Ertekin, F. (2012). Effect of stema baking on acrylamide formation and browning kinetics of cookies. Journal of Food Science, n.0, 10.1111/j.1750-3841.2012.02912.x.

35. Kim, H.W., Lee, I.J., Park, S.M., Lee, J.H., Nguyen, M.H., Park, H.J. (2019). Effect of hydrocolloid addition on dimensional stability in post-processing of 3D printable cookie dough. LWT, 101, 69-75, https://doi.org/10.1016/j.lwt.2018.11.019.

36. Kim, S.M., Woo, J.H., Kim, H.W., & Park, H.J. (2021). Formulation and evaluation of cold-extruded chocolate ganache for three-dimensional food printing. Journal of Food Engineering, 314 110785, https://doi.org/10.1016/j.jfoodeng.2021.110785.

37. Kumbhar, B.K., Agarwal, R.S., & Das, K. (1981). Thermal properties of fresh and frozen fish. International Journal of Refrigeration, 4(3), 143-146.

38. Ling, K.C.L., Yee, A.Z.H., Leo, C.H. Chua, C.K. (2022). Understanding 3D food printing technology: An affordance approach. Materials Today: Proceedings. https://doi.org/10.1016/j.matpr.2022.08.564.

39. Liu, W., Wang, D., Huang, J. Wei, Y., Xiong, J., Zhu, W., Duan, L., Chen, R., & Wang, D. (2017). Low-temperature deposition manufacturing: A novel and promising rapid prototyping technology for the fabrication of tissue-engineered scaffold. Materials Science and Engineering: C, 70 (2), 976-982,https://doi.org/10.1016/j.msec.2016.04.014
40. .
41. Liu, Z., Bhandari, B., Prakash, S., Mantihal, S., & Zhang, M. (2019). Linking rheology and printability of a multicomponent gel system of carrageenan-xanthan-starch in extrusion based additive manufacturing. Food Hydrocolloids, 87, 413-424, https://doi.org/10.1016/j.foodhyd.2018.08.026.

42. Liu, Z., He, C., Guo, C., Chen F., Bhandari, B. & Zhang, M. (2021). Dehydration-triggered shape transformation of 4D printed edible gel structure affected by material property and heating mechanism. Food Hydrocolloids. 115, 106608, 10.1016/j.foodhyd.2021.106608

43. Lowke, D., Dini, E., Perrot, A., Weger, D., Gehlen, C., & Dillenburger, B. (2018). Particle-bed 3D printing in concrete construction – Possibilities and challenges. Cement and Concrete Research, 112, 50-65, https://doi.org/10.1016/j.cemconres.2018.05.018

44. Mattas, K., Tsakiridou, E., Karelakis, C., Lazaridou, D., Gorton, M., Filipovic, J., Hubbard, C., Saidi, M., Stojkovic, D., Tocco, B., Tregear, A., & Veneziani, M. (2022). Strengthening the sustainability of European food chains through quality and procurement policies. Trends in Food Science &Technology, 120, 248-253. https://doi.org/10.1016/j.tifs.2021.11.021

45. Menold, J. (2017). Prototype for X (PFX): A prototyping framework to support product design. The Pennsylvania State University.

46. Mundt, S., & Wedzicha, B.L. (2007). A kinetic model for browning in the baking of biscuits: effects of water activity and temperature. LWT – Food Science and Technology, 40, 1078-1082. doi:10.1016/j.lwt.2006.07.011.

47. Nijdam, J.J., Agarwal, D., & Schon, B.S. (2021). Assessment of a novel window of dimensional stability for screening food inks for 3D printing. Journal of Food Engineering, 292, 110349, https://doi.org/10.1016/j.jfoodeng.2020.110349.

48. Oral, M.O., Derossi, A., Caporizzi, R., & Severini, C. (2021). Analyzing the most promising innovations in food printing. Programmable food texture and 4D foods. Future Foods, 4, 100093, https://doi.org/10.1016/j.fufo.2021.100093.

49. Oral, O.M., Derossi, A., Caporizzi, R., & Severini, C. (2021). Analyzing the most promising innovations in food printing. Programmable food texture and 4D foods. Future Foods, 4, 100093,https://doi.org/10.1016/j.fufo.2021.100093.

50. Paolillo, M., Derossi, A., van Bommel, K., Noort, M., & Severini, C. (2021). Rheological properties, dispensing force and printing fidelity of starchy-gels modulated by concentration, temperature and resting time. Food Hydrocolloids, 117,106703, https://doi.org/10.1016/j.foodhyd.2021.106703.

51. Phuhongsung, P., Zhang, P., & Devahastin, S. (2020). Influence of Surface pH on Color, Texture and Flavor of 3D Printed Composite Mixture of Soy Protein Isolate, Pumpkin, and Beetroot. Food and Bioprocess Technology, 13 (9), 1600-1610, 10.1007/s11947-020-02497-8

52. Prasittisopin, L., Pongpaisanseree, K., Jiramarootapong, P., & Snguanyat, C. (2020). Thermal and Sound Insulation of Large-Scale 3D Extrusion Printing Wall Panel. In: Bos, F., Lucas, S., Wolfs, R., Salet, T. (eds) Second RILEM International Conference on Concrete and Digital Fabrication. DC 2020. RILEM Bookseries, vol 28. Springer, Cham. https://doi.org/10.1007/978-3-030-49916-7_111.

53. Pulatsu, E., Su, J.W., Lin, J., & Lin, M. (2022) Utilization of ethyl cellulose in the osmotically-driven and anisotropically-actuated 4D printing concept of edible food composites. Carbohydr. Polym. Technol. Appl. 3, 100183. https://doi.org/10.1016/j.carpta.2022.100183

54. Pulatsu, E., Su, J.W., Kenders, S.M., Lin, J., Vardhanabhuti, B., & Lin, M.(2022b). Restructuring cookie dough with 3D printing: Relationships between the mechanical properties, baking conditions, and structural changes. J. Food Eng. 319, 110911. https://doi.org/10.1016/j.jfoodeng.2021.110911

55. Pulatsu, E., & Lin, M. (2021). A review on customizing edible food materials into 3D printable inks: Approaches and strategies. Trends Food Sci. Technol. 107, 68-77. https://doi.org/10.1016/j.tifs.2020.11.023

56. Qiu, R. Wang, K., Tian, H., Liu, X., Liu, G., Hu, Z., & Zhao, L. (2022). Analysis on the printability and rheological characteristics of bigel inks: Potential in 3D food printing. Food Hydrocolloids, 129 107675, https://doi.org/10.1016/j.foodhyd.2022.107675.

57. Rahman, J.M.H., Shiblee, M.DN.I., Ahmed, K., Khosla, A., Kawakami, M., Furukawa, H. (2020). Rheological and mechanical properties of edible gel materials for 3D food printing technology. Heliyon, 6 (12), e05859, https://doi.org/10.1016/j.heliyon.2020.e05859.

58. Rando, P. & Ramaioli, M. (2021). Food 3D printing: Effect of heat transfer on print stability of chocolate,Journal of Food Engineering, 294, 110415, https://doi.org/10.1016/j.jfoodeng.2020.110415.
59. Rayna, T. & Striukova, L. (2016). From rapid prototyping to home fabrication: How 3D printing is changing business model innovation. Technological Forecasting and Social Change, 102 214-224, https://doi.org/10.1016/j.techfore.2015.07.023.

60. Shibukawa, S., Sugiyama, K., & Yano, T. (1989). Effect of heat transfer by radiation and convection on browning of cookies at baking. Journal of Food Science, 54, 621-624

61. Siacor, F.D.C., Chen, Q., Zhao, J.Y., Valino, A.D., Taboada, E.B., Caldona, E.B., & Advinculan, R.C. (2021). On the additive manufacturing (3D printing) of viscoelastic materials and flow behavior: From composites to food manufacturing. Additive Manufacturing, 45, 102043, https://doi.org/10.1016/j.addma.2021.102043.

62. Sun, J., Xiao, J., Li, Z., & Feng, X. (2021). Experimental study on the thermal performance of 3D printed concrete prototype building. Energy Building, 241, 110965.

63. Teng, X., Zhang, M., & Mujumdar, A.S. (2021). 4D printing: recent advantages and proposals in the food sector. Trends Food Science and Technology, 110, 349-363, 10.1016/j.tifs.2021.01.076

64. Tomašević, I, Putnik, P, Valjak, F., Pavlić, B., Šojić, B., Markovinović, A.B., Kovačević, D.B. (2021). 3D printing as novel tool for fruit-based functional food production. Current Opinion in Food Science, 41, 138-145, https://doi.org/10.1016/j.cofs.2021.03.015.

65. Vasilescu, S.A., Bazaz, S.R., Jin, D., Shimoni, O., & Warkiani, M.E. (2020). 3D printing enables the rapid prototyping of modular microfluidic devices for particle conjugation. Applied Materials Today, 20, 100726, https://doi.org/10.1016/j.apmt.2020.100726.

66. Venezia, P., Ronsivalle, V., Rustico, L., Barbato, E., Leonardi, R., & Lo Giudice, A. (2022). Accuracy of orthodontic models prototyped for clear aligners therapy: A 3D imaging analysis comparing different market segments 3D printing protocols. Journal of Dentistry, 124, 104212, https://doi.org/10.1016/j.jdent.2022.104212.

67. WHO (2019). Sustainable Healthy diets. Guiding Principles. https://www.who.int/publications/i/item/9789241516648.

68. Wong, G.H.C., Pant, A., Zhang, Y., Kai Chua, C., Hashimoto, M., Huei Leo, C., Tan, U.W. (2022). 3D food printing sustainability through food waste upcycling. Materials Today: Proceedings, In press, https://doi.org/10.1016/j.matpr.2022.08.565

69. Zhang, C., Wang, C.S., Therriault, D., & Heuzey, M.C. (2022). Development of aqueous protein/polysaccharide mixture-based inks for 3D printing towards food applications. Food Hydrocolloids, 131, 107742, https://doi.org/10.1016/j.foodhyd.2022.107742.

70. Zhang, J., Li, Y., Cai, Y., Ahmad, I. Zhang, A., Ding, Y. Qiu, Y., Zhang, G., Tang, G.Z., Lyu, F. (2022). Hot extrusion 3D printing technologies based on starchy food: A review. Carbohydrate Polymers, (294), 119763. https://doi.org/10.1016/j.carbpol.2022.119763.

71. Zhang, L., Zaky, A.A., Zhou, C., Chen, Y., Su, W., Wang, H., El-Aty, A.M.A., & Tan, M. (2022). High internal phase Pickering emulsion stabilized by sea bass protein microgel particles: Food 3D printing application. Food Hydrocolloids, 131, 107744, https://doi.org/10.1016/j.foodhyd.2022.107744.

72. Zonder, L., & Sella, N. (2013). Precision prototyping: the role of 3D printed molds in the injection molding industry White Paper 9-13, Stratasys, Eden Prairie, MN 55344, USA (2013)

Chapter 5 Unconstrained freedom for designing and building 3D printed food.

Oral, M.O., Derossi, A., Gleadall[ᵡ], A., Jekle[Φ], M., Caporizzi, R., Severini, C.

Department of Agriculture, Food, Natural Resources and Engineering, University of Foggia, Italy.

[ᵡ]Wolfson School of Mechanical and Manufacturing Engineering, Loughborough University, Loughborough, Leicestershire, LE11 3TU, UK

[Φ]Department of Plant-Based Foods, Institute of Food Science and Biotechnology, University of Hohenheim, Stuttgart, Germany.

Abstract

We present the results on the use of the innovative FullControl GCode designer to improve the quality and efficiency of the 3D food printing process. A dough was used to create three-dimensional structures with increased morphological complexities. The novel method was compared with the conventional approach consisting of the use of CAD and slicing softwares. Being needed to combine multiple geometric structural elements the conventional approach required 6-8 h to create the digital models instead of 30-90 min for the FullControl Gcode designer. In addition, the conventional approach generates two printing movements for each segment of the structures reducing the printing quality and the efficiency of the process. The novel method reduced the printing time by 18-36% and reduced the number of non-printing movements limiting the structural defects and the amount of food materials deposited for each segment with additional advantages on the sustainability of 3D food printing process.

Keywords: 3D printed food; printing path; CAD models; slicing, GCodes; unconstrained structures; printing efficiency.

Corresponding author: antonio.derossi@unifg.it

1. Introduction

In recent years the academia and food industries have shown an extraordinary interest in the adoption of Additive Manufacturing. For the years 2023 and 2022, a total of 19 and 330 scientific documents have been retrieved by using the keywords [3D Printing] AND [Food] on the database SCOPUS on date 1 Dec 2022. 3D Food printing (3DFP) is a disruptive technology which, for the first time, enables the transition of any digital design to a physical and edible object through a layer-by-layer deposition process of food materials; also, these food materials require specific rheological properties, globally referred as 'the printability of food' (Severini et al, 2018; Le Bail et al. 2020) to be successfully printed. A growing body of 3DFP experiments demonstrate the potential applications of 3D printing in the food sector by deeply studying the printability of food formula (Yu et al., 2022; Maldonado-Rosas et al., 2022; Cheng et al., 2022), the creation of personalized food (Pattarapon et al., 2022; Qiu et al., 2023), the new application of 4D and 5D food printing (Nida et al., 2022), modulation of calories intake (Kim et al., 2022), reuse of nutrients from food waste and by-products (Wong et al., 2022, Donn et al., 2022) as well as the design of sensory perceptions by controlling the infill level and infill path of material deposition (Fahmy et al., 2021). Other interesting readings may be found in Pant et al., (2021), Lui et al. (2018), Pulatsu, (2020), Feng et al., (2020), Severini et al. (2018), Derossi et al., (2020a; 2020b), Park et al. (2020) while industrial applications have been developed by BluRhapsody (https://blurhapsody.com/), Nourishment (https://get-nourished.com/blogs/nourished/3d-printing), etc. However, the current technological development level is limited to the following aspects: 1. Improvement of the printability of food formula; 2. Optimization of printing variables; 3. 3D printing of different kinds of food materials (fish, vegetables, cereals, etc.). However, it is growing interest in creating ambitious and complex 3D printed food structures delivering desired/personalized sensory perception and functionalities (Juravle et al., 2022) as well as the recent discovery in the new field of gastrophysics (Spence, 2017) which demonstrated the potential benefits of designing specific food texture, visual aspects, sound, and also inhomogeneous morphology (Juravle, 2022) on the improvement of the diet habits and in reducing the intakes of salt and sugar (Reinoso Charvalho et al., 2017). Given the linkage between these physico-chemical-sensory attributes, the question regarding the possibility of unconstrained freedom of design food structures is becoming a technological challenge with disruptive potential. In this context, there is a need for novel approaches to better replicate the shape and internal structure of digital food projects. The 3D printing is a multi-phase methodology consisting of some digital steps involving computer-aided design, from CAD to STL file conversion, a slicing step responsible for designing the paths for manufacturing, i.e. the path planning of 3DP, and finally the GCode generation (Jian et al., 2020). Such digital steps are widely considered essentials for the quality of the 3D printed objects (Duong et al., 2018; Gleadall, 2021;Manford et al., 2022) being responsible for the balance between printing movements and materials deposition, and several papers have been focused, in the non-food sector, on different algorithms used to define and optimize the printing path, (Volpato et al., 2013; Ma et al., 2022). Unfortunately, the food sector has completely undervalued these aspects and the majority of the experiments use conventional approaches and softwares showing several limitations and many errors (Wang et al., 2018;

Armstorng et al., 2022; Masbernat et al., 2021; Liu et al., 2018; Chen et al., 2019; Lee et al., 2019); in addition, sometimes information regarding the digital design and path planning are only limitedly reported (Lille et al., 2018; Maldonato-Rosas et al., 2022) or completely neglected (Chen et al., 2021). In this context, suitable new methodologies should be studied and established to fully exploit the great potential of additive manufacturing (Loh et al., 2018). Gleadall (2021) developed a new generic approach for unconstrained design printing paths and associated printing parameters. The approach is a FullControl GCode designer which allows to directly generate the GCode of the print-path for each segment including all printing parameters such as direction, print speed, extrusion rate, acceleration, jerk, nozzle temperature, etc. The approach extends the possibilities to design complex structures by using novel mathematically defined lattice structures, and previously inconceivable 3D geometries and functionalities for traditional slicing software to achieve (Gleadall, 2021). Considering the above-mentioned research on gastrophysics and the multisensory perception (Spence, 2020) of taste, visual aspect, texture, sounds, etc., this novel design method could represent a disruptive turning point for the creation of innovative food with features never thought before, with several social and economic benefits such as food waste reduction, sustainable diet, reduced energy intakes, etc. The focus of the present study is to investigate the potential of the novel FullControl GCode method in the food sector, in terms of improving printing fidelity (e.g. morphological features, etc.) and the efficiency of the printing process (i.e. printing time, the total number of non-printing movements, etc.) compared to the conventional approach of CAD and slicing softwares.

2. Material and methods

2.1. Preparation of the edible-ink

Wheat flour of type '00' was used for the edible-ink preparations. The flour was mixed with water in a ratio of 85 g and 100 g of water and then mixed for 7 min in a planetary kneader (model cooking chef, Kenwood Ltd. UK) at a speed level of 2. Then the dough was left to rest for 15 min at room temperature.

2.2. Digital models, GCodes development and printing experiments

Two different approaches were used to create GCodes for printing: the conventional approach of CAD and slicer software, and the newly developed approach of FullControl GCode Designer. The conventional approach consisted of two. First, a 3D digital model was designed by using popular free CAD software, Tinkercad, Autodesk Inc 2022 (https://www.tinkercad.com/) by which a STL file was created. We choose to use such software considering it enables complete beginners to create 3D digital models on the base of Constructive Solid Geometry (CSG) which enable to combine in the 3D space, single basic solid geometries to create the desired 3D structures. More specifically, during the creation process, one can add 'solid objects' and 'transparent objects' where the latter are used to remove undesired parts of the construct somewhat similar to digital subtracting manufacturing.
The STL file was sliced by using the slicing software CURA ver 3.6.0 (https://ultimaker.com/it/software/ultimaker-cura). The following printing conditions were used for all the experiments: nozzle diameter, D=0.84 mm; printing speed: 9.8 mm/s; layer

height, LH= 0.67 mm; non-printing/travel speed, TS = 10 mm/s. Also, the firmware Marlin (https://marlinfw.org/) was used to drive the 3D printer by managing all the activities and coordinating stepper motors, display, sensors, etc.

In the innovative approach, i.e. FullControl GCode designer (Gleadall, 2021), the printing path was directly defined along all printing parameters such as printing speed, layer height, extrusion rate, jerk/acceleration, etc. All printing parameters can be precisely defined, i.e. customized for each segment of the printing-path, allowing significantly improve the optimization of the printing process. The designer defines a sequence of 3D lines specifying all printing variables. mathematical equations can be used to design complex printing paths with high efficiency and non-geometric Gcode commands can be designed to control other aspects of the printing process.

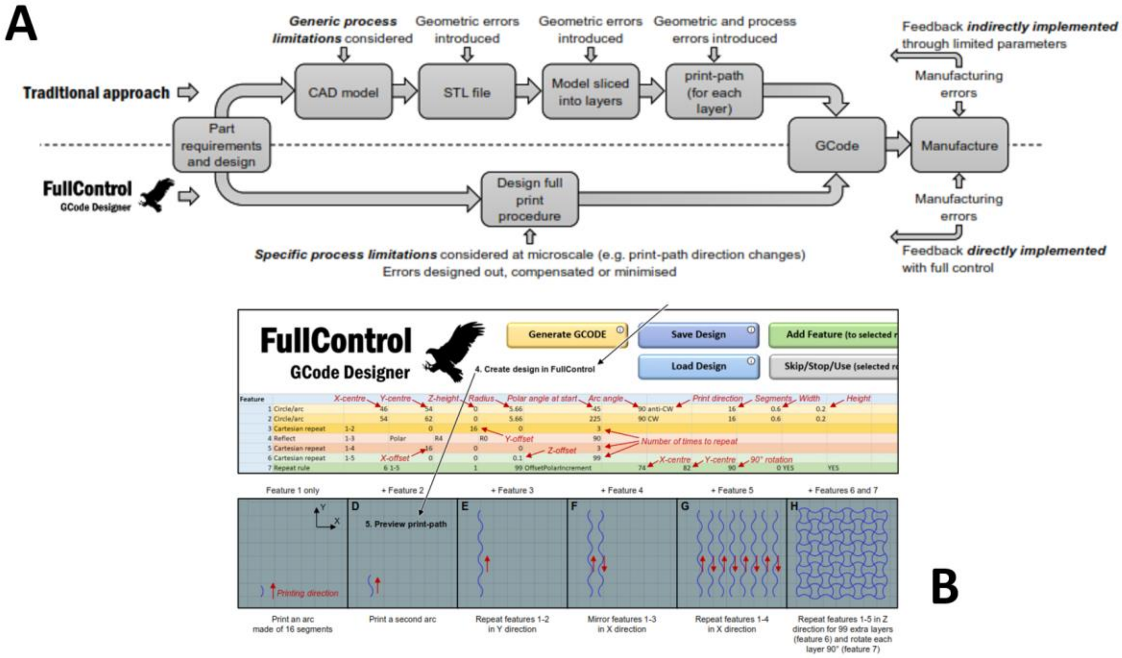

Figure 1 shows the schematic representation of the FullControl GCode Designer.

More specifically Figure 1b shows an example of the creation of the printing-path by using specific features such as 'circle/arc' and the corresponding values defining the dimension of such circles. Interestingly the feature 'Cartesian repeat' enables to replicate aforementioned features in different directions or positions significantly reducing the time for designing complex 3D digital models.

In the field of food manufacturing, this approach widely extends the opportunity of creating foods with innovative, never thought, and personalized food products due to the possibility to precisely defining the printing paths in the internal part of the object, instead of using few and constrained printing path, i.e. the 'infill paths', available on commercial slicing software. The software Repetier-Host (https://www.repetier.com/) was used to visualize the 3D digital models based on the obtained Gcodes.

To analyze the potential differences when using both approaches, we compared 3D structures designed by increasing the level of structural complexities such as: simple (S), medium (M) and complex (C) 3D structures. Samples S, M and C were designed with the following dimensions (figures 2 and 4): structure S, 40.00 mm x 72.50 mm; structure M, 40.00 mm x 65.00 mm; structure C, 65.00 mm x 65.00 mm.

All printing experiments were performed by using a FOODBOT 3D printer (A FOODBOT-MF 3D food printer (Hangzhou Shiyin Technology Co., Ltd., Zhejiang, China). Finally, the specific properties of the computer used for digital design, the slicing and the GCode creation are reported as follows: digital model design and slicing programs are, RAM: 32GB, CPU: Intel(R) Core (TM) i7-10750H CPU @ 2.60GHz (12 CPUs), 2.6GHz, GPU: NVIDIA GeForce RTX 2070 Super 8GB.

2.3. 3D printing quality and printing efficiency

A preliminary visual analysis of the printing quality was performed highlighting the existence of undesired lines of the food ink on the designed structures as a result of oozing problems during non-printing movements. The weight of the printed samples was also compared for each structure. Furthermore, the efficiency of the 3D printing process was evaluated by comparing the following main parameters: 1. working time (h) necessary to create the 3D digital models; 2. printing time per layer, PT_L; 3. the total Gcode lines per layer, GCode_L; 4. the non-printing (G0) lines of the Gcode per layer, G0_L; 5. the printing movements per layer, PM_L; 6. the travel movements per layer, TM_L; 7. the size (kb) of the Gcodes. More specifically, for G0_L, both the total number of G0 lines per layer and the fraction of G0 lines per layer, were analyzed. The latter was computed by the following equation:

$$G0\_L(\%) = \left[\sum G0\ lines\ per\ layer \Big/ \sum(G0 + G1\ lines\ per\ layer)\right] * 100 \quad \textbf{(Eq.1)}$$

Similarly, for PM_L both the global length of printing movements and the fraction of length were analyzed. The latter, was computed as follow:

$$PM\_L(\%) = \left[\sum printing\ movement\ per\ layer \Big/ \sum(printing + nonprinting\ movemmets\ per\ layer)\right] * 100 \quad \textbf{(Eq.2)}$$

Finally, the same approach was used to compute the parameter TM_L per layer:

$$TM\_L(\%) = \left[\sum travel\ movement\ per\ layer \Big/ \sum(printing + travel\ movemmets\ per\ layer)\right] * 100 \quad \textbf{(Eq.3)}$$

In addition, the printing fidelity was evaluated by comparing the 3D digital model and the 3D printed structure for some morphological features as reported in **Figure 2**.

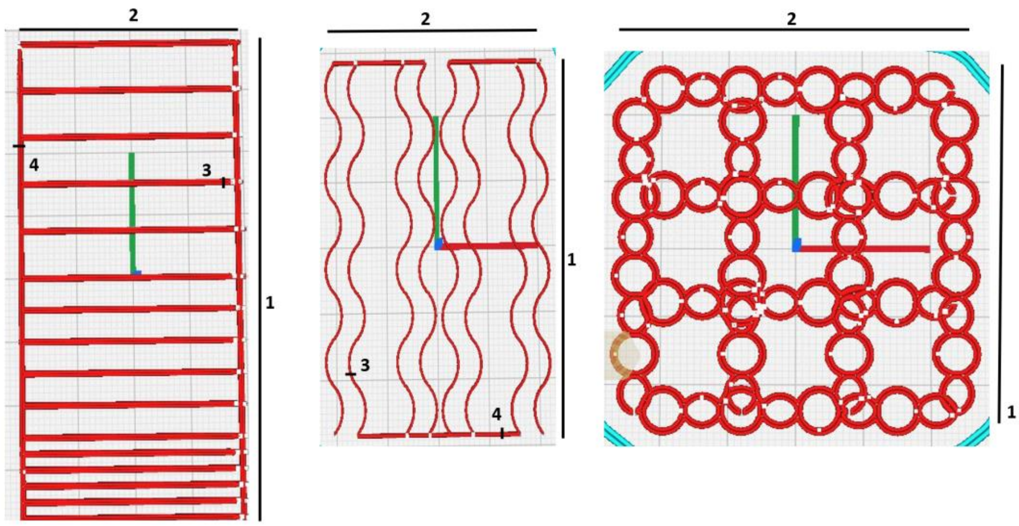

Figure 2 – Main morphological features measured on the 3D printed samples.

ImageJ software (https://imagej.nih.gov/ij/download.html) was used to calibrate the photographic images of the samples and to measure the above features. At least three replicates were always performed.

3. Results and Discussion

Initially, we analyzed the printing paths generated by using the two approaches used to design the GCodes. Although starting by analyzing the simple structure, S, the conventional approach created a printing path consisting of two lines (Fig 3c) for each segment characterizing the 3D digital structure, S (Fig 3a). Also, some non-printing movements were exhibited after the slicing step, as visualized by the blue lines in Figure 3b.

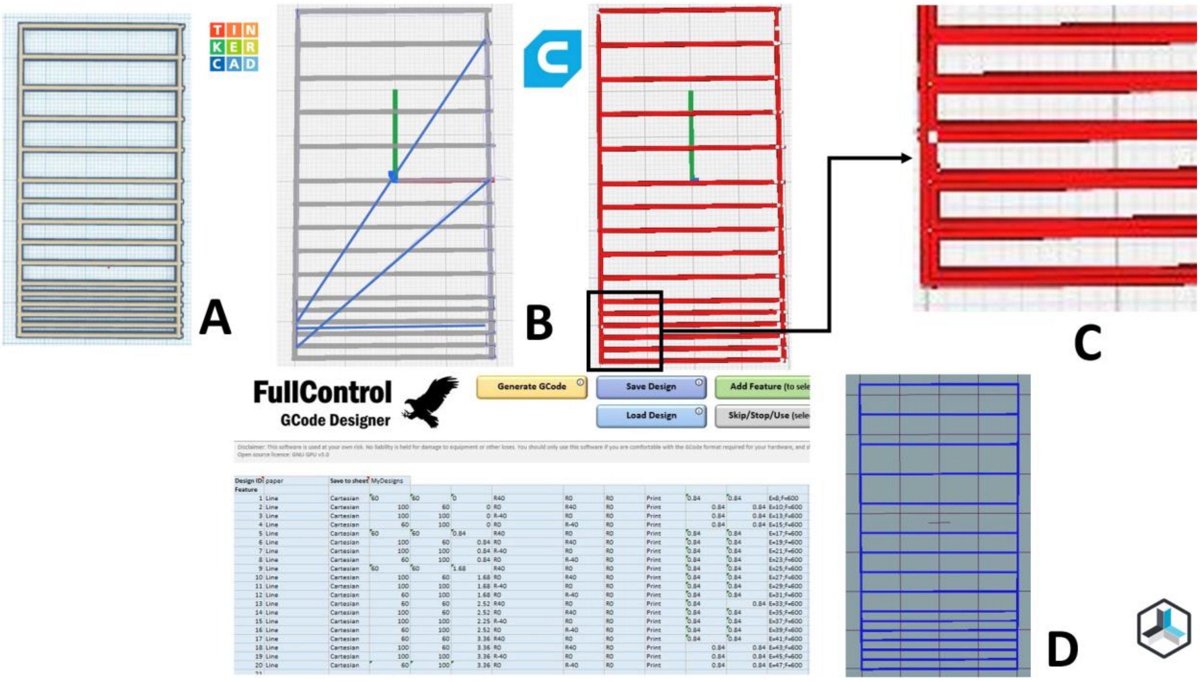

Figure 3. Schematic representation of the differences in printing paths of the simple structure (S) obtained by using the conventional approach and the FullControl Gcode designer. A) designed structure by Tinkercad; B) Sliced structure and non-printing movements; C) particular of the sliced structure showing the double printing movements per segment; D) printing path obtained by the novel approach.

These and other errors and limitations have been previously reported when utilizing the conventional approach (Tronvol et al., 2018). The observed discrepancy between the digital model and the printing path can be attributed to some procedures which are out of the control of the user: first, the conversion of the digital model into a STL file by dividing all the object surfaces into a set of triangular facets (Asianbanpour et al., 2004; Manmadhacharay et al., 2016) each of which is described by one vector and three vertices with corresponding X, Y, Z coordinates. Then, the slicing step creates a slicing plane parallel to X, Y plane that splits the STL file into several layers while such a slicing plane is moving within a specified range of Z-coordinates (Adnan et al., 2018). Finally, the coordinates of the intersecting points between the slicing plane and each facet are recorded and used to generate the printing paths (Jiang et al., 2021) based on different kinds of slicing algorithms.

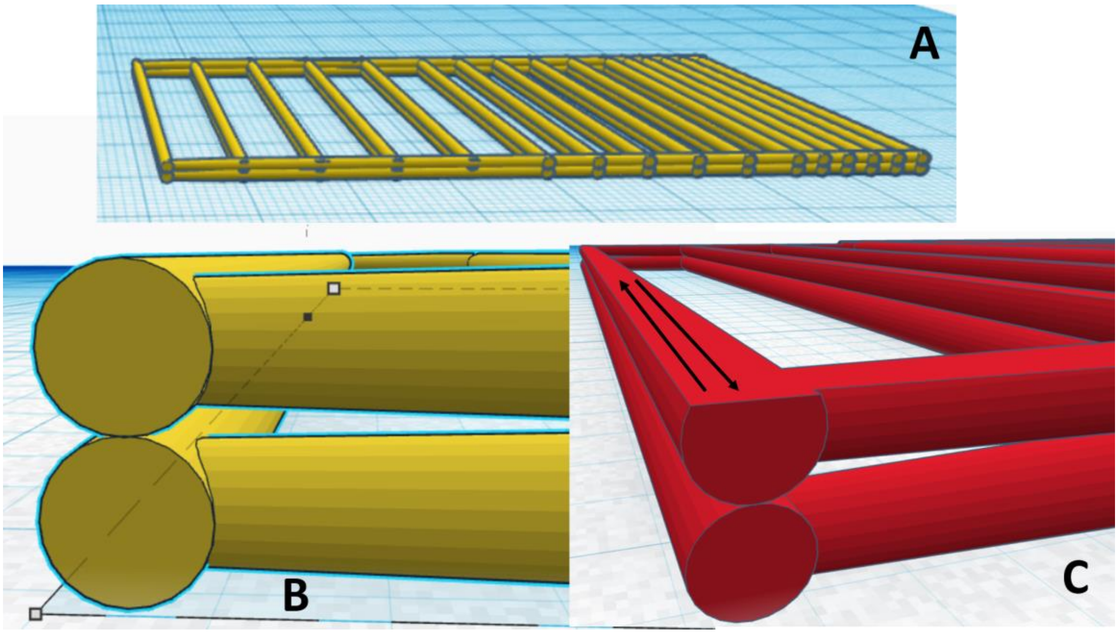

Figure 4. Creation of 3D digital model by Tinkercad. Example of combining 3D cylinders to build the digital structure, S. A) whole structure; B) Cylindrical elements combined to create the structure S; C) Schematic representation of the double printing path generated by conventional approach.

The structure S was digitally created by combining several cylinders of diameter of 0.84 mm and different lengths in the X, Y, Z plane (figure 4a and 4b). Each cylinder may be considered a solid object with many surfaces which were tessellated for generating the STL file and, later, such objects were sliced in the Z axis. In this context, the slicing step created a printing path consisting of two movements in opposite directions - i.e. forward and backward movements – for any segment of the structure S (figure 4c). The video showing such double-printing movements is reported in Supplementary materials, S1. Notice that by using parallelepipeds instead of cylinders as 3D digital elements to design the structure S, we obtained similar results with two printing movements in opposite directions for each segment (data not shown). Furthermore, the printing path depends on many variables related to the tessellation methodologies (Hallgren et al., 2016; Yang et al., 2021; Manmadhacharay et al., 2016) and slicing algorithm as well as on the use of different optimization approaches for the path-planning (Volpato et al. 2013, Yang et al., 2021). However, due to the paucity of 3D food printing experiments on these aspects, in this paper, we explored the salient differences occurring when the conventional approach and the innovative FullControl GCode designer were used. Next, by using a novel GCode designer it was possible to define the printing path of each 3D line of the structure S sequentially (figure 3D) and to define any specific information such as direction, printing speed, extrusion rate, and many other relevant properties (e.g. acceleration, jerk, etc.). This procedure facilitated the improvement of the quality and the efficiency of the process (**Table 1**) given by the following primary advantages: 1. to eliminate the phase of 3D digital design; 2. to keep only one printing movement for each segment of the structure; 3. to significantly reduce the non-printing movements. The video showing the 3D printing movements of the structure S generated by the innovative approach is shown in the Supplementary material (S1).

Such augmented printing efficiency is of extreme importance, especially in the food sector, considering the global challenges for limiting food waste and the linkages with the Sustainable Development Goals (SDGs) of the United Nations (UN, 2015). With the purpose of better demonstrating how it could be discouraging to digitally design intricate structures and, after creating the GCodes by the conventional approach, discover that the printing paths could contain significant limitations and could produce several printing errors, **Figure 5** shows the digital models and some representative slices of the structures M and C.

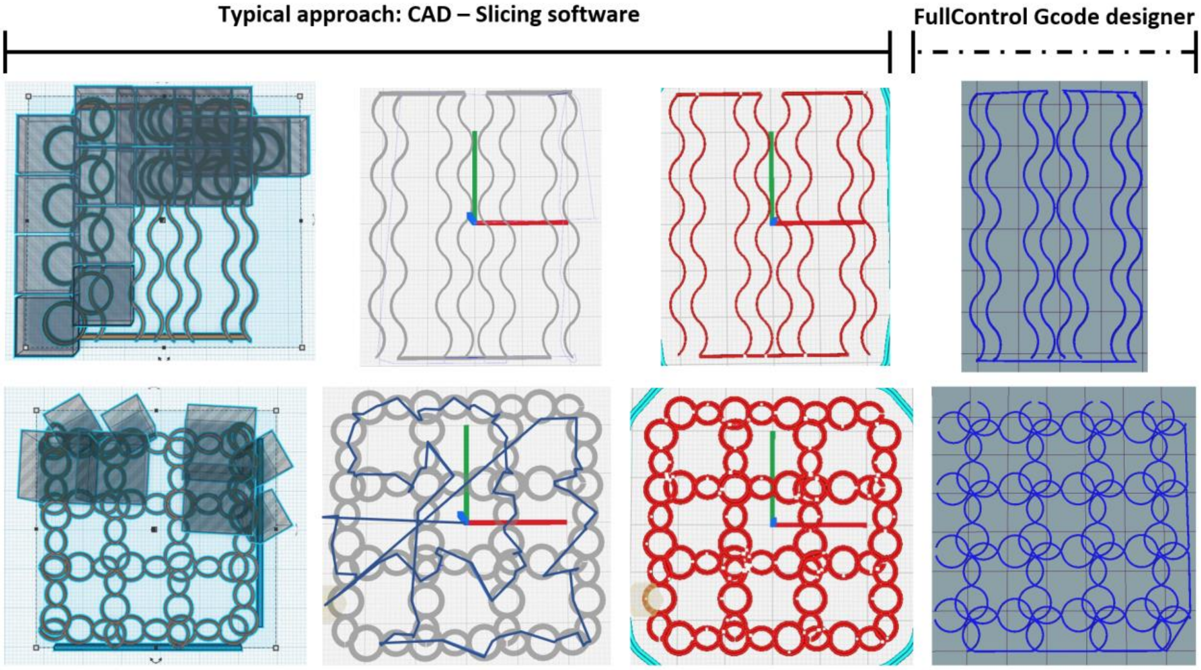

Figure 5 - 3D digital models and representation of the corresponding printing path of the structures M and C obtained by using the conventional approach and the FullControl Gcode designer.

The complexity of these structures is clearly visible. In these cases, such models were obtained by combining 3D circles with a diameter of 0.84 mm which were precisely allocated in X, Y, Z space; also, several transparent 3D circles were used as cutting tools to remove undesired parts. First, we want to answer the question of how long it took to generate the GCode. For the structures M and C respectively 6h and 8h were needed by using the conventional approach while only 30 and 90 minutes were necessary by using the Full Control designer; also, the use of the novel approach does not require programming skills (Moetazedian et al, 2020). As in the previous sample, the conventional approach produced a double printing line - i.e. forward and backward movements - for each segment of the structure. The videos of the corresponding 3D printing processes are shown in supplementary material, S1. In addition, dozens of non-printing/travel movements have been generated in the printing-path, especially for structure C as highlighted by the blue lines in **figure 5**. Furthermore, it is worth noting that for the complex structures M and C the time for the slicing steps increased exponentially due to the time needed to calculate all the intersections for one slice and then sort them into a continuous contours method. King et al. (2021) reported that this conventional procedure becomes less efficient as the complexity of the structure increases. In this condition, we decided to slice half of the height of the 3D digital model with the purpose to complete the slicing process for at least one layer and maintain the opportunity to study the potential limitations and errors generated by the printing path. On the other hand, when using the innovative approach only a single printing line was obtained for each segment of the structures M and C (Supplementary materials S1). Interestingly, the overall number of G0 commands per layer, i.e. the non-printing/travel movements, for the structure S, M and C were respectively N. 1, 8 and 8 for the FullControl GCode designer and N. 50, 15, and 887 for the conventional approach. More specifically, the fraction of travel movements per layer on the total amount of printing movements were 1.47, 0.386 and 0.386% for the FullControl GCode designer and 8.91%, 1.48% and 13.71% for the conventional approach (**Table 1**). Notice, however, that the data for the structures M and C regards only one layer for the conventional approach, by sustaining the greater efficiency of the innovative method.

Few non-printing movements increased the overall quality of the printed samples by reducing the risk of the structural defects often observed in 3D printed food due to the oozing of the food formula or the nozzle touching the other parts of the printed structure during the travel movements (Severini et al., 2018; Liu et al., 2018) and, in addition, as a consequence the touching of the previous layers considering the setting the layer height at 80% of the nozzle size to increase the adhesion of overlaying layers (Lee et al., 2019; Severini et al., 2018; Dick et al., 2019). **Figure 6** displays the photographic images of the three different 3D printed samples obtained by using the different approaches to create the GCodes. Although for structure S only minor differences can be observed, for structures M and C significant structural errors were observed when the conventional approach was utilized. More specifically, the figure shows as two unexpected diagonal lines were deposited for the structure M caused by the oozing of the food-ink during non-printing/travel movements. Differently, structure C was characterized by several and large defects such as crossing lines, increased thickness of the circles, fractures, etc. There are no doubts about the low printing fidelity when the conventional approach was used to build structure C (**Figure 6c**).

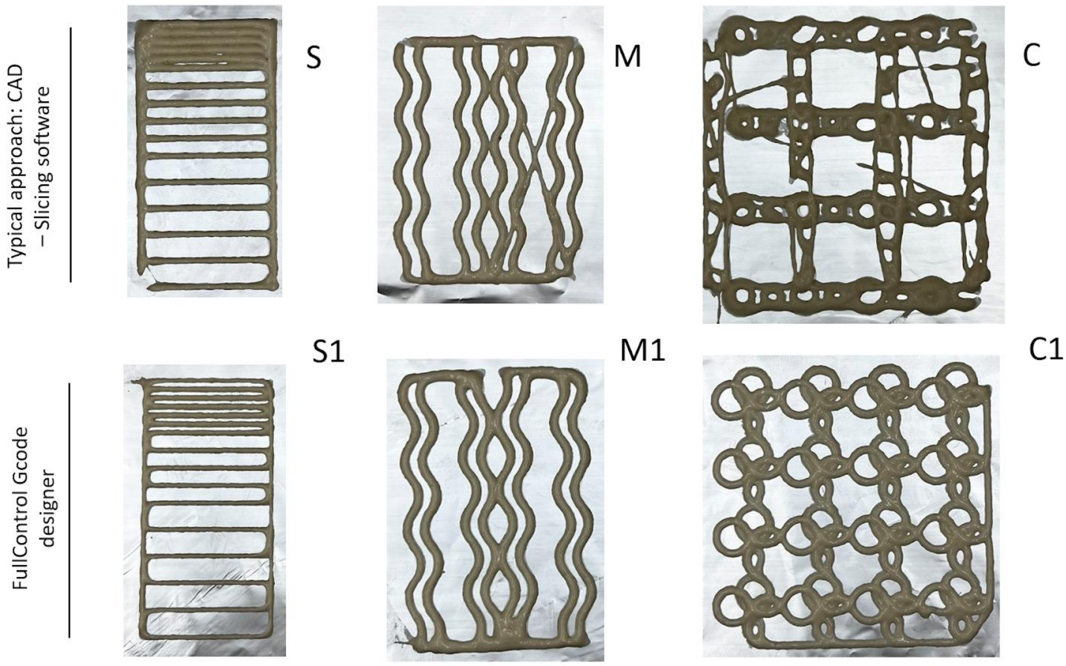

Figure 6. Photographic images of the 3D printed structures. S, M and C represent the samples obtained by using the conventional approach; S1, M1 and C1 represent the samples obtained utilizing the FullControl GCode method.

Considering the printing fidelity, the use of the FullControl designer better replicated the main dimensions of the digital models. For instance, the structures S and M showed average dimensions of 78.04±0.40, 40.76±0.36, 2.66±0.07, 59.60±0.30, 45.78±0.19, 2.84±0.19 and 2.79±0.31 mm respectively for the features n.1, 2, 3 and 4 when the conventional approach was used to generate the Gcodes. The use of FullControl Gcode designer enabled to obtain average values of 74.42±0.46, 41.61±0.28, 1.32±0.07, 1.45±1.28 mm and 67.25±0.20, 42.86±0.08, 169±0.39 and 1.747±0.134 mm were measured for the structure S and M, respectively for the features n.1, 2, 3 and 4 (data not shown). Furthermore, when considering structure C average values of 62.64±1.56 and 62.85±1.58 mm and 66.58±.45 and 65.64±0.15 mm were respectively measured for the samples obtained by utilizing the conventional and the Full Control Gcode designer, respectively. Considering the dimensions of the designed 3D digital models, The greater discrepancies obtained by using the conventional approach were the results of the above-discussed non-printing movements, nozzle touching other printed lines, and oozing effects, all of these generated dimensional changes such as stretches and compression of the printed structure.

Table 1 shows the data regarding the main properties of the 3D printed food and the primary indexes describing the process's efficiency. The measured weights were always greater for the samples obtained by using the conventional approach; for example, average values of 1.67 and 2.67 g were measured respectively for the samples created by using the FullControl Gcode designer and the conventional approach. This result supports the aforementioned benefits of the novel approach on the sustainability of the 3D printing process, proving the opportunity to deposit less food while assuring a high printing fidelity, as a result of the greater efficiency of the printing path. Despite not showing relevant differences in weight for the structures M and C between the two approaches (**Table 1**), results similar to the structure S may be envisioned considering that, for the conventional approach, the data refers to the weight of only one layer. So, by computing the weight of the whole structure consisting of two layers, a weight of 2.72 and 4.76 g would be expected for the structures M and C respectively which is much greater than the measured weight of samples obtained by using FullControl GCode designer.

Table 1 – Main quality and efficiency parameters of the 3D printed structures obtained by the conventional approach and FullControl Gcode method. Average values of and corresponding standard deviation.

Samples	Weight (g)	PT_L* (min)	GCode_L [ɤ]	G0_L [ꝛ] (N; %)	PM_L [Φ] (m; %)	TM_L [¥] (m; %)	File dimension (kb)
FullControl Gcode Designer							
Structure S	1.67±0.06	1.75±0.2	34	1; 1.47%	0.92m; 45.6%	0.08 m; 4.20%	3.00
Structure M	1.90±0.11	1.37±0.2	1032	8; 0.39%	0.71m; 49.5%	0.07 m; 0.45%	55
Structure C	2.34±0.04	1.50±0.15	1032	8; 0.38%	0,785m; 49.5%	0.08 m; 0.5%	53
Conventional approach							
Structure S	2.67±0.17	2.75±0.1	283	50; 8.9%	1.51m; 43.8 %	0.21 m; 6.15%	28.50
Structure M	1.36±0.17	2.20±0.2	1014	15; 1.5%	1.15m; 76.9%	0.30 m; 23.10 %	42.30
Structure C	2.43±0.04	3.50±0.5	6471	887; 13.71%	2.36m; 70.2%	1 m; 29.80 %	360

In the last section of the paper, we want to discuss more in detail the efficiency of the printing process. So, the printing time per layer was considered at first given its primary importance in 3D food printing (Derossi et al., 2020). Average printing times of 2.75 and 1.75 minutes were measured when the simpler structure S was printed respectively using the conventional and the FullControl Gcode method: this delivers evidence of a reduction of 36% of the printing time for the novel approach. Similarly, printing times of 1.37 and 2.20 minutes were measured for structure M, and 1.5 and 3.5 minutes for structure C, respectively when the innovative and the traditional approach were used. In the end, we want to consider the lower number of the GCode lines per layer which were of a total of N. 34, 1032 and 1032 and of 283, 1014 and 6471 for the structures S, M and C, respectively for the printing paths generated by using the conventional and the FullControl GCode approach.

4. Conclusion

We applied and tested an innovative approach for creating GCodes in the field of 3D Food Printing to explore new avenues to improve the quality of 3D printed food as well as to increase the efficiency of the process. Also, the experiments were carried out to improve our understanding of the digital phases of the 3D food printing process which, more than often, are undervalued. Our study provides evidence of the improvement of the printing fidelity of the structures when the FullControl GCode designer was utilized while highlighting the limits and the errors occurring by using the conventional approach consisting of two common steps: 1. Creation of a digital model by using CAD software; 2. Slicing the STL and generating the GCode. Three main problems were encountered by using the conventional approach, especially for the more complex structures M and C: 1. It took many hours to digitally design the structures and to create the STL file; 2. The failure of completing the slicing step in the appropriate time; 3. The slicing generated dozens of undesired travel moments with negative effects on the printing time and the worst printing quality. The use of novel approaches could be a good candidate to improve the overall printing fidelity and the efficiency of the process both in terms of printing time and fidelity. Furthermore, the proposed approach could open innovative food products, never thought before, with inhomogeneous structures capable to convey new sensory perceptions to facilitate mastication for elderly people, also in line with the idea of personalized food structure.

5. Literature

1. Adnan, F.A., Romlay, F.R.M., & Shafiq, M. (2018). Real-time slicing algorithm for stereolithography (STL) CAD model applied in additive manufacturing industry. *IOP Conf. Ser. Mater. Sci. Eng.*, 342, 102016. doi:10.1088/1757-899X/342/1/012016

2. Armstrong, C.D., Yue, L., Deng, Y., & Qi, H.J. (2022). Enabling direct ink write edible 3D printing of food puress with cellulose nanocrystals. *Journal of Food Engineering*, 330, 111086. https://doi.org/10.1016/j.jfoodeng.2022.111086

3. Asiabanpour, B., Khoshenevis, B. (2004). Machine path generation for the SIS process. *Robotics and Computer Manufacturing*, 20, 167-175. https://doi.org/10.1016/j.rcim.2003.10.005

4. Carvalho, F.R., Wang, Q., van Ee, R., Persoone, D., & Spence, C. (2017). 'Smooth operator': music modulates the perceived creaminess, sweetness, and bitterness of chocolate. *Appetite*, 108, 383-390. https://doi.org/10.1016/j.appet.2016.10.026

5. Chen, J., Mu, T., Goffin, D., Blecker, C., Richard, G., Richel, A., & Haubruge, E. (2019). Application of soy protein isolate and hydrocolloids based mixture as promising food material in 3D food printing. *Journal of Food Engineering*, 261, 76-86. https://doi.org/10.1016/j.jfoodeng.2019.03.016

6. Chen, Y. Zhang, M., & Phuhongsung, P. (2021). 3D printing of protein-based composite fruit and vegetable gel system. *LWT Food Science and Technology*, 141, 110978. https://doi.org/10.1016/j.lwt.2021.110978

7. Cheng, Y., Fu, Y., Yap, LP.L., Losic, D., Wang, H., & Zhang, Y. (2022). Rheology of edible food inks from 2D/3D/4D printing, and its role in future 5D/6D printing. *Food Hydrocolloids*, 132, 107855. https://doi.org/10.1016/j.foodhyd.2022.107855

8. Derossi, A., Caporizzi, R., Oral, M.O., & Severini, C. (2020). Analyzing the effects of 3D printing process per se on the microstructure and mechanical properties of cereal

food products. *Innovative Food Science and Emerging Technology*, 66, 102531. https://doi.org/10.1016/j.ifset.2020.102531

9. Derossi, A., Caporizzi, R., Paolillo, M., & Severini, C. (2020b). Programmable texture properties of cereal-based snack mediated by 3D printing technology. *Journal of Food Engineering*, 289, 110160. https://doi.org/10.1016/j.jfoodeng.2020.110160

10. Derossi, A., Paolillo, M., Caporizzi, R., & Severini, C. (2020). Extending the 3D food printing test at high speed. Material deposition and effect of non-printing movements on the final quality of printed structures. *Journal of Food Engineering*, 275, 109865. https://doi.org/10.1016/j.jfoodeng.2019.109865

11. Derossi, A., Paolillo, M., Caporizzi, R., & Severini, C. (2020a). Extending the 3D food printing test at high speed. Material deposition and effect of non-printing movements on the final quality of printed structures. *Journal of Food engineering*, 275, 109865. https://doi.org/10.1016/j.jfoodeng.2019.109865

12. Dick, A., Bhandari, B., Prakash, S. (2019). 3D printing of meat. *Meat Science*, 153, 35-44. https://doi.org/10.1016/j.meatsci.2019.03.005

13. Donn, P., Prieto, M.A., Mejuto, J.C., Cao, H., & Simal-Gandara, J. (2022). Functional foods based on the recovery of bioactive ingredients from food and algae by-products by emerging extraction technologies and 3D printing. *Food Bioscience*, 49, 101853. https://doi.org/10.1016/j.fbio.2022.101853

14. Duong, T.H., Jaksic, N.I., DePalma, J.L., Ansaf, B., Daniel, D.M., Armijo, J., & Galavi, M. (2018). G-code visualization and editing program for inexpensive metal 3D Printing. *Procedia manufacturing*, 17, 22-28. https://doi.org/10.1016/j.promfg.2018.10.007

15. Fahmy, A.R., Amann, L.S., Dunkel, A., Frank, O., Dawid, C., Hofmann, T., Becker, T., & Jekle, M. (2021). Sensory design in food 3D printing – Structuring, texture

modulation, taste localization and thermal stabilization. *Innovative Food Science and Emerging Technology*, 72, 102743. https://doi.org/10.1016/j.ifset.2021.102743

16. Feng, C., Zhang, M., Bhandai, B., & Ye, Y. (2020). Use of potato processing by-product: effects on the 3D printing characteristics of the yam and the texture of air-fried yam snacks. *LWT – Food Science and Technology*, 125, 109265. https://doi.org/10.1016/j.lwt.2020.109265

17. Gleadall, A., (2021). FullControl GCode Designer: open-source software for unconstrained design in additive manufacturing. *Additive Manufacturing*, 46, 102109. https://doi.org/10.1016/j.addma.2021.102109

18. Halllgren, S., Pejryd, L., & Ekengren J. (2016). 3D data export for additive manufacturing – improving geometric accuracy. *Procedia CIRP design conference*, 50, 518-523. https://doi.org/10.1016/j.procir.2016.05.046

19. Jiang, J., & Ma, Y. (2020). Path planning strategies to optimize accuracy, quality, build time and material use in additive manufacturing: A review. *Micromachines*, 11(7), 633. https://doi.org/10.3390/mi11070633

20. Jiang, J., & Ma, Y. (2020). Path planning strategies to optimize accuracy, quality, build time and material use in additive manufacturing: A Review. *Micromachines*, 11, 633. https://doi.org/10.3390/mi11070633

21. Juravle, G., Olari, E.L., & Spence, C. (2022). A taste for beauty: On the expected taste, hardness, texture, and temperature of geometric shapes. *I-Perception*. https://doi.org/10.1177%2F20416695221120948

22. Kim, S.M., Wen, Y., Kim, H.W., & Park, H.J. (2022). Textural and sensory qualities of low-calories surimi with carrageenan inserted as a protein substitute using coaxial extrusion 3D food printing. *Journal of Food engineering*, 333, 111141. https://doi.org/10.1016/j.jfoodeng.2022.111141

23. King, B., Rennie, A., & Bennet, G. (2021). An efficient triangle mesh slicing algorithm for all topologies in additive manufacturing. *The International Journal of Advanced Manufacturing Technology*, 112, 1023-1033. https://doi.org/10.1007/s00170-020-06396-2

24. Le-Bail, A., Chieregato Maniglia, B., & Le-Bail, P. (2020). Recent advances and future perspective in additive manufacturing of foods based on 3D printing. *Current Opinion in Food Science* 35, 54-64. https://doi.org/10.1016/j.cofs.2020.01.009

25. Lee, J.H., Won, D.J., Kim, H.W., & Park, H.J. (2019). Effect of particle size on 3D printing performance of the food-ink system with cellular food materials. *Journal of Food Engineering*, 256, 1-8. https://doi.org/10.1016/j.jfoodeng.2019.03.014

26. Lille, M., Nurmela, A., Nordlund, E., Metsa-Kortelainen, S., & Sozer, N. (2018). Applicability of protein and fiber-rich food materials in extrusion-based 3D printing. *Journal of Food Engineering*, 220, 20-27. https://doi.org/10.1016/j.jfoodeng.2017.04.034

27. Liu, Z., Bhandari, B., Prakash, S., & Zhang, M. (2018). Creation of internal structure of mashed potato construct by 3D printing and its textural properties. *Food Research International*, 111, 534-543. https://doi.org/10.1016/j.foodres.2018.05.075

28. Liu, Z., Bhandari, B., Prakash, S., & Zhang, M. (2018a). Creation of internal structure of mashed potato construct by 3D printing and its textural properties. *Food Research International*, 111, 534–543. https://doi.org/10.1016/j.foodres.2018.05.075

29. Loh, G.H., Pein, E., Harrison, D., & Monzon, M.D. (2018). An overview of functionally graded additive manufacturing. *Additive Manufacturing*, 23, 34–44. https://doi.org/10.1016/j.addma.2018.06.023

30. Ma, Z., Wan, W., Song, L., Liu, C., Liu, H., & Wu, Y. (2022). An approach of path optimization algorithm for 3D concrete printing based on graph theory, *Applied Science*, 12, 113115. https://doi.org/10.3390/app122211315

31. Maldonado-Rosas, R, Tejada-Ortigoza, V., Cuan-Urquizo, E., Mandoza-Cachù, D., Morelas-de la Pena, M., Alvarado-Orozco, J.M., & Campanella, O.H. (2022). Evaluation of rheology and printability of 3D printing nutritious food with complex formulations. *Additive Manufacturing*, 58, 103030. https://doi.org/10.1016/j.addma.2022.103030

32. Maldonado-Rosas, R., Tejada-Ortigoza, V., Cuan-Urquizo, E., Mendoza-Cachù, Morales-de la Pena, M., Alvarado-Orozco, J.M., Campanella, O.H. *Additive Manufacturing*, 58, 103030.

33. Manford, D., Budinoff, H.D., Bushra, J. (2022). Accuracy of build time and mass estimates from slicer software for fused filament fabrication. *Manufacturing letter*, 33, 752-757. https://doi.org/10.1016/j.mfglet.2022.07.092

34. Manmadchary, A., Ravi Kumar, Y., & Krishananand, L. (2016). Improve the accuracy, surface smoothing and material adaptation in STL file for RP medical models. *Journal of Manufacturing Processes*, 21, 46-55. http://dx.doi.org/10.1016%2Fj.jmapro.2015.11.006

35. Masbernat, L., Berland, S., Leverrier, C., Moulin, G., Michon, C., & Almeida, G. (2021). Structuring wheat dough using a thermomechanical process, from liquid food to 3D-printable food material. *Journal of Food Engineering*, 310, 1106969. https://doi.org/10.1016/j.jfoodeng.2021.110696

36. Moetazedian, A., Gleadall, A., Han, X., & Silberschmidt, V. (2020). Effect of environment on mechanical properties of 3D printed polylactide for biomedical applications. Journal of the mechanical behavior of biomedical materials. 102, 103510. https://doi.org/10.1016/j.jmbbm.2019.103510

37. Nida, S., Moses, J.A., & Anandharamakrishnan, C. (2022). Emerging applications of 5D and 6D printing in the food industry. *Journal of Agriculture and Food Research*, 10, 100392. https://doi.org/10.1016/j.jafr.2022.100392

38. ONU (2015). Transforming our world: the 2030 Agenda for Sustainable development. https://documents-dds ny.un.org/doc/UNDOC/GEN/N15/291/89/PDF/N1529189.pdf?OpenElement

39. Pant, A., Lee, A.Y., Karyappa, R., Lee, C.P., An, J., Hashimoto, M., Tan, U.X., Wong, G., Chua, C.K., & Zhang, Y. (2021). 3D food printing of fresh vegetables using food hydrocolloids for dysphagic patients. Food Hydrocolloids, 114, 106546, https://doi.org/10.1016/j.foodhyd.2020.106546

40. Park, S. M., Kim, H. W., & Park, H. J. (2020). Callus-based 3D printing for food exemplified with carrot tissues and its potential for innovative food production. *Journal of Food Engineering*, 271, 109781. https://doi.org/10.1016/j. jfoodeng.2019.109781

41. Pattarapon, P., Zhang, M., & Mujumdar, A. (2022). Application potential of 3D food printing to improve the oral intake for immunocompromised patients: A review. *Food Research International*, 160, 111616. https://doi.org/10.1016/j.foodres.2022.111616

42. Pulatsu, E., Su, J.W., Lin, J., & Lin, M. (2020). Factors affecting 3D printing and post-processing capacity of cookie dough. *Innovative Food Science & Emerging Technologies*, 61, 102316. https://doi.org/10.1016/j.ifset.2020.102316

43. Qiu, L., Zhang, M., Bhandari, B., Chitrakar, B., & Chang, L. (2023). Investigation of 3D printing of apple and edible rose blends as a dysphagia food. Food Hydrocolloids, 135, 108184. https://doi.org/10.1016/j.foodhyd.2022.108184

44. Severini C, Azzollini D, Albenzio M, & Derossi, A. (2018). On printability, quality and nutritional properties of 3D printed cereal-based snacks enriched with edible insects. *Food Research International*, 106, 666-676. https://doi.org/10.1016/j.foodres.2018.01.034

45. Severini, C., Azzollini, D., Albenzio, M., & Derossi, A. (2018). On printability, quality and nutritional properties of 3D printed cereal based snacks enriched with edible insects. *Food Research International*, 106, 666–676. https://doi.org/10.1016/j.foodres.2018.01.034

46. Severini, C., Derossi, A., Ricci, I., Caporizzi, R., & Fiore, A. (2018). Printing a blend of fruit and vegetables. New advances on critical variables and shelf life on 3D edible objects. *Journal of Food Engineering*, 220, 89-100. https://doi.org/10.1016/j.jfoodeng.2017.08.025

47. Severini, C., Derossi, A., Ricci, I., Caporizzi, R., & Fiore, A. (2018). Printing a blend of fruit and vegetables. New advances on critical variables and shelf life of 3D edible object. *Journal of Food Engineering*, 220, 89-100. https://doi.org/10.1016/j.jfoodeng.2017.08.025

48. Spence, C. (2017). Gastrophysics: The New Science of Eating. Viking Penguin, London, UK (2017)

49. Tronvoll, S.A., Elverum, C.W., & Welo, T. (2018). Dimensional accuracy of threads manufactured by fused deposition modeling, *Procedia Manuf*. 26 (2018) 763–773. https://doi.org/10.1016/j.promfg.2018.07.088

50. Volpato, N., Franzoni, A., Luvizon, D.C., & Schramm, J.M. (2013). Identifying the directions of a set of 2D contours for additive manufacturing process planning, *International Journal Advaced Manufacturing Technologies*, 68, 33-43. https://doi.org/10.1007/s00170-012-4706-y

51. Wang, L., Zhang, M., Bhandari, B., & Yang, C. (2018). Investigation of fish surimi gel as promising food material for 3D printing. *Journal of Food Engineering*, 220, 101-108. https://doi.org/10.1016/j.jfoodeng.2017.02.029

52. Wong, G.H.C., Pant, A., Zhang, Y., Chua, C.K., Hashimoto, M., Leo, C.H., & Tan, U.X. (2022). 3D food printing- sustainability through food waste upcycling. *Materialstoday*, 70, 627-630. https://doi.org/10.1016/j.matpr.2022.08.565

53. Yang, M., Lai, M., & Liu, S. (2021). 3D Printing path planning algorithm for thin walled and complex devices. *Science and Engineering of Composite Materials*, 28, 327-334. https://doi.org/10.1515/secm-2021-0032

54. Yu, J., Wag, X.y., Li, D., Wang, L., & Wang, Y. (2022). Development of soy protein isolate emulsion gels as extrusion-based 3D food printing inks: Effect of polysaccharides incorporation. *Food Hydrocolloids*, 131, 107824. https://doi.org/10.1016/j.foodhyd.2022.107824

Chapter 6 Utilising Mediterranean food waste and by-products in 3D Food Printing

Oral, M.O, Derossi, A., Caporizzi, R., Severini, C.

Department of Agriculture, Food, Natural Resources and Engineering, University of Foggia, Italy.

Abstract

This study explores the transformative potential of 3D Food Printing (3DFP) in utilizing food waste and non-edible by-products for the creation of personalized food products. The research highlights the process of drying and grinding these by-products into powder form, enabling their reintegration into the food supply chain, thus contributing to a circular economy. The study further underscores the capacity of 3DFP to manipulate the texture and flavour of food, opening new possibilities for personalized nutrition. The research also suggests that the use of food by-products in 3DFP could enhance food diversity and security by reducing reliance on a limited number of staple crops. However, the study identifies the need for further research to optimize the particle size and concentration of different food by-products, ensure the printability and sensory acceptability of the printed food, and explore the nutritional implications of using food by-products in 3DFP. The findings of this study underscore the potential of 3DFP as a tool for sustainable and personalized food production, paving the way for a future where food waste is minimized, dietary needs are met with precision, and the enjoyment of food consumption is enhanced through innovative and appealing designs.

Keywords: 3D Food Printing, Personalized Nutrition, Sustainability, Food Waste, Non-Edible By-Products

1. INTRODUCTION

The imperative to mitigate food loss and waste across all stages of the food supply chain is a pressing concern for the European Union. This urgency is underscored by the most recent and pertinent data, which reveals that billions of tons of food, equating to approximately one-third of all food produced annually for human consumption, are wasted yearly (FAO, 2011; Girotto et al., 2015). Food wastage is a pervasive issue that permeates every step of the food chain, from agricultural production (Johnson et al., 2019) to processing (Redlingshöfer et al., 2017; Garcia-Herrero et al., 2019), retail (Lebersorger et al., 2014), and household consumption (van Dooren et al., 2019; Revilla et al., 2018; Philippidis et al., 2019). For instance, food loss at the agricultural level manifests when edible products remain unharvested or are harvested but not sold or donated, leading to surplus crops. A more granular analysis reveals that approximately 45% of roots and tubers, 45% of fruits and vegetables, and 35% of fish and seafood are lost in North America (Levis et al., 2010; FAO, 2011; Johnson et al., 2019). When translated into nutritional terms, this equates to a daily loss of 40-47% of the calories and protein from seafood products (Love et al., 2015), along with significant quantities of vitamin C, iron, dietary fiber, amino acids, and fatty acids in the EU (Fusion project, www.eu-fusions.org). Beyond the direct loss of food, food waste also represents a significant resource and sustainability challenge. The processes involved in food production consume substantial resources, including land, water, energy, and fertilizers, while simultaneously engendering environmental adversities such as biodiversity and habitat loss, soil and water degradation, and greenhouse gas emissions. Therefore, addressing food waste is a matter of food security and a crucial component of sustainable resource management.

3D Food Printing, 3DFP, is the only technique capable to translate digital images into tangible food products and this opens the way for innovative and intricated shapes and dimensions maximizing food's eye appeal, helping in differentiating or identifying food products, and improving the overall enjoyment of food consumption. However, 3DFP is well beyond the creation of food with fascinating visual aspects, with many additional benefits on the food chain, nutritional/healthy and sensory properties, satiety, consumer's behavior, and sustainability (German et al., 2011; Oral et al., 2021). For instance, personalized 3D printed food considers the production of foods with desired sensorial and nutritional properties offering many solutions to contribute to the better health status of consumers. In addition, at its core 3DFP has the process of dispensing/depositing small amounts of food material per unit of time offering the opportunity of dosing each ingredient with high accuracy to modulate the content of macro- and micronutrients according to the needs of each individual or small group of consumers. Finally, though, 3DP supports the decentralization of food manufacturing and the consumer-centric system of production allowing the manufacture of products close to the final customer (Chan et al., 2018). By reducing the tight dependence on the supply chain, it would be possible to increase the overall sustainability of the food system with a significant reduction in energy consumption and gas emission generated by transportation, and the amount of food waste or food loss at industrial or home level (Derossi et al. 2021).

This study seeks to broaden the conventional application of 3D Food Printing Technology, venturing into the emerging domains of sustainable food systems and bespoke food manufacturing. Specifically, the research aims to repurpose surplus crops and by-products from Mediterranean food production, leveraging their rich nutrient and bioactive compound content to create innovative personalized food products. This endeavor will be achieved by exploiting the vast potential of 3D Food Printing, both in terms of nutritional and sensory properties.
A distinctive feature of this study lies in its approach to raw material utilization. Instead of relying on extracted phytochemicals, the research will employ whole raw materials abundant in bioactive compounds. This innovative method, combined with the transformative potential of 3D Food Printing, aims to contribute significantly to personalized nutrition and sustainable food production.

2. Material & Methods

2.1. Determination of the Raw Material

Artichokes, eggplants, and tomatoes have been purchased locally and the external leaves (for artichokes) or the skin (for eggplant and tomato) usually treated as food waste were reserved for the drying process. A commercial air-drier (Klarstein-Arizona Jerky food dehydrator) was used for drying experiment at temperatures of 30°C, 40°C, and 50°C. During the drying process, the samples were analysed every 30 min for weight and moisture content (AOAC, 2005). Water activity was measured by water activity meter (Decagon device).

The initial phase of the experiment, spanning the first 30 minutes, was characterized by a higher frequency of data collection, with three replicate samples being taken at 15-minute intervals. This approach was designed to capture the rapid changes in moisture content that typically occur in the early stages of the drying process.

Following the initial phase, the sampling frequency was reduced to every 30 minutes, with three replicates taken at each time point. This adjustment in sampling frequency was predicated on the understanding that the rate of change in moisture content tends to decrease as the drying process progresses, thus necessitating less frequent measurements. The data collected from these measurements were instrumental in establishing the drying curves for each vegetable at each temperature condition. This rigorous and systematic approach obtained a comprehensive dataset, capturing the dynamic changes in moisture content throughout the drying process. This dataset provides a robust foundation for analyzing drying kinetics in these vegetables and developing predictive models for moisture loss under different temperature conditions. This research contributes to the broader understanding of food preservation techniques, with potential implications for improving the efficiency and sustainability of food processing operations.

2.2. Description of dehydration kinetics

The drying curves were obtained by plotting the moisture ratio (MR) as a function of time as reported by (Jangam et al., 2010). The moisture ratio was computed by using Eq. 1;

$$M_R = \frac{M_t - M_e}{M_i - M_e} = \exp(-Kt) \quad \text{(Eq.1)}$$

Where M_R is the dimensionless moisture ratio, M_t is the moisture content at any time of drying (kg water/kg dry matter), M_i is the initial moisture content (kg water/kg dry matter) and M_e is the equilibrium moisture content (kg water/ kg dry matter).
The M_R values as a function of time were modelled by using the Page's model:

$$MR = \exp(-kt^n) \quad \text{(Eq.2)}$$

Where k (1/min) is the rate constant, *t* (min) and *n* are a fitting parameter (dimensionless). The fitting was performed by using the packages of STATISTICA.

Furthermore, the moisture content as a function of water activity was modelled by using the GAB equation:

$$EMC = \frac{x_0 * C * K * a_w}{(1 - k * a_w) * (1 + (c-1) * k * a_w} \quad \text{(Eq.3)}$$

X_0: Moisture content in the monolayer
C: Guggenheim constant
K: Related to the heat of adsorption in the multilayer

where EMC is the equilibrium moisture content of the material, X_0 is the monolayer moisture content, which is the amount of water that can be adsorbed in a single layer on the surface of the material, C and K are constants that depend on the nature of the material and the temperature, a_w is the water activity.

2.3. Ink-Gel Preparation and 3D Printing Process

2.3.1 Ink-gel preparation

The starch gel was formulated utilizing wheat starch sourced from Sigma-Aldrich, USA. An exact quantity of 12 grams of wheat starch was meticulously measured using an Accuris Analytical Balance. This starch was amalgamated with 88 grams of distilled water in a beaker. Throughout this process, continuous stirring was employed to guarantee the attainment of a homogeneous mixture.
Utilizing a Velp Scientifica brand ARE Heating Magnetic Stirrer, a mixture of pure water, and wheat starch was subjected to a carefully controlled thermal and mechanical treatment. The stirrer was set to a precise temperature of 90°C, and the mixture was agitated at 400 rpm for 15 minutes. This meticulous process facilitated the formation of a homogeneous starch gel. Upon

completion of the stirring process, the gel was promptly removed from the stirrer, and its temperature was reduced to 55°C. To preserve the uniformity of the gel, it was promptly transferred into 3D printer syringes while still at this temperature. Subsequently, the gel-filled syringes were allowed to equilibrate to ambient temperature.
Throughout this procedure, the temperature and duration of heating were meticulously monitored to circumvent any potential thermal degradation of the starch. The cooling of the starch gel to room temperature prior to its utilization in the 3D printing process was a critical step to ensure the preservation of its structural integrity and homogeneity.

2.3.2 Digital Model Design

A three-dimensional digital model of a square was meticulously crafted using Tinkercad (Autodesk, USA), a user-friendly, web-based 3D modelling platform. The model was designed with precise dimensions of 30mm x 30mm x 10mm. The design process entailed the creation of a fundamental square shape, followed by carefully adjusting its dimensions to meet the desired specifications.

After the design phase, the model was exported as an STL file, a standard file format in 3D printing and an acronym for Stereolithography, from Tinkercad to the CURA slicing software. Within the CURA interface, an array of printer settings was meticulously calibrated, including parameters such as print speed, set at 8 mm/s, flow rate, adjusted to 6 mm/s, and layer height, established at 0.84 mm. Furthermore, two distinct model versions were generated, each with a different infill level: one with a 10% infill and the other with a 40% infill. The adjustment of the infill level was executed within the Tinkercad settings prior to the exportation of the model for printing.

Upon the completion of these adjustments, the file was exported in the gcode format, a widely utilized numerical control programming language, to the 3D printer, thereby commencing the printing process.

2.3.3 3D Printing Experiments

The 3D printing process was carried out using the prepared starch gel as the printing material. The 3D printer (FoodBot 3D printer) was set up with the following printing conditions: a printing speed of 10 mm/s, a flow speed of 8 mm/s, and a layer height and width of 0.8 mm. These settings were chosen to ensure a high-quality print while minimizing the risk of print failure.

3. Results and Discussion

3.1. Dehydration of the Vegetable and Obtain the Deyhdration Curves

The experimental procedure was meticulously conducted across four distinct vegetable types, i.e., the not edible parts of artichokes, eggplants, lemons and tomatoes. These were subjected to varying temperature conditions, specifically at 30°C, 40°C, and 50°C, to investigate the impact of temperature on the kinetic of drying. The initial phase of the experiment, spanning the first 30 minutes, was characterized by a higher frequency of data collection, with three replicate samples being taken at 15-minute intervals. This approach was designed to capture the rapid changes in moisture content that typically occur in the early stages of the drying process.

Following the initial phase, the sampling frequency was reduced to every 30 minutes, with three replicates taken at each time point. This adjustment in sampling frequency was predicated on the understanding that the rate of change in moisture content tends to decrease as the drying process progresses, thus necessitating less frequent measurements. The data collected from these measurements were instrumental in establishing the drying curves for each vegetable at each temperature condition. This rigorous and systematic approach obtained a comprehensive dataset, capturing the dynamic changes in moisture content throughout the drying process. This dataset provides a robust foundation for analyzing drying kinetics in these vegetables and developing predictive models for moisture loss under different temperature conditions. This research contributes to the broader understanding of food preservation techniques, with potential implications for improving the efficiency and sustainability of food processing operations.

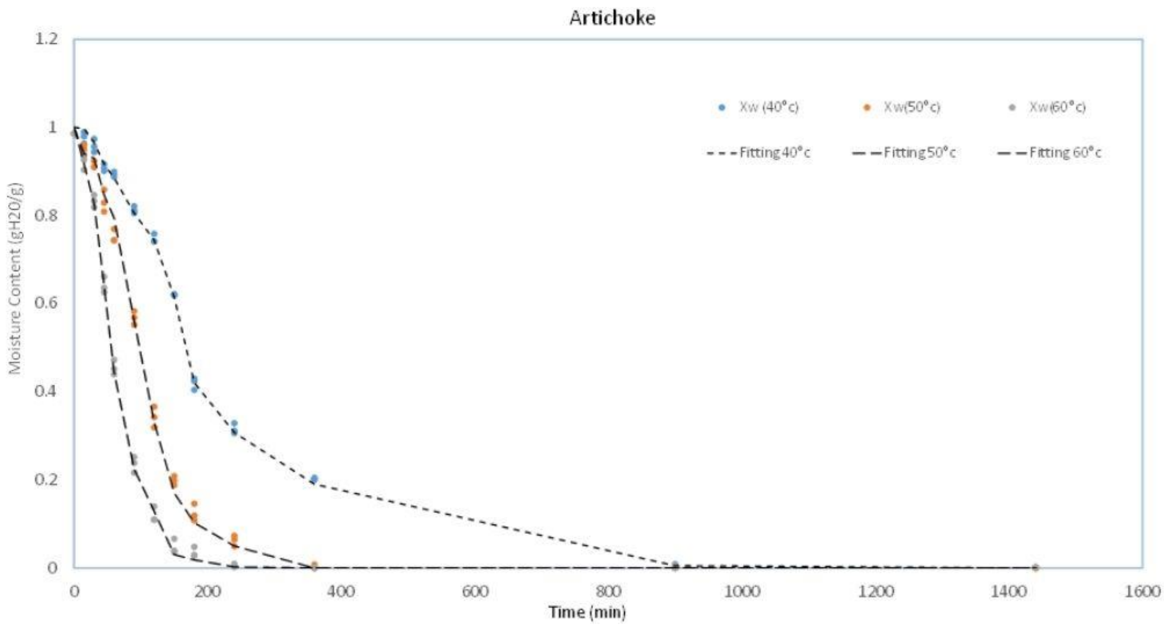

Figure 1: Dehydration curve of artichoke by-products

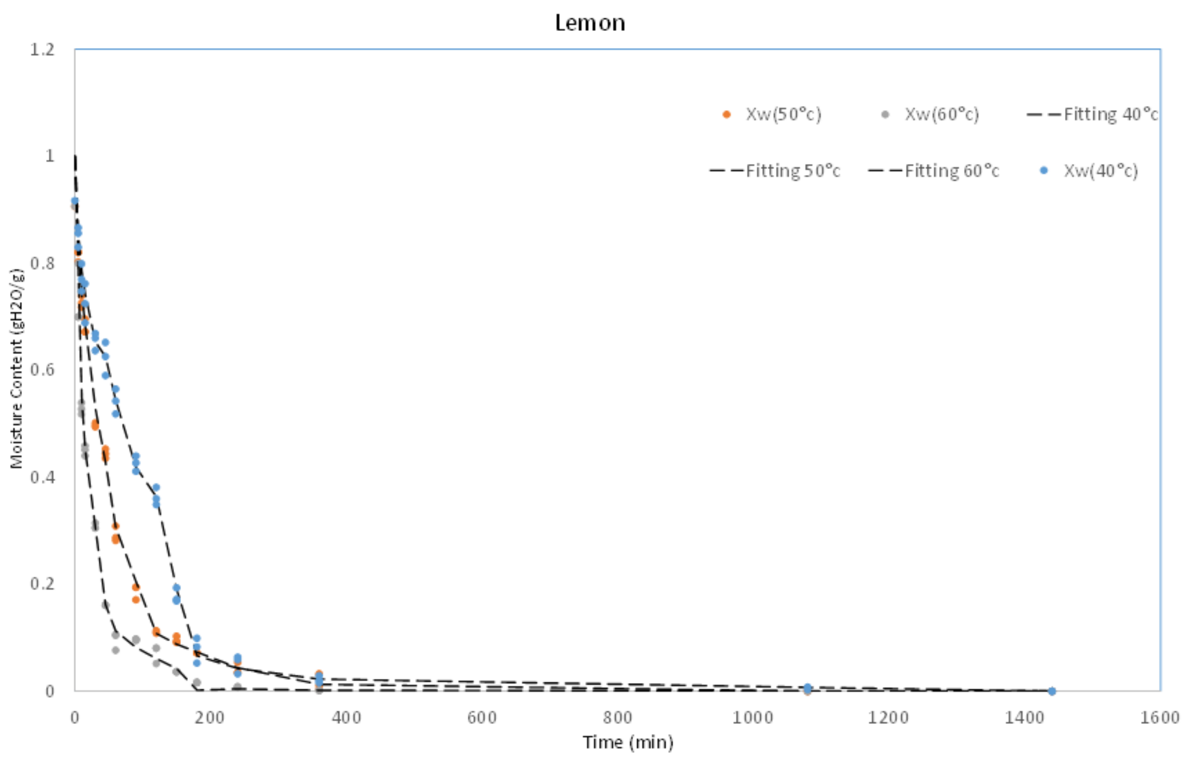

Figure 2: Dehydration curve of lemon by-products

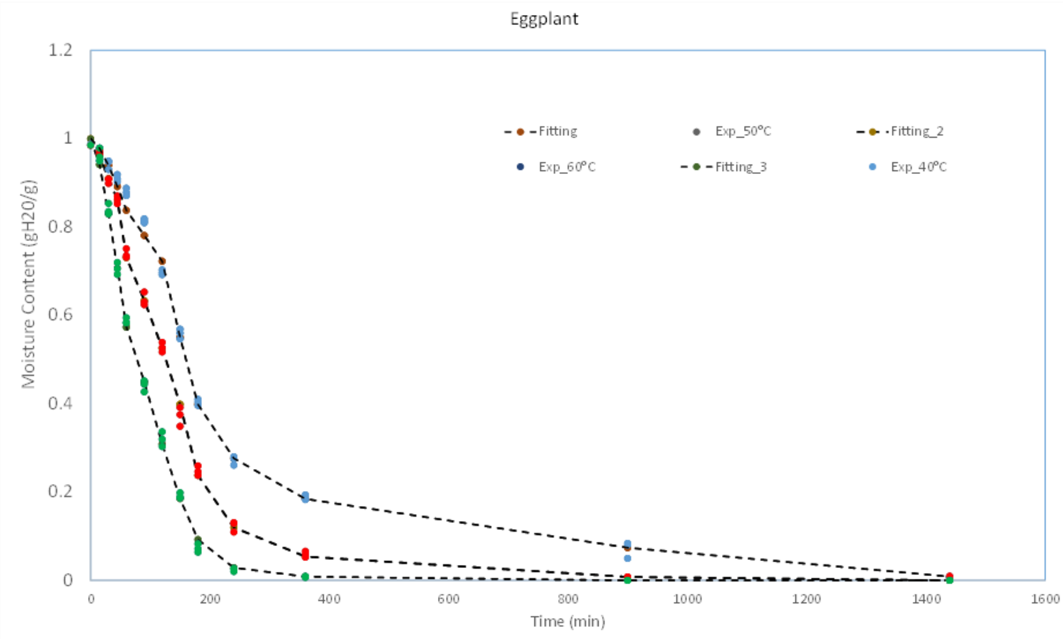

Figure 3: Dehydration curve of eggplant by-products

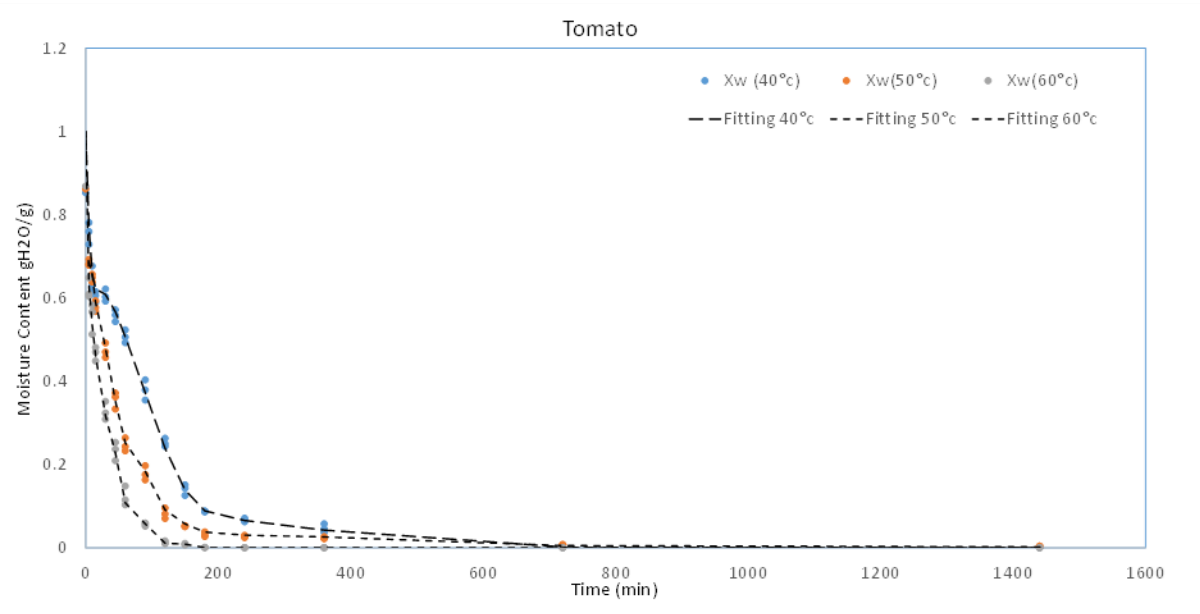

Figure 4: Dehydration curve of tomato by-products

The dehydration curves generated from the experimental data were effectively characterized using Page's model. This model demonstrated a high degree of fit, as evidenced by the correlation coefficients, r^2, exceeding 0.98 across all temperature conditions and for each type of vegetable. This high correlation coefficient signifies a strong linear relationship between the observed and predicted values, underscoring the model's robustness and ability to represent the drying kinetics of the vegetables accurately.

Table 1 – Estimated parameters regarding the dehydration treatment of external leaves, no edible, of artichokes.

Parameters	*Estimates*	*Standard error*	*p-value*	r^2
Dehydration temperature = 40°C				*0.985*
k (min^{-1})	*0.000588*	*0.00028*	*<0.01*	
n (dimensionless)	*1.41*	*0.0916*	*<0.01*	
Dehydration temperature = 50°C				*0.992*
k (min^{-1})	*0.000587*	*0.000270*	*<0.01*	
n(dimensionless)	*1.63*	*0.1014*	*<0.01*	
Dehydration temperature = 60°C				*0.992*
k (min^{-1})	*0.000551*	*0.000356*	*<0.01*	
n(dimensionless)	*1.83*	*0.161*	*<0.01*	

Table 2 – Estimated parameters regarding the dehydration treatment of external leaves, no edible, of eggplants.

Parameters	*Estimates*	*Standard error*	*p-value*	r^2
Dehydration temperature = 40°C				*0.987*
k (min^{-1})	*0.000023*	*0.00001*	*<0.01*	
n (dimensionless)	*2.39*	*0.2725*	*<0.01*	
Dehydration temperature = 50°C				*0.991*
k (min^{-1})	*0.000338*	*0.000270*	*<0.01*	
n(dimensionless)	*1.74*	*0.1014*	*<0.01*	
Dehydration temperature = 60°C				*0.998*
k (min^{-1})	*0.000004*	*0.000*	*<0.01*	
n(dimensionless)	*3.165*	*0.2586*	*<0.01*	

Table 3 – Estimated parameters regarding the dehydration treatment of external leaves, no edible, of lemons.

Parameters	*Estimates*	*Standard error*	*p-value*	r^2
Dehydration temperature = 40°C				*0.989*
k (min^{-1})	*0.03853*	*0.006497*	*<0.01*	
n (dimensionless)	*0.6599*	*0.03529*	*<0.01*	
Dehydration temperature = 50°C				*0.992*
k (min^{-1})	*0.034*	*0.006403*	*<0.01*	
n(dimensionless)	*0.753830*	*0.04153*	*<0.01*	
Dehydration temperature = 60°C				*0.9995*
k (min^{-1})	*0.04054*	*0.00739*	*<0.01*	
n(dimensionless)	*0.8145*	*0.04418*	*<0.01*	

Table 4 – Estimated parameters regarding the dehydration treatment of external leaves, no edible, of tomatoes.

Parameters	*Estimates*	*Standard error*	*p-value*	r^2
Dehydration temperature = 40°C				*0.991*
k (min^{-1})	*0.1199*	*0.01636*	*<0.01*	
n (dimensionless)	*0.614948*	*0.036725*	*<0.01*	
Dehydration temperature = 50°C				*0.993*
k (min^{-1})	*0.1277*	*0.01697*	*<0.01*	
n(dimensionless)	*0.581469*	*0.034707*	*<0.01*	
Dehydration temperature = 60°C				*0.9992*
k (min^{-1})	*0.130931*	*0.01832*	*<0.01*	
n(dimensionless)	*0.5613*	*0.035762*	*<0.01*	

Figure 5 visually represents the dehydration curve and the desorption isotherm for the outer leaves of artichokes subjected to air-drying at 50°C for 7 hours.

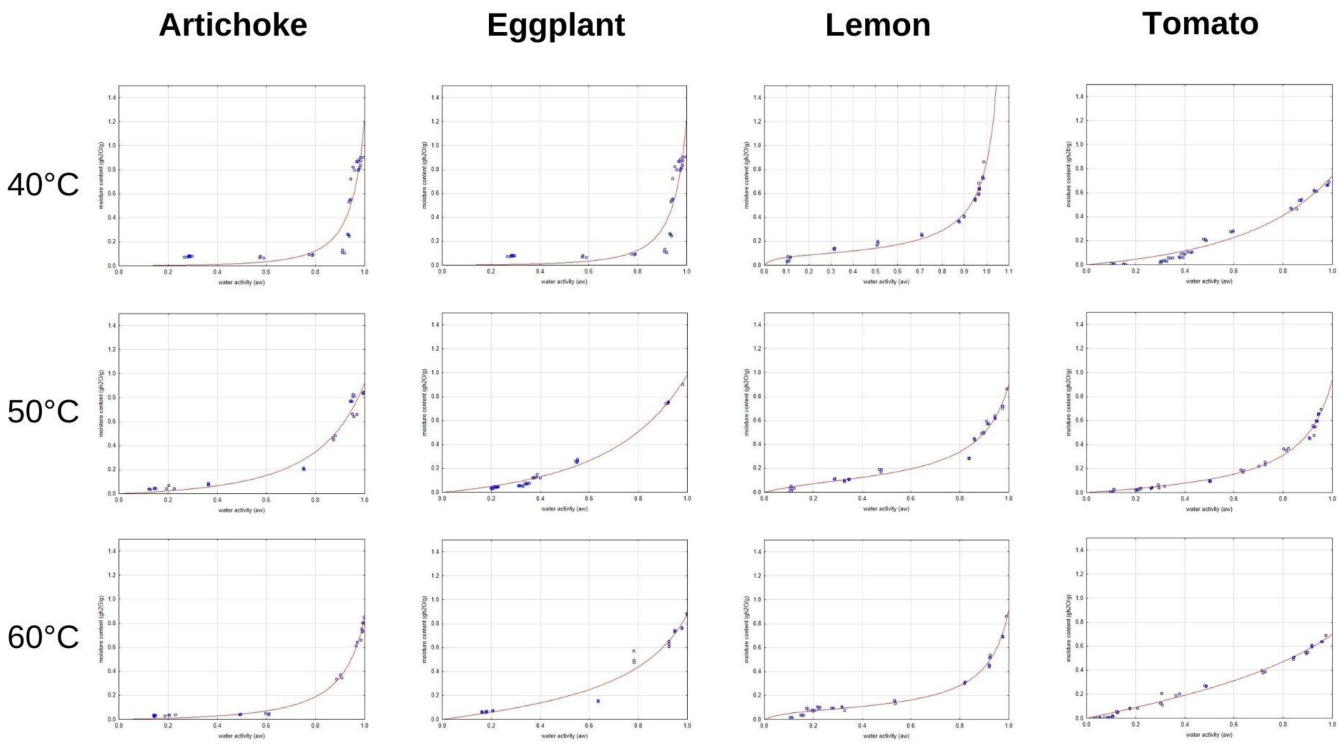

Figure 5: Dehydration and isotherm curves for external not-edible artichokes leaves.

The successful application of Page's model to the experimental data, coupled with the high correlation coefficients obtained, underscores the Page model's efficacy in describing the dehydration process of artichokes, eggplants, lemons, and tomatoes.

Following the mathematical characterization of the drying process, subsequent experiments were conducted to validate the estimated results derived from the Page's model. This validation process was designed to ascertain the model's predictive accuracy. These validation experiments focused on determining the drying times required to achieve a target moisture content of 5% and a water activity below 0.4. These targets were chosen based on standard industry benchmarks for safe and effective food preservation.

Table 5 – Estimated parameters of lemon samples regarding the GAB equation.

Parameters	*Time (minute)*	*m*	*C*	*K*
Dehydration temperature = 40°C				
Estimated	*121.795*	*0.08087*	*21.71266*	*0.90636*
Actual	*120*			
Dehydration temperature = 50°C				
Estimated	*78.9964*	*0.106293*	*5.683932*	*0.884748*
Actual	*80*			
Dehydration temperature = 60°C				
Estimated	*45.974*	*0.7807*	*13.76783*	*0.91572*
Actual	*45*			

Table 6 – Estimated parameters of tomato samples regarding the GAB equation.

Parameters	*Time (minute)*	*m*	*C*	*K*
Dehydration temperature = 40°C				
Estimated	*32.0147*	*3.335902*	*0.106221*	*0.525559*
Actual	*31*			
Dehydration temperature = 50°C				
Estimated	*29.6080*	*0.117188*	*1.417087*	*0.885706*
Actual	*28*			
Dehydration temperature = 60°C				
Estimated	*23.2852*	*0.437504*	*1.766968*	*0.563633*
Actual	*21*			

Table 7 – Estimated parameters of eggplant samples regarding the GAB equation.

Parameters	*Time (minute)*	*m*	*C*	*K*
Dehydration temperature = 40°C				
Estimated	*181.06*	*2.088959*	*0.008917*	*0.885226*
Actual	*185*			
Dehydration temperature = 50°C				
Estimated	*128.875*	*2.95722*	*0.11567*	*0.58223*
Actual	*125*			
Dehydration temperature = 60°C				
Estimated	*98.7020*	*0.229672*	*1.574782*	*0.778402*
Actual	*95*			

Table 8 – Estimated parameters of artichoke samples regarding the GAB equation.

Parameters	*Time (minute)*	*m*	*C*	*K*
Dehydration temperature = 40°C				
Estimated	*178.4354*	*2.449740*	*0.005561*	*0.902097*
Actual	*180*			
Dehydration temperature = 50°C				
Estimated	*91.495*	*3.466957*	*0.035534*	*0.705283*
Actual	*90*			
Dehydration temperature = 60°C				
Estimated	*66.687*	*0.109082*	*0.360787*	*0.902046*
Actual	*65*			

The outcomes of these validation experiments demonstrated the robustness and reliability of the mathematical modeling approach employed in this study. The discrepancies observed between the estimated and experimental data were minimal, indicating the Page's model's high degree of predictive accuracy.

Mathematical modeling has elucidated that the discrepancies between the forecasted and computed durations are remarkably minimal.
An examination of the mean difference across all temperature times can be conducted to succinctly comprehend the variations between the predicted and calculated times. This mean difference is 2 min 5 sec for lemon, 2 min 40 sec for tomato, 3 min 40 sec for eggplant, and 2 min 30 sec for artichoke. Considering the duration required to attain an identical water activity at disparate temperatures, the time differences procured testify to the mathematical computation's precision and accuracy. These differences underscore the satisfactory nature of the mathematical model in accurately predicting the time required to reach a specific water activity level for different food items.

3.2 Food powders and food-ink preparation

After dehydration, the by-products from artichokes, eggplant and tomatoes have been submitted to the grinding process by using a semi-professional grinder that allows modulating the particle size. More specifically three different levels of grinding have been used – fine (<200μm), medium (<400μm) and ground (<600μm) – and the obtained powders have been studied by analyzing the granulometric curve (Analysette, 22, Fritsch).

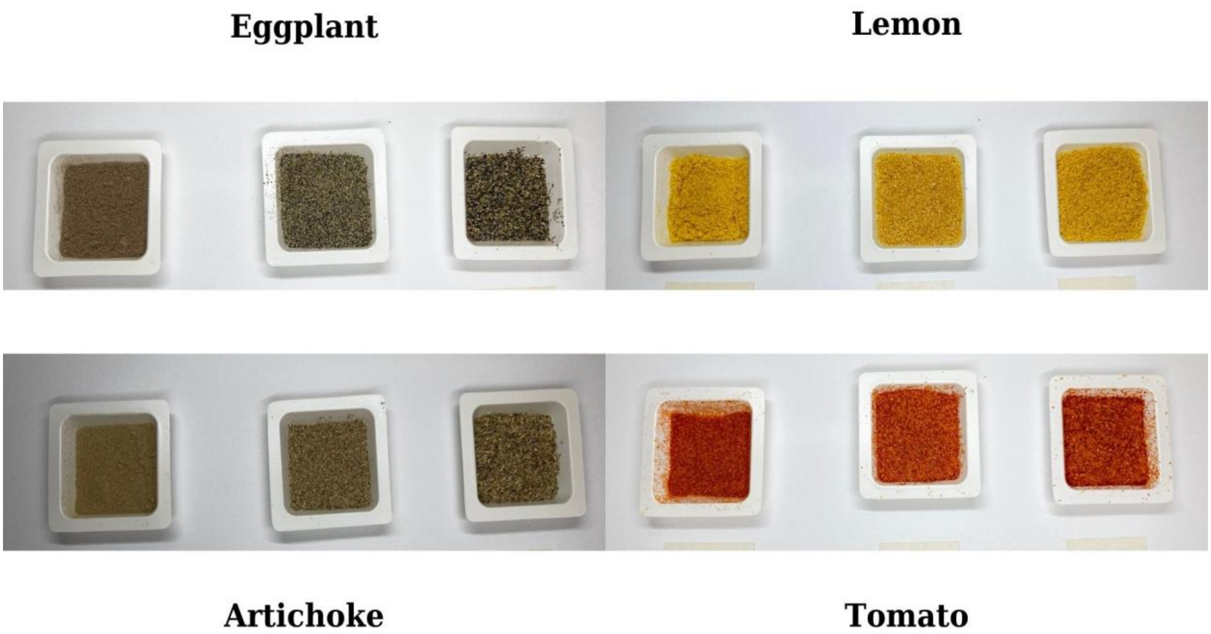

Figure 2: Eggplant, lemon, artichoke and tomato powder samples sieved from left to right as fine (<200μm), medium (<400μm) and ground (<600μm).

Food-inks have been intended as new food formulas obtained in different modalities and served as two different levels of complexity which is food-inks based on hydrocolloids enriched with innovative food powders.

The following figures show the results of the granulometric analysis of different particle sizes distribution of the powders.

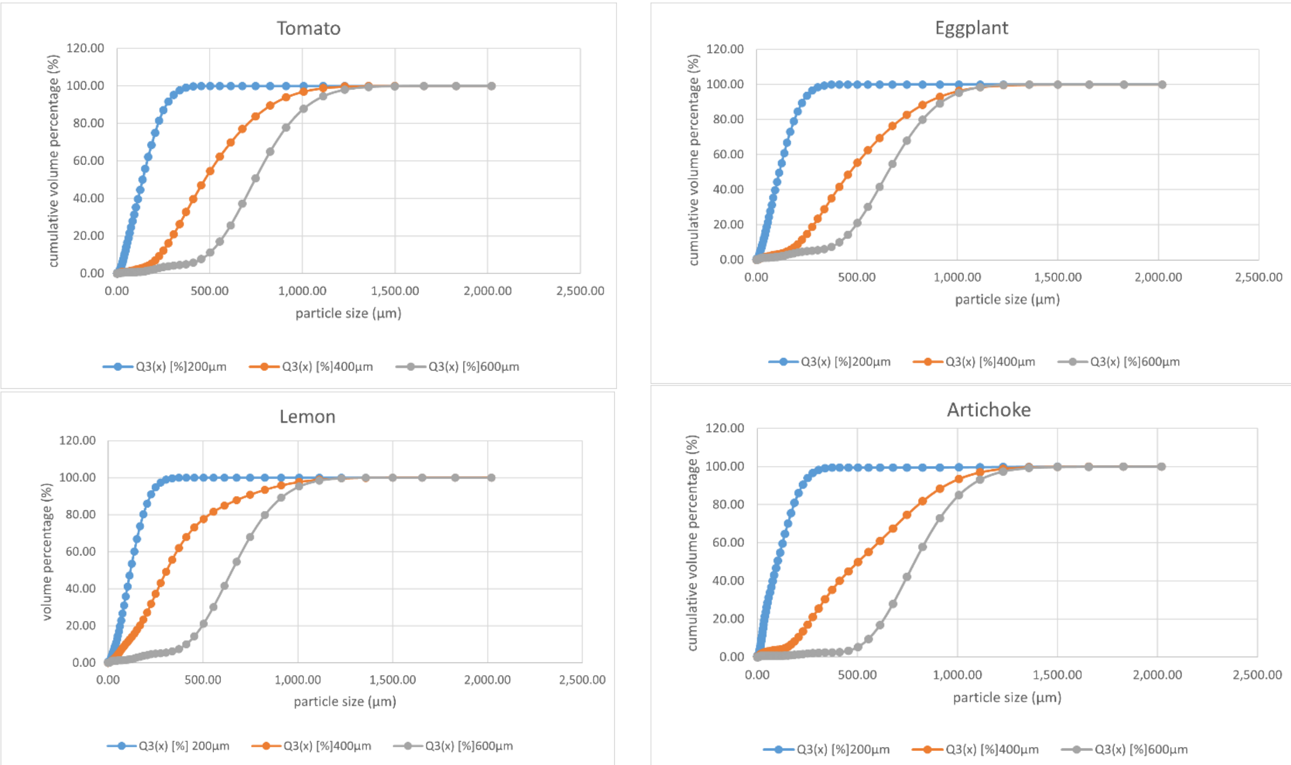

Figure 3a: Cumulative volume distribution function graphics.

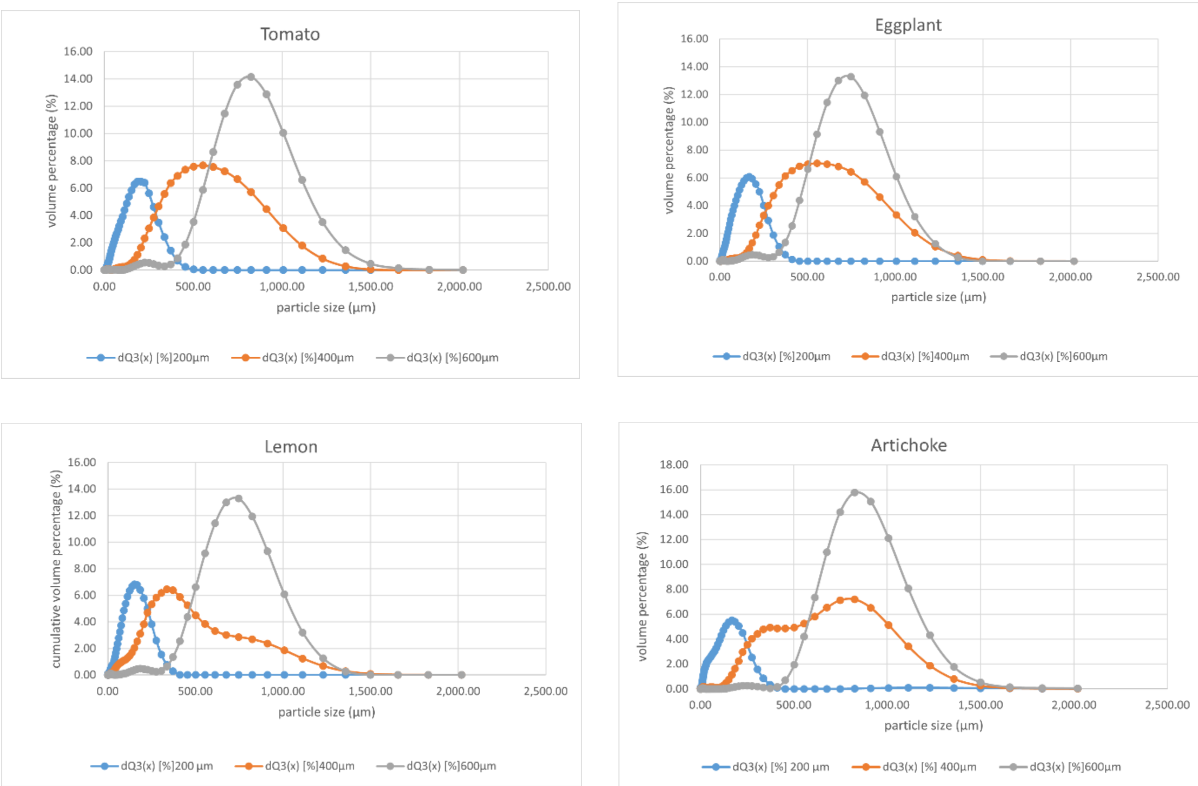

Figure 3b: Differential volume distribution function.

The granulometric analysis of Artichoke, Eggplant, Tomato, and Lemon samples at three different sizes (200 µm, 400 µm, and 600 µm) provides valuable insights into the particle size distribution within these materials. The data indicate a general trend of increasing particle size distribution from the 200µm to the 600µm samples across all materials, suggesting that the materials' preparation method or inherent properties may influence the particle size range.

The analysis also reveals that the distribution of particle sizes within each sample is unique, with each material showing different patterns of differential and cumulative volume percentages.

3.2. 3D Printing Experiments with vegetable and fruit Powder

A study investigated the potential of incorporating dried tomato and lemon peel powders into a gel matrix prepared from wheat starch for 3D food printing applications. The powders derived from these food by-products were categorized into three distinct sizes: fine (<200µm), medium (<400µm), and ground (<600µm). Each size was separately mixed with the gel matrix at two different concentrations, 2% and 5%, to examine the effect of particle size and concentration on the printability of the gel.

Subsequently, 3D models were printed using these gel mixtures at two different infill levels, 10% and 40%. The results showed that the gels prepared with 2% and 5% of fine (<200µm) and medium (<400µm) powders were successfully printed without any issues (Figure 4). This successful printing was attributed to the nozzle size being larger than the particle size, allowing for a smooth flow of the gel mixture through the nozzle.

However, the printing process did not yield satisfactory results when the ground (<600µm) powder was used for gel preparation. The larger particle size led to particles clumping together within the nozzle, causing blockages (Figure 4). This resulted in excessive or insufficient flow, leading to significant discrepancies between the digital design and the printed product.

As demonstrated in this study, the utilization of food waste and non-edible by-products in 3D food printing offers a promising avenue for developing personalized food solutions. This innovative approach promotes sustainability through waste reduction and provides a unique opportunityto tailor food products to individual dietary needs and preferences.

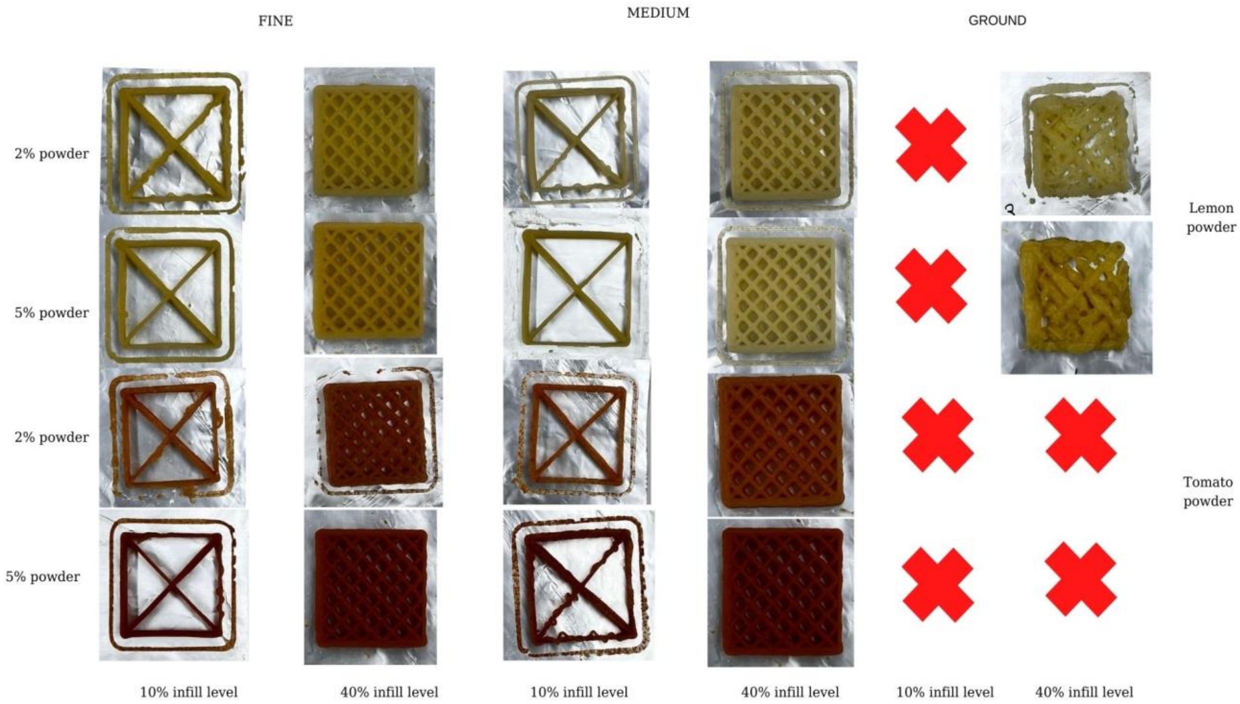

Figure 4: 3D food printer printing results of rectangular shapes with 10% and 40% infill levels with gels prepared from lemon and tomato powders in sizes (<200µm), medium (<400µm), and ground (<600µm).

4. Conclusion

This study comprehensively examines the drying kinetics of non-edible parts of artichokes, eggplants, lemons, and tomatoes under varying temperature conditions. The meticulous experimental procedure and rigorous data collection strategy have yielded a robust dataset, effectively characterised using Page's model. The high correlation coefficients obtained across all temperature conditions and vegetable types underscore the model's robustness and ability to represent the drying kinetics of the vegetables accurately. The validation experiments further attest to the model's high predictive accuracy, with minimal discrepancies between the estimated and experimental data.

The subsequent processing of the dehydrated by-products into food powders and their incorporation into food inks for 3D food printing applications represents an innovative approach to food waste utilisation. The granulometric analysis of the powders and the successful 3D printing of gels prepared with fine and medium powders demonstrate the potential of this approach in developing personalised food solutions. However, the challenges encountered with the ground powder highlight the need for further particle size optimisation for successful 3D printing.

Overall, this research contributes significantly to the broader understanding of food preservation techniques and the potential of food waste utilisation in 3D printing. It provides a robust foundation for future studies to improve the efficiency and sustainability of food processing operations and develop personalised food solutions.

5. References

1. Chan, H. K., Griffin, J., Lim, J. J., Zeng, F., & Chiu, A. S. (2018). The impact of 3D Printing Technology on the supply chain: Manufacturing and legal perspectives. International Journal of Production Economics, 205, 156-162. https://doi.org/10.1016/j.ijpe.2018.09.009

2. Derossi, A., Caporizzi, R., Oral, M., & Severini, C. (2020). Analyzing the effects of 3D printing process per se on the microstructure and mechanical properties of cereal food products. Innovative Food Science & Emerging Technologies, 66, 102531. https://doi.org/10.1016/j.ifset.2020.102531

3. Derossi, A., Caporizzi, R., Paolillo, M., Oral, M. O., & Severini, C. (2021). Drawing the scientific landscape of 3D Food Printing. Maps and interpretation of the global information in the first 13 years of detailed experiments, from 2007 to 2020. Innovative Food Science & Emerging Technologies, 70, 102689. https://doi.org/10.1016/j.ifset.2021.102689

4. Derossi, A., Paolillo, M., Verboven, P., Nicolai, B., & Severini, C. (2022). Extending 3D food printing application: Apple tissue microstructure as a digital model to create innovative cereal-based snacks. Journal of Food Engineering, 316, 110845. https://doi.org/10.1016/j.jfoodeng.2021.110845.

5. German, J. B., Zivkovic, A. M., Dallas, D. C., & Smilowitz, J. T. (2011). Nutrigenomics and Personalized Diets: What Will They Mean for Food? https://doi.org/10.1146/annurev.food.102308.124147.

6. Gholamipour-Shirazi A, Kamlow MA, T Norton I, Mills T. (2020). How to Formulate for Structure and Texture via Medium of Additive Manufacturing-A Review. Foods. 2020 Apr 15;9(4):497. https://doi.org/10.3390/foods9040497

7. Girotto, F., Alibardi, L., & Cossu, R. (2015). Food waste generation and industrial uses: A review. Waste Management, 45, 32-41. https://doi.org/10.1016/j.wasman.2015.06.008.

8. Jangam SV, Law CL, Mujumdar AS (2010) Drying foods, vegetables and fruits (Vol.1, p.202) Singapore:TPR Group).

9. Johnson, L.K., Bloom, D.J., Dunning, R.D., Gunter, C.C., Boyette, M.D., Creamer, N.G., 2019. Farmer harvest decisions and vegetable loss in primary production. Agricultural systems, 176, 102672. https://doi.org/10.1016/j.agsy.2019.102672.

10. Lebersorger, S., Schneider, F., 2014. Food loss rates at the food retail, influencing factors and reasons as a basis for waste prevention measures. Waste management, 34, 1911 – 1919. https://dx.doi.org/10.1016/j.wasman.2014.06.013.

11. Levis, J.W, Barlaz, M.A., Themelis, N.J., Ulloa, P., 2010. Assessment of the state of food waste treatment in the United States and Canada. Waste Manage, 30, 1486-1494.

12. Love, D.C., Fry, J.P., Milli, M.C., Neff, R.A., 2015. Wasted seafood in the United States: quantifying loss from production to consumption and moving toward solutions. Global environmental change, 35, 116 – 124. https://doi.org/10.1016/j.gloenvcha.2015.08.013.

13. Maroulis, Z., Tsami, E., Marinos-Kouris,D., & Saravacos, G. (1988). Application of the GAB model to the moisture sorption isotherms for dried fruits. Journal of Food Engineering, 7(1), 63-78. https://doi.org/10.1016/0260-8774(88)90069-6

14. Nguyen VA, Ramanathan M. (2020). Application of Brunauer-Emmett-Teller (BET) theory and the Guggenheim-Anderson-de Boer (GAB) equation for concentration-dependent, non-saturable cell-cell interaction dose-responses. J Pharmacokinet Pharmacodyn. 2020 Dec;47(6):561-572. doi: 10.1007/s10928-020-09708-x.

15. Oral, M., Derossi, A., Caporizzi, R., & Severini, C. (2021). Analyzing the most promising innovations in food printing. Programmable food texture and 4D foods. Future Foods, 4, 100093. https://doi.org/10.1016/j.fufo.2021.100093

16. Philippidis, G., Sartori, M., Ferrari, E., M'Barek, R., 2019. Waste not, want not: a bio-economic impact assessment of household food waste reductions in the EU. Resources, conservation & recycling, 146, 514 – 522. https://doi.org/10.1016/j.resconrec.2019.04.016.

17. Redlingshöfer, B., Coudurier, B., Georget, M., 2017. Quantifying food loss during primary production and processing in France. Journal of cleaner production, 164, 703 – 714. https://dx.doi.org/10.1016/j.clepro.2017.06.173.

18. Revilla, B.P., Salet, W., 2018. The social meaning and function of household food rituals in preventing food waste. Journal of cleaner production, 198, 320 – 332. https://doi.org/10.1016/j.clepro.2018.06.038.

19. Severini, C., Caporizzi, R., Fiore, A. G., Ricci, I., Onur, O. M., & Derossi, A. (2020). Reuse of spent espresso coffee as sustainable source of fibre and antioxidants. A map on functional, microstructure and sensory effects of novel enriched muffins. LWT, 119, 108877. https://doi.org/10.1016/j.lwt.2019.108877

20. van Dooren, C., Janmaat, O., Snoek, J., Schrijnen, M., 2019. Measuring food waste in Dutch households: A synthesis of three studies. Waste management, 94, 153 – 164. https://doi.org/10.1016/j.wasman.2019.05.025.

Chapter 7 Conclusion

This doctoral research journey has ventured into the uncharted territories of 3D Food Printing (3DFP), a transformative technology with the potential to redefine the food industry. The primary objective was to investigate the potential of 3DFP in creating personalized foods with unique sensorial and nutritional properties and to identify the current challenges and opportunities in this field.

The research has unequivocally demonstrated that 3DFP is not merely a tool for producing intricate geometries but a technology capable of reducing food waste and creating customized food products. Exploring recent advancements in this field, such as 4D Food Printing and programmable food texture, has broadened the horizons for creating food products with a wide range of quality parameters and addressing mastication and swallowing difficulties in vulnerable consumers.

The global production of scientific documents on 3DFP was analyzed in depth, revealing that while China and New zealand are leading in this field, there needs to be more international collaborations. This research underscores the need for fostering global partnerships to expedite the implementation of 3D printing in the food sector. The need for further experiments on alternative printing methods like Selective Laser Sintering (SLS) and Hot Air Sintering (HAS) was also highlighted, which could introduce novelties such as fast printing and dehydrated food-ink with extended shelf life.

The empirical investigations have provided valuable insights into the impact of the 3D printing process on the morphological, microstructural, and mechanical properties of food products. The discrepancies between 3D printed snacks and the virtual model, mainly in the top layers, were attributed to the imbalance between the layer height and extrusion rate, affecting the distribution of pores in the 3D printed samples.

Furthermore, the research has demonstrated the potential of 3D printing technology in defining optimal 3D food architectures to minimize the baking time of biscuits. A significant reduction in the acrylamide content in the 3D-printed biscuits was also found, indicating the potential of 3D printing to improve the chemical safety of food products.

During this research, an innovative approach for creating GCodes in 3DFP was tested, which improved the quality of 3D-printed food and increased the efficiency of the process. This novel approach could lead to innovative food products with inhomogeneous structures capable of conveying new sensory perceptions and facilitating mastication for elderly people.

In conclusion, integrating food waste and non-edible by-products into 3DFP presents a transformative approach to food production and consumption. This innovative methodology addresses food waste and provides a unique platform for creating personalized food products tailored to individual dietary needs and preferences. As we look to the future, optimizing the particle size and concentration of different food by-products will be crucial to ensure the printability and sensory acceptability of the printed food. This research underscores the potential of 3DFP as a tool for sustainable and personalized food production, paving the way for a future where food waste is minimized, dietary needs are met with precision, and the enjoyment of food consumption is enhanced through innovative and appealing designs.