





EDITOR-IN-CHIEF'S WORD

In this issue the HATZ Bulletin presents scientific and professional achievements of our members too, especially in important areas of the development of the Croatian and world society. That is why we have asked our member Prof. Stela Jokić PhD, full professor at the Faculty of Food Technology of the University J.J. Strossmayer Osijek, to present in this issue the current considerations and works of her team in the field of food industry as an essential part of human survival. I believe that the contributions presented in this issue will be useful and interesting for the readers to gain new insights about this field.

Editor-in-Chief Vladimir Andročec, President of the Croatian Academy of Engineering



EDITOR'S WORD

Dear readers.

It is my pleasure to present in this issue of the HATZ-Bulletin Engineering Power the continuation of our efforts to present the activities of the research groups on various topics carried out by members of the Academy from various Croatian universities.

The guest editor of this issue is Stela Jokić, Full Professor at the Faculty of Food Technology, Josip Juraj Strossmayer University of Osijek and Associate Member of the Academy in the Department of Bioprocess Engineering. The topic of the issue relates to current trends in food science and technology.

Editor

Zdravko Terze, Vice-President of the Croatian Academy of Engineering



FOREWORD

Dear Readers,

Never before have there been so many demands on the food industry as today. On the one hand, in addition to the production of safe and tasty food worldwide, the production of "functional food" or food that has a positive effect on human health is in the focus of the development of new products in the food industry. The production of food/products with added value in terms of maintaining the health and improved consumers' diets and promotion of good health and nutrition is a must.

On the other hand, an important aspect today is "zero-waste approach" and a high interest in the possible use and application of different by-products, especially from the food industry. By-product utilization has become one of the fastest growing areas of research because it represents a cheap and nutritiously highly valuable raw material, along which efficient waste management is achieved through their use in subsequent industrial processes. Food by-products often contain significantly high amounts of health-promoting compounds (bioactive components, vitamins, minerals, etc.) due to which they represent a highly valuable raw material for the production and development of new products.

For all the above reasons, scientists work with the food industry to solve the problems related to the consumption of unhealthy foods and to produce new products with increased nutritional value, while achieving a "zero waste" approach.

Bearing in mind that this is a very dynamic and vast area, and although the authors of this issue have made great efforts to present the latest trends in this field, only a few of the areas are presented. This special issue on "Trends in food science and technology" therefore highlights some of the current challenges facing the food industry and trends towards their solution. Topics include new or novel raw materials including bioactive compounds such as marine macroalgae as "superfoods" due to their nutritional value; some innovations in the food industry; the use of waste as a potential source of functional food ingredients; and finally three-dimensional (3DP) food printing as a potential solution to further develop current food processing techniques.

I would like to thank all the authors who contributed to this special issue and to my colleague Prof. Đurđica Ačkar PhD for her support and help in collecting the papers.

I hope you will enjoy reading these articles! Yours sincerely

Guest-Editor

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Challenges Facing the Food Industry

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Abstract

Never before have so many demands been made on the food industry as today. In addition to producing safe and tasty food, the food industry faces the challenge of making food functional, easy to prepare and convenient, with a "zero waste" approach, both in the use of raw materials and in packaging. Food is not only fuel, but it has been given a role in preventing various diseases and improving human health, preferably finely tuned to personal needs and desires. For all these reasons, scientists are working with the food industry to solve problems associated with the consumption of unhealthy foods and to produce new products with increased nutritional value. Furthermore, several new strategies have recently been implemented to address the problem of negative environmental impacts and to maintain sustainability in the food industry. Therefore, the aim of this paper is to describe some of the contemporary challenges of the food industry and trends towards their solution.

1. Introduction

The food industry is one of the most important branches of the national economies, in the European Union as well as in the World in general, which plays a central role for the processing of agricultural raw materials and food supply (Bigliardi and Galati, 2013). This industry has changed and developed over the decades in order to satisfy customer needs and consumer behavior. It is characterized by a complex system of activities concerning supply, consumption and delivery of food products across the entire globe. While people in the past had to fight to find their daily meals and used to eat only local and seasonal products, nowadays people in the Western world are surrounded by different types, qualities and prices of food products (Recordati, 2015). Furthermore, the situation has changed drastically, especially in the 20th century, as a result of civilization, industrialization, technological innovations, mechanization, the economic growth as well as increasing the world's population. The world's population is now more than 7.7 billion persons, and this number is presently growing at a rate of around

1.07% per year (Fasolin et al., 2019), which generates a growing demand for food globally (Ramos et al., 2020). While on the one hand food industry must produce enough quantity of products, on the other hand it is well established that poor diet is a major contributor to many conditions including cardiovascular disease, obesity, type 2 diabetes, dental caries, and some cancers (Crawford, 2020). According to all the above, the aim of this paper is to highlight some of the challenges facing the food industry today and trends in addressing them.

2. Contemporary challenges of the food industry

Until the end of the 20th century, the prevalent policy was mainly to increase food production, without improving the efficiency of the food systems (Biglardi and Galanakis, 2020). This fact caused an increase in food loss and food waste, where the amount of lost or wasted food accounts 1.3 billion tonnes globally each year

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(Galanakis, 2020). This situation has triggered a series of environmental challenges that could threaten human survival. Climate change, loss of biodiversity, accumulation of waste, resource depletion (water, fossil fuels, etc.), growing disease pandemics, and the modification of natural ecosystems, are some of the results of human activities (Khoo and Knorr, 2014; Ramos et al., 2020). One of the main manufacturing industries is the agrifood industry, which is responsible for 25% of the total emissions of greenhouse gases (Ramos et al., 2020). Therefore, improving food production systems is one of the major actions that needs to be done urgently to reduce the global impacts on the environment.

Recently, three main innovation directions for the future food systems were highlighted: 1) sustainability and higher efficiency – production at the lowest possible cost; 2) innovation opportunity – refer to new scientific and technical approaches in food processing, and to the introduction of novel foods; and 3) development of functional foods focused on the health and well-being after consumption (Fasolin et al., 2019).

2.1. Sustainability and higher efficiency

Sustainable food systems are those that ensure food security and nutrition for all in a way that economic, social and environmental sustainability is not compromised for future generations (Fasolin et al., 2019). Therefore,

maintaining sustainability in food industry requires the maximum utilization of all raw materials and integration of activities throughout all the production-to-consumption stages. The production stage is the first one, where the efforts begin with activities of reducing postharvest losses and increment of waste valorisation. Furthermore, efforts are underway to ensure that the energy, water, and other resources are used most efficiently, and environmental impacts are minimized (Biglardi and Galanakis, 2020). One of the new strategies which could be used to reduce environmental impacts is Cleaner Production (CP) methodology, which in food industries focuses on minimization of resource consumption, reduction of the waste generation, a better use of food by-products looking for increasing the process efficiency (Ramos et al., 2020). On the other hand, future perspectives for the food industries is the application of Industry 4.0 concept, which proposes an interconnected production, through the information exchange (smart data) of all the links of the supply chain, connecting workers, devices, materials, processes, logistics or consumers. According to this strategy, the company could provide and receive information in real time about their own processes and the rest of the links of the value chain, adjusting their production to unforeseen events (Figure 1). This leads to the optimization of the inputs needed for the production and thus to the minimization of environmental impacts related to the efficiency of water, energy or raw material (Ramos et al., 2020).

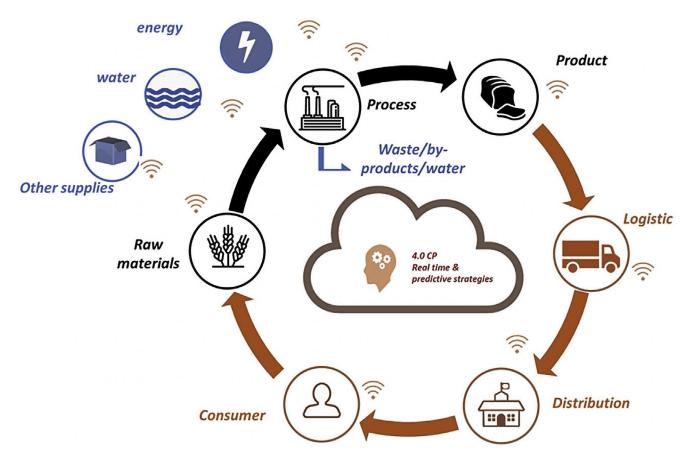


Fig. 1. Application of Industry 4.0 strategy in food industry (adopted from Ramos et al., 2020)

2.2. Innovation in the food industry

In the past, the food industry has traditionally focused on minimizing production line costs, giving little attention to the real needs of consumers. Thereafter, the food chain is reversed, the need for food, based on the offer was transferred to a concept based on demand. So consumers tell producers what they want to eat. According to this trend, modern innovations in food companies are implemented in various ways (Biglardi and Galanakis, 2020). Innovations can occur in all parts of the food chain and the possible classification of food innovations is as follows: (1) new food ingredients and materials, (2) innovations in fresh foods, (3) new food processing techniques, (4) innovations in food quality, (5) new packaging materials and methods, and (6) new methods of distribution or retailing (Bigliardi and Galati, 2013). However, most research follows the classification in process and product innovations. Process innovations represent the customization of existing production lines, as well as the installation of a completely new infrastructure and the implementation of new technologies that enable the creation of new products. Their main aims are to improve product quality and production process in terms of time, cost and flexibility (Biglardi and Galanakis, 2020). In this regard, so-called emerging (mainly non-thermal) processing technologies have been gaining interest among food researchers, due to their lower impact on nutritional and sensory properties of the products (Barba et al., 2016). The most popular emerging technologies examined in the of food science are: pulsed UV-light, pulsed electric fields (PEF), irradiation, cold plasma (CP), high hydrostatic pressure (HPP) and ultrasound (US) (Zhang et al., 2018), as well as radio-frequency drying, electro-osmotic dewatering, pressurized extraction, high voltage electrical discharge, nanoencapsulation and others (Biglardi and Galanakis, 2020). Another important example of innovations in the food industry is known as "food waste recovery". This kind of innovations is based on valorisation of food industry by-products, as a source of high added-value micromolecules (e.g., antioxidants, polyphenols, carotenoids) and macromolecules (e.g., dietary fiber, pectin, β-glucan, proteins). In connection with this, it is important to highlight two recently published books: "Some possibilities for utilization of food industry by-products" (Šubarić, 2017) with 13 chapters and "Some possibilities for utilization of food industry by-products – Book 2" with 18 chapters (Šubarić and Babić, 2019), outlining the potential for utilization of various food by-products.

2.3. Development of functional foods

Although the food is primary fuel and one of the major human needs which is used to eliminate hunger and obtain energy for the body, in recent years the lifestyle of people and desire for a healthier life have changed the philosophy of food. Thus, the food has been given a role in preventing various diseases and improving human health. These foods are called functional foods. Functional foods are defined as the foods or food elements that provide additional benefits for human physiology and metabolic functions and help to reduce the occurrence of disease (Santeramo et al., 2018; Lule et al., 2019). Functional foods can be the foods which are natural. fortified, enriched, or contain functional ingredients (Guo, 2009). Functional foods have been developed almost in all food categories. However, among all the food products, functional foods are mainly launched in dairy, confectionery, soft drinks, bakery and baby foods (Bigliardi and Galati, 2013). There are different classifications of functional foods in the literature, whereby the classification from the product point of view includes the following types: 1) fortified products - food fortified with additional nutrients (vitamin C, vitamin E, folic acid, zinc, calcium, etc.); 2) enriched products - food with additional new nutrients or components normally not found in a particular food (probiotics and prebiotics); 3) altered products - food from which a harmful component has been removed, reduced or replaced by another with beneficial effects (dietary fibers as fat replacers); 4) enhanced commodities - food in which one of the components have been naturally enhanced (eggs with increased omega-3 content) (Bigliardi and Galati, 2013). Regardless of the type of functional food, it is important to emphasize that awareness for health benefits of some functional foods are gaining ground. Guo (2009) found that the sources of information about these benefits come primarily from different media, accounting for 72%, while medical sources ranked second with 44%, and 20% obtained from friends and family or self, while diet and health books represented 13%. Among the functional products that have been particularly attractive in recent times, a distinction could be made between products with an increased content of proteins, dietary fibres or active components such as polyphenols.

In the case of protein, when thinking about replacing conventional proteins (e.g., meat and egg proteins), the main groups of emerging and sustainable proteins are: vegetable proteins (seeds, legumes, nuts, fruits and vegetables or vegetable-based by-products such as apple pomace, orange pulp, oat bran, sugar beet pulp and brewer's spent grain); insect proteins (*Coleoptera* (31%), *Lepidoptera* (18%), *Hymenoptera* (14%), *Orthoptera* (13%) and *Hemiptera* (10%) (Sun-Waterhouse et al., 2016)); microbial proteins (microalgae, fungi and bacteria) and milk proteins (whey proteins) (Fasolin et al., 2019).

It is known that the consumption of dietary fibers in sufficient quantity is necessary for the function of the gastrointestinal tract. Furthermore, the reductions in LDL-cholesterol, attenuating glycemic and insulin response, increasing stool bulk, and improving laxation have been associated with dietary fiber intake through the consumption of foods rich in this dietary component. It includes polysaccharides, lignin and associated plant substances, whereby as the most important components

in the production of new products can be highlighted: resistant starch, pectins, guar gum, gum arabic, fructans, galactooligosaccharides, lactulose and other wide variety of oligosaccharides (Guo, 2009).

The disease prevention and health enhancement of active plant-derived ingredients or plant-based foods have widely been examined and many of them have demonstrated positive effect on human health (Carrillo et al., 2019; Dimou et al., 2019; Pawlowska et al., 2019; Santamarina et al., 2019; Kalt et al., 2020). There have been a several attempts to enrich different food with polyphenols such as fruit smoothie enriched with polyphenols and fibre by adding high concentration of fruit, snack bars with added fruit fiber and polyphenols, a drinking yoghurt and functional breads enriched with fruit polyphenols etc. Some of the developed functional foods enriched with fruit polyphenols are shown in **Table 1**.

The food industry has also faced this challenge by recovering of polyphenols from the by-products of plant food processing with possible potential applications in the food, pharma and cosmeceutical industries (Ibrahim et al., 2017). In **Figure 2** a schematic representation summarizing key technical development factors and potential applications of plant by-products valorisation is provided. Fruit processing involves numerous phases, including peeling, trimming, deseeding, cutting and pressing which results in high amounts of by-products rich in polyphenols.

Fruit pomace is one of the most common by-products in the fruit industry. Some fruits such as apple, grape and/ or berry pomace are known to possess high antioxidant activities. Today, these value-adding components are isolated and exploited as natural antioxidants for the formulation of functional foods or food ingredients (Ibrahim et al., 2017). However, future innovations in the field of functional foods enriched with polyphenols and other antioxidants should focus on exploring synergistic interactions between added bioactive substances and other food ingredients in selected products during processing and storage.

Table 1. Fruit-related functional foods (adopted from Sun-Waterhouse, 2011)

| Fruit material | Food format | Bioactives or mechanism of interest | |
|--|--|--|--|
| Blueberry | Blueberry fruit drinks | Not available | |
| Berries | Blended berry fruit juices | Flavonoids | |
| Fruit and vegetable juice powders | Enrichment for cereal products | Flavonoids, fibre and vitamins; Use as processing aids, colorants, flavorings. | |
| Chokeberry, elderberry, blackcurrant redcurrant, red grape, cherry, strawberry, raspberry plum | Antioxidant functional juices | Anthocyanins, flavonols, flavan-3-ols, phenolic acids | |
| Fruit powder (blueberry, cranberry, Concord grape, and raspberry) | Naturally colored breakfast cereals (extruded) in opaque bags | Soluble phenolics and anthocyanins (survive extrusion and retain some antioxidant activity) | |
| Acerola extracts | An isotonic soft drink containing anthocyanin extracts from acerola and from acai | The highest stability was correlated to high flavonoid content and absence of ascorbic acid. The degradation of anthocyanins occurred in the presence or absence of light. | |
| Citrus fruits, such as lemons or oranges | Fruit juice and skim milk mixtures | Antioxidant capacity of new fruit juice and skim milk | |

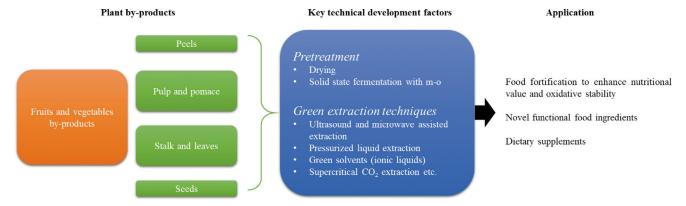


Fig. 2. Schematic representation summarizing key technical development factors and potential applications of plant by-products valorisation (adopted from Ben-Othman et al., 2020)

3. Conclusion

Health and nutrition are the most demanding and challenging field in this era and would continue to be in the future as well. During the last decades, the global dynamics in food production and consumption have changed significantly. While in the past the prevalent policy was mainly to increase food production, today the food industry is focused on the production in accordance with consumer demands as well as assuring the sustainability of the food sector. Finally, it can be concluded that there is an extensive need in food industries to introduce innovations associated with the implementation of emerging technologies, food waste recovery and production of new functional food products in order to survive market competition.

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Onion Solid Waste as a Potential Source of Functional Food Ingredients

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Abstract

Onion (Allium cepa L.) is one of the most basic and most consumed vegetables in human diet with a long tradition of use in food and medicine. It is grown and traded worldwide for culinary purpose because of its distinctive aroma and high nutritive value. Nowadays, onion production is increasing as well as the demand for processed onion, leading to the generation of a substantial amount of industrial waste. The waste biomass is mainly composed of outer scales, skin, roots, tops of the bulbs, and deteriorated bulbs. The use of onion waste as a potential source of functional food ingredients is one way of its valorization, highly in line with the current trends in the food industry to improve by waste reuse while responding to the increasing demand for functional food. Onion waste components exhibiting functional potential are: (i) flavonols – quercetin and quercetin glucosides, known for antioxidant activity; (ii) fructooligosaccharides, well-established prebiotics; (iii) cell-wall polysaccharides, as a part of dietary fibre, or specifically pectin as a known food additive and/or a potential source of prebiotic oligomers; and (iv) organosulfuric compounds. The use of onion waste in food production is still in the research stage, but shows promising results in the incorporation of powdered onion waste concentrates and extracts into various food matrices to obtain innovative products with improved antioxidant and prebiotic quality.

Keywords: fructooligosaccharides, functional food, onion solid waste, quercetin, valorization

1. Introduction

Industrial food production is steadily increasing with the growth of world's population as well as with changes in consumers' behaviour, seeking for "instant", "ready to eat" and "fast" food (Chandrasekaran, 2013). Food processing inevitably generates by-products of varying composition and structure, depending on the primary raw material and the technological process involved. In case of plant-origin food, agro-industrial residues usually comprise peels, shells, skins, pods, stems, straw, stones, pits, seeds, exhausted pulp, pomace and other non-edible and/or non-usable plant parts (Galanakis, 2012; Chandrasekaran, 2013). The management of such materials usually includes composting and animal feed; however, only a limited type of plant residue is suitable for these uses, while the others become waste Inadequate disposal of agro-industrial wastes leads to serious problems at environmental, economic and socialy-responsable levels (Chandrasekaran, 2013). Nowadays, a concept of valorization is intensively being introduced to the agro-industrial waste management, by which the residues are regarded as valuable secondary raw materials for the production of value-added products (Galanakis, 2012; Matharu et al., 2016). New ways of agro-industrial residues use are focused on the recovery of their interesting and rich chemical composition, namely of target micro- or macro- components with certain potential (Laufenberg et al., 2003; Galanakis, 2012; Otles et al., 2015). The advantage of waste materials as secondary sources of valuable components is promoted by their low initial value, abundance and availability, renewability and non-competitiveness to food.

In view of the fact that plant residues were originally a part of food raw materials, their possible reuse again in food production is possibly the most interesting valorisation option. For that matter, constituents with potential health-promoting properties are the most interesting target molecules for the recovery from agro-industrial wastes (Kumar et al., 2017). Their incorporation into food products falls into a specific segment of food industry – production of functional food. The market for functional food is continuously increasing and it is believed to reach 305.4 billion dollar value in 2020 (Bogue et al., 2017). Key drives for the accelerated development of the functional food market are the strong interest of consumers for the prevention of certain health problems, raising costs of health care, prolonged life expectancy and the trend of an ageing population towards a better quality of life (Bigliardi and Galati, 2013).

Many different waste materials are being studied (Mirabella et al., 2014) as potential sources of functional food ingredients. The aim of this paper is to provide a systematic review of the most important functional constituents of onion waste which enable its potential reuse in innovative functional food development, as well as to report on the current uses of this waste material for such a purpose.

2. Onion – importance of the crop, its production, and waste generation

Onion (*Allium cepa* L.), also known as common, bulb onion, or Egyptian onion, is one of the world's most important food crops. Its cultivation has been known for more than 4000 years, believed to originate from central

Asia, but nowadays spread all over the world owing to the high adaptability of the plant to different climates and water supply (Halnet, 1990; Griffiths et al., 2002; Lewande, 2012). Onion is a biennial plant, although mostly grown as an annual crop due to the finished formation of its most usable part - the bulb, by the end of the first year of growth (Slimestad et al., 2007). Morphologically, the bulb consists of cylindrically oriented leaf bases, called scales, attached to a short disc-like stem, called the basal plate. The scales accumulate water and products of photosynthesis which causes them to swell, thus forming the bulb. As the bulb ripens, the outer scales develop into a dry and impermeable skin, which helps in preventing drying-out (Teshika et al., 2019). Onion is considered to be among the oldest vegetables and traditionally has been used in folk medicine and for culinary proposes since ancient times. Today it is one of the most important crops and ranks third after potato and tomato as the most important food crop (Teshika et al., 2019). Onions are grown in a variety of forms, including shape, colour, size, dry matter content and pungency, to meet specific culinary and nutritive requirements and so have become an almost universal ingredient in food preparation worldwide. Onion production falls into three major product segments: (i) bulbs for fresh market, (ii) dehydrated onions for food processing, and (iii) green, salad onions for fresh consumption (Griffiths et al., 2002). World production of onion bulbs (dry onions) in 2018 was estimated at 96.7 million tonnes (FAO, 2020). The biggest producers are Asian countries, accounting for 67.5 % of world production, referring primarily to China and India. European countries contribute with 9.3 % to the world production, 66 % of which comes from EU, with leading Spain and The Netherlands (FAO, 2020). If refrigeration or controlled atmosphere storage is available, onions can be stored for up to 9 months, which, as a major advantage, adds up to their massive

production and prevalence all over the world (Griffiths et al., 2002). Still, long storage of onions in bulks can results in notable losses, up to 25-30 % (Lewande, 2012). Therefore, processing of onions ensures product stability while protecting its specific sensory quality. The international market is increasingly focused on dehydrated products such as flakes, rings, granules, powder, etc., and processed onion as frozen or canned, or onions in vinegar and in brine (Lewande, 2012). Processing of onions generates waste material mainly composed of onion skins, outer fleshy scales (usually 2), roots and tops of the bulbs, resulting from the mechanical peeling of the bulbs, as well as of deformed, damaged or rotten bulbs, not suitable for usage (Benítez et al., 2012; Sharma et al., 2016; El Mashad et al., 2019), as can be seen in Figure 1.

Approximately 38 % of the fresh weight of processed onion is not suitable for consumption and discarded as waste. In addition, approximately 10 % of harvested onions are worthlessly marketed, meaning that they do not meet the quality standards for marketing (El Mashad et al., 2019). In the available literature it is reported that annual generation of onion waste in the EU is as high as 500 000 tonnes, dominantly in countries with significant onion production, such as Spain, The Netherlands and United Kingdom (Waldron, 2001; Sharma et al., 2016). Onion waste disposal poses certain problems for conventional ways of vegetable waste disposal, namely: (i) onion solid waste cannot be used as fodder due to the specific aroma or possible toxicity; (ii) also its composting is limited due to the high susceptibility for the growth of phytopathogen Sclerotium cepivorum (white rot) and high content of sulfur-containing compounds; as well, (iii) the combustion of onion waste is rather expensive due to the high moisture content (Jaime et al., 2002; Benítez et al., 2012).



Fig. 1. Onion solid waste generation by discarding unusable parts of the bulb (Sharma et al., 2016)

Onion demand and production is steadily increasing; in the last 10 years, from 2008 to 2018, onion production has increased by about 30% (FAO, 2020). Due to the increasing production, combined with a relatively large amount of generated solid waste without adequate ways of its disposal, onion waste has become an attractive challenge for researchers trying to establish efficient reusing strategies for this biomass. These mainly focus on the production of value-added products, such as functional food in the food industry sector.

3. Constituents of onion waste with bioactive potential

The potential for valorization of onion waste as a source of functional ingredients lies within its interesting and rich chemical composition as well as its well-known health-promoting properties. Onion composition varies with cultivar, stage of maturation, environmental and agro-technical conditions, storage time and bulb section (Jaime et al., 2002; Benítez et al., 2012). Since onion waste is composed of different parts of the bulb in different proportions, and taking into account the above mentioned differences, onion waste chemical composition can notably vary and therefore it is necessary to determine the content of target compounds from case to case. However, onion waste has generally been identified as a source of flavour compounds, fibre, non-structural carbohydrates and polyphenols (Benítez et al., 2012; Sharma et al., 2016) which opens possibilities for its further employment as a source of functional food ingredients for enhancing antioxidant and prebiotic quality of novel products.

3.1 Polyphenols – flavonoids

Flavonoids are a group of polyphenolic compounds known for their strong antioxidant potential. Most important dietary sources of flavonoids are fruits and vegetables, with onion being one of the richest. In addition, flavonoids are dominant polyphenolic compounds found in onion, represented by 2 major subgroups: (i) flavonols, characteristic for yellow varieties, and (ii) anthocyanins, found in red/purple varieties (Griffiths et al., 2002). Representation of flavonol groups, as well as individual compounds within a specific group, is highly dependent on onion variety, as well as on bulb portion and onion growing conditions (Benítez et al., 2011). Main flavonols in onion are quercetin and its glucoside derivatives, with dominant quercetin-4'-glucoside and quercetin-3,4'-diglucoside, which represent up to 80 % of total flavonoids in onion (Benítez et al., 2012). The monoglucoside:diglucoside ratio varies among bulb portions in favour of monoglucoside content towards the outer scales (Benítez et al., 2011). It is believed that quercetin diglucoside is formed during storage and its higher contents are expected in inner scales (Slimestad et al., 2007). Generally, the content of quercetin glucosides decreases towards the outer scales, while the content of aglycone quercetin increases in the same direction, reaching its maximum in the brown onion skin. The increase in aglycone content results from deglucosilation of quercetin glucosides, probably in the presence of sunlight (Higashio et al., 2005). Further oxidation of quercetin by peroxidase results in formation of protocatehuic acid (Takahama and Hirota, 2000) which exhibits antifungal properties. This suggests an enzymatic formation of quercetin-derived defence substances against infection in the dry onion skin (Slimestad et al., 2007). Quercetin, its glucoside derivatives, protocatehuic acid as well as their oxidation products are responsible for yellow and brown shades of onion skins. Slimestad et al. (2007) report that outer dry skins contain 2.5-6.5 % of flavonols by weight, with 67-86 % being quercetin aglycone while quercetin-4'-glucoside only to a smaller extent. On the other hand, a study by Benítez et al. (2011) found guercetin-4'-glucoside dominant over quercetin aglycone in the brown onion skin. As well, in the mentioned study, the outer scales contained the highest content of flavonols, dominantly quercetin-4'-glucoside and quercetin-3,4'-diglucoside. Other quercetin derivatives, besides 4'-glucoside and 3,4'-diglucoside, also can be found in some onion varieties, namely quercetin-3-glucoside (isoquercetin) and quercetin-7,4'-diglucoside, but to a minor extent. Of other flavonols, derivatives of kaempferol, isorhamnetin and possibly myricetin, as well as their aglycones, can be found at low contents (Griffiths et al., 2002; Slimestad et al., 2007). In addition to the mentioned flavonols, dihydroflavonols, such as taxifolin (dihydroquercetin) and its glucosides, also contribute to the polyphenolic profile of onion, while their concentration highly depends on onion cultivar (Slimestad et al., 2007).

Quercetin and its derivatives expose a number of biological activities, including antioxidant, anti-inflammatory, anticancer properties and prevention of cardiovascular diseases (Griffiths et al., 2002; Wang et al., 2016). Therefore, many extraction approaches are being investigated for their efficiency in quercetin recovery (Ren et al., 2020). Moreover, postharvest storage manipulations and processing, such as temperature control, UV irradiation and high pressure have shown a potential in increasing, or at least preserving, quercetin levels in onions (Ren et al., 2020). These measures are being investigated to enhance the bioactive quality of onion as a raw material, and consequently of its waste.

Besides flavonols, a number of anthocyanins has been detected in some onion varieties (red onions), contributing to their distinctive colour in shades of red and purple. These comprise cyanidin-3-glucoside, cyanidin-3-(3"-glucosylglucoside) (also known as cyanidin 3-laminariobioside), cyanidin 3-(6"-malonylglucoside), and cyanidin 3-(3"-glucosyl-6"-malonylglucoside) as the main anthocyanins in most of the cultivars (Slimestad et al., 2007). Most of the anthocyanins reported to occur in various cultivars of red onion are cyanidin derivatives, although minor amounts of peonidin derivatives have

been identified, as well as of delphinidin and petunidin derivatives in some cultivars (Slimestad et al., 2007).

3.2. Other constituents

3.2.1 Organosulfuric compounds

Organosulfuric compounds in onion are carriers of its specific aroma and flavour. They derive from non-volatile precursor sulphuric compounds - S-alk(en)yl-L-cystein sulfoxides (ACSO), commonly known as aliin (Rose et al., 2005). Most relevant ACSO in onion are (+)-S-methyl-L-cysteine sulfoxide (metiin), (+)-S-propyl-L-cysteine sulfoxide (propiin) and trans-(+)-S-(propen-1-yl)-Lcysteine sulfoxide (izoaliin), the latter being most dominant, representing up to 80 % of ACSO in onion, and thus most responsible for onion characteristic flavour (Griffiths et al., 2002; Benítez et al., 2012). Common to all Allium species is the enzyme allimase that catalyses decomposition of ACSO into pyruvate, ammonia and sulfenic acids. In intact onion tissues, the enzyme is located in the vacuoles, while the ACSO in the cytoplasm. Upon cell disruption, for example by cutting, the enzyme and ACSO come in contact generating sulfenic acids which further spontaneously react among themselves and with other compounds to produce various other volatile organosulfuric compounds, such as thiosulfinates (Griffiths et al., 2002; Rose et al., 2005). Aroma of onion is therefore very complex and changes with time as the volatiles generate and decompose. Besides their importance as aroma compounds, organosulfuric compounds of onion exhibit many health promoting effects as summarized in papers of Griffiths et al. (2002), Rose et al. (2005) and Sharma et al. (2016). In addition, low molecular sulphuric compounds, such as thiols, can act as polyphenol oxidase inhibitors, which opens up the possibility of being used as preservatives and anti-browning agents in the food industry (Sharma et al., 2016).

3.2.2 Carbohydrates

Carbohydrates found in onion can be divided into two major groups: (i) non-structural carbohydrates, and (ii) cell wall polysaccharides. Non-structural carbohydrates in onion comprise glucose, fructose, sucrose and fructooligosaccharides (FOS) which constitute a large portion of onion's dry matter, up to 65 % (Griffiths et al., 2002; Sharma et al., 2016). While glucose, fructose and sucrose contribute to nutritive and caloric value of onions, FOS are considered as non-nutritive functional components by being a part of dietary fibre and furthermore well-known and recognized prebiotics. FOS (polyfructosylsucrose) are a sub-group of fructans - complex polymers made of fructose monomers linked with sucrose molecule, which can be found in a wide range of structural diversity and degree of polymerisation (Benkeblia, 2013). In comparison to other fructans, FOS are generally characterized by lower degree of polymerisation (DP). Although, oligomeric and polymeric forms are not clearly distinguished until today so the literature provide different data on the classification of FOS according to degree of polymerisation (Benkeblia, 2013). FOS content in onion, determined as a sum of kestose (GF2 – G-glucose; F-fructose; DP3; β-(1,2) bond), nystose (GF3; DP4) and fructofuranosyl nystose (GF4; DP5), exhibited 30-60 % of total fructans, depending on the onion variety, with kestose being dominant among the three (Jaime et al., 2001). This implies that onion FOS are mainly inulin type oligomeric (DP3-DP5) fructans (Jaime et al., 2001; Benítez et al., 2012). Further studies showed as well the presence of other FOS forms, namely inulin neoseries (Benkeblia, 2013). The FOS content in onion is generally relatively decreasing towards the outer scales (Benítez et al., 2011). Tops and bottoms of the bulbs, as well as dry brown skins show low contents of FOS (Jaime et al., 2001; Benítez et al., 2011). However, since onion solid waste usually comprises a part of fleshy scales, it can be regarded as a potential novel source of FOS. Dietary importance of FOS results from their significant prebiotic effect (Roberfroid, 2007).

Cell wall polysaccharides of onion waste, determined by monomeric sugars analysis upon complete hydrolysis of the material, reveal the dominance of uronic acids (galacturonic acid) and glucose, exhibiting 21 % and 27 % dry matter basis, respectively (Vojvodić et al., 2016), which is correlated to the presence of pectic substances and cellulose (Ng et al., 1998; Jaime et al., 2002; Vojvodić et al., 2016). Dietary fibre analysis revealed predominance of insoluble fibre in onion solid waste, especially in the dry brown skin, tops and bottoms of the bulb (Jaime et al., 2002), but as well in the waste containing fleshy scales, exhibiting approximately 59 % dry matter basis (Vojvodić et al., 2016). High insoluble fibre content is related to the increased content of lignin in the outer parts of the bulb (Jaime et al., 2002). Regarding the high content of galacturonic acid in onion waste, the low soluble fibre content is possibly unexpected. However, these results indicate on structural characteristics of onion waste pectin, making it insoluble for the applied fibre analysis (Vojvodić et al., 2016). Indeed, onion waste pectin is largely chelator-soluble and therefore cannot be readily solubilised using water or diluted acids (Babbar et al., 2015). The application of onion waste pectin and fibre has not been sufficiently supported by the available data so far. However, limited studies suggest the use of solid onion waste (paste and bagasse) in the form of dietary fibre concentrates to enrich foods with dietary fibre and, in addition, to achieve possible additional functionality with regard to hypoglycaemic effects (Benítez et al., 2017). Lecain et al. (1999) suggest onion waste as a potential novel source of cheap pectin. Innovative ways of onion waste pectin valorization include the production of potentially prebiotic pectic oligosaccharides (Babbar et al., 2016; Baldasaree et al., 2018). Valorisation of the second most dominant cell wall polysaccharide – cellulose, could be directed towards its fermentation and bioconversion potential (Choi et al., 2015).

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4. Perspectives for onion waste uses

Due to its rich chemical composition, onion waste offers a number of possibilities for reuse. One of the most perspective uses is within food industry as a source of functional food ingredients. For this purpose, onion waste can be used in powdered form, but also in a form of various extracts rich in target compounds or microencapsulated extracts designed to preserve specific quality of the extract (Table 1). As can be seen from Table 1, potential food uses focus on the enrichment of conventional food products with dietary fibre and polyphenols, while food matrices can be very different, from bakery and cereal-based products and meat products to confectioneries. In addition, onion waste as well could be used as flavouring and colouring agent for some food products.

With respect to the carbohydrate content of onion waste, specifically non-structural carbohydrates, onion waste has shown potential as a raw material for vinegar production (Hiriouchi et al., 2000).

Prior to implementation into food products, onion waste needs to be adequately stabilized in microbiological terms, as well as to protect the material from degradative processes that could decrease material's quality. In addition, stabilization is mandatory to ensure waste material's preservation over the time gap between waste generation and its further processing. As reported by Sharma et al. (2016) stabilizing techniques commonly used are sterilization, pasteurization, and freezing, presenting crucial steps for valorization of onion waste as a safe food ingredient, since the waste involves risk for microbiological growth and decomposition of value-added compounds. Since these methods can affect quantitative and qualitative profile of bioactive constituents, the choice should be made with respect to the aim of waste usage.

Besides stabilization, further steps of onion waste preparation also must be carefully planned. This is particularly important in the preparation of onion waste extracts where it is desired to have efficient extraction of target compounds while reducing negative environmental impact and hazard. From the life-cycle assessment approach, conventional extraction strategies for the recovery of quercetin and FOS from onion waste resulted in significant environmental burdens and therefore the development of alternative extraction techniques should be encouraged (Santiago et al., 2019).

Table 1. Summary table of potential food uses of different forms of onion waste

| Form of onion waste | Application matrix /final product | Functional/technological benefits | Reference |
|---|--|---|-----------------------------------|
| onion peel powder | fermented onion peel | functional food based on fermented onion peel rich in flavonoids and prebiotics | Kimoto-Nira et al., 2019 |
| onion peel powder | wheat flour extrudates | extrudates with higher antioxidant activity, smaller diameter and pore size | Tonyali and Sensoy, 2017 |
| onion peel extract | emulsion pork sausage | product with improved quality characteristics and higher shelf life stability | Lee et al., 2015 |
| onion peel powder | bread | bread with enhanced antioxidative properties | Gawlik-Dziki et al., 2013 |
| onion peel extract | film from Artemisia sphaerocephala Krasch gum (ASKG) | onion peel extract-containing ASKG films express potential as intelligent packaging materials and gas-sensing labels with pH indicator | Liang et al., 2018 |
| onion peel powder | Hanwoo Tteokgalbi | natural preservative and antioxidant properties of product with great water retention | Chung et al., 2018 |
| onion peel powder | gluten-free bread | consumption of enriched bread increased antioxidant activity of consumers' blood | Bedrnicek et al., 2020 |
| onion peel extract | wheat bread | bread with higher antioxidant activity and phenolic content | Piechowiak et al., 2020 |
| onion peel extract | bean paste | paste with higher antioxidant activity and phenolic content | Sęczyk et al., 2015 |
| onion peel extract | hard candy and glazing jelly | natural food colorant | Om-Hashem et al., 2016 |
| onion skin powder | wheat pasta | enhancement of nutritional and antioxidant quality while maintaining sensory acceptability; authors recommend onion skin powder as a possible additive in pasta making | Michalak-Majewska et al., 2020 |
| microencapsulated onion peel extract | cake | cake of higher specific volume, enhanced moisture absorption and texture as well as phenolic content | Elsebaie and Essa (2018) |

5. Conclusions

Increasing world production and processing of onion results in a substantial amount of generated waste. Conventional ways of vegetable waste disposal seem to be inadequate in the case of onion so other options are intensively being explored. Due to its abundance in components with bioactive potential, the reuse of onion waste for the production of enriched food products possibly presents the best valorization strategy. In this way, the low-value material is reduced as waste and simultaneously upgraded to a valuable secondary raw material.. By its reuse in food, one part of the originally produced biomass intended for food, could be brought back to the food chain while additionally exhibiting added value through the health-promoting effects of its bioactive constituents. The most important potential ingredients from onion waste to consider are quercetin and its glucosides, and dietary fibre – as a whole or separated fractions of FOS and pectin. However, onion waste reuse in practical large-scale terms is not easy to achieve. Many researchers worldwide try to find solutions for efficient stabilization of the material, efficient and environmentally acceptable extraction techniques of target compounds, preservation of extracts' functional characteristics through various delivery forms, optimal ways of implementation into food products and finally, ways to prove health claims. To the present day, many studies have prepared the ground for presenting onion waste as a material to be explored. Indeed, onion waste has shown a great potential to be incorporated into different food matrices in various forms: as a powdered concentrate, an extract, or most innovatively as microencapsulates, in order to obtain innovative food products with enhanced antioxidant and prebiotic quality. There are still many unexplored possibilities for food implementation and research to discover its full potential.

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Macroalgae in the Food Industry – Opportunities and Challenges

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Abstract

The industry recognized macroalgae not only as a very important source of bioactive compounds but also as 'superfoods' due to their nutritional value. They can be used as raw food acting as an alternative source of vegetable proteins, fibres, vitamins and minerals but also they can be commercialized in many forms and incorporated in other food products. The aim of this review is to emphasize the importance of macroalgae application in the food industry. They are already used for different purposes, from food products to medicine. Among mentioned compounds, the special emphasis was given to macroalgal polysaccharides that are present in many products consumed on daily basis. Their importance in texture stability of the food products, as well as their functional properties are evaluated.

Keywords: macroalgae, food industry, high added-value products, macroalgal polysaccharides

1. Introduction

Oceans are considered as the "lungs of the Earth" due to the presence of the algae and Cyanobacteria which provide up to 80 % of the atmospheric oxygen. Among organisms that provide oxygen, macroalgae are very important for human health as they are important for the Earth. Generally, they are divided into three groups based on the colour of the thallus, as follows: Chlorophyta (green algae), Rhodophyta (red algae), and Ochrophyta Phaeophyceae (brown algae) (Barsanti and Gualtieri, 2014). Since they are sessile organisms, they have to adapt to extreme and hostile environmental conditions such as temperature fluctuations, salinity, UV radiation and various pollutants. During the adaptation, they are producing a wide range of secondary metabolites, such as pigments, phenolic compounds, sterols and other bioactive agents. Besides, they represent a rich source of proteins, saturated/unsaturated fatty acids and polysaccharides which makes them important at the commercial level. A few years ago, interest in the exploitation of macro-algae increased and they are already used for various purposes (Pereira, 2016). Although there is still much to investigate about macroalgae and their compounds, it is known that several of the metabolites they synthesize have great potential to be used in pharmaceutical, cosmetics and the food industry. As their interest, cultivation and applications increase, their value in the market rises too. It is estimated that the value of the macroalgae market worldwide in 2024 will exceed twice the achieved in 2017 (Statista 2019). Recently, Europe has been highlighted as one of the most innovative regions regarding the use of macroalgae as a food ingredient with the exponential increment of new products on the European market by 147 % between 2011 and 2015 (Afonso et al., 2019).

This overview provides an insight into the importance of macroalgae as part of the human diet as well as into the components of food. The studies reported in this review showed that the addition of macroalgae, in powder or as extracts, can improve the nutritional and textural properties of food products. Macroalgal polysaccharides are the most important compounds incorporated into products so their role in the food industry is specially emphasized in this review.

2. Macroalgae as part of everyday diet

Since ancient times until the beginning of the 19th century, people in the East regarded seaweed as a food of great delicacy, while the Greeks and Romans thought differently. After 1800, the boom in the macroalgae industry began, especially in the 20th century (Zeneveld, 1959). It is recognised that edible macroalgae, which are categorised in more than 600 species, have a great nutritional value which can be influenced by geographical location, growth stage, season, etc. (Kim, 2011). Even though they are known as low caloric food, rich in vitamins and minerals, they need to be evaluated before being used as supplements (Leandro et al., 2020).

Although the consumption of macroalgae is not as widespread in Europe as in Asia, they have attracted attention because their bioactive compounds have earned them a reputation as the new 'superfoods' (Cofrades et al., 2017). Brown algae are the most consumed (66.5 %), followed by red (33 %) and green (5 %) (Lorenzo et al., 2017). Brown macroalgal species considered safe for human consumption are Fucus vesiculosus, Fucus serratus, Himanthalia elongata, Undaria pinnatifida, Ascophyllum nodosum, Laminaria

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digitata, Laminaria saccharina, Laminaria japonica and Alaria esculenta (CEVA, 2014). Studies have shown that the macroalgae contain a higher content of fibres (Grateloupia filicina, Chondrus crispus, Ulva lactuca) than the plants (Holdt and Kraan, 2001; Yuan et al., 2009) which can contribute to general human health. It is known that dietary fibres stimulate the growth of beneficial gut bacteria, reduce the risk of diabetes, obesity and hypercholesterolaemia due to their capacity for absorbing organic compounds such as glucose and cholesterol (Bocanegra et al., 2009). Macroalgae can be used as alternatives to vegetable sources of proteins (red algae: Pyropia tenera, Grateloupia filicina), as well as for the formation of protein balanced diets with low-costs due to their high content present in macroalgae, similar to those found in legumes such as peas and beans (Rodrigues et al., 2015; Yuan et al., 2009). Although a small amount of total lipids is present in macroalgae, their qualitative profile is of particular interest from the nutritional point of view due to the presence of ω -3 and ω -6 polyunsaturated fatty acids (PUFAs) which exhibit activities such as cardiovascular and coronary protection, reduction of arterioscelorsis, etc. (Khan et al., 2007; Plaza et al., 2008). It was found that the ashes of edible macroalgae contained higher contents of macro- and microelements than those reported for edible plants where the amounts of Na, K, Ca and Mg were in range of 8.083-17.875 mg/100 g and of Fe, Zn, Mn and Cu were between 5.1-15.2 mg/100 g. According to this fact, brown and red macroalgae can be used as food supplements to reach the recommended daily intake of minerals and trace elements. For instance, the consumption of 10 g of *Ulva* lactuca provides 70 % of the daily Mg and over a half of Fe requirements (Yuan et al., 2009; Ruperez, 2002). Himanthalia elongata is the most exploited due to its mineral composition and it is a good candidate for the salt replacer as it reduces high salt consumption and health-related complications (Circuncisao et al., 2018).

3. Macroalgae as functional health-promoting ingredients of food products

Because of the bioactivities mentioned for the fatty acids ω -3 and ω -6, they were used in infant foods and formulas, leading to an improvement in infants' cognitive performance and visual acuity (Jensen et al., 2005). Compounds derived from macroalgae can behave as functional ingredients of various food products which has been demonstrated in the production of fortified healthier snacks with algal oil. Healthy adults that consumed these snacks showed higher levels of docosahexaenoic acid (DHA; 22:6) in plasma (Arterburn et al., 2007). Algal oil was also added to ice creams to increase the ω-3 fatty acid content, but the results of the sensory evaluation showed that the fishy taste was stronger than ice cream taste (Chee et al., 2007). When the food is enriched with ω-3 fatty acids, its oxidative stability decreases and it can lead to the development of undesirable flavours and

reduction of the shelf-life (Jacobsen et al., 2000). However, O'Sullivan et al. (2014, 2016) observed that the incorporation of *F. vesiculosus* ethanol extract improved the shelf-life of milk and yogurt which indicated the presence of the compounds with the ability to reduce lipid oxidation that is known as antioxidants.

Antioxidants present in macroalgae can serve as alternative replacements of synthetic antioxidants such as butylated hyroxyltoluene (BHT) and butylated hydroxianisole (BHA) without any side effects on human health (Roohinejad et al., 2017). Also, they have ability of the extension of shelf-life during storage by inhibition or postponement of lipid oxidation (Cornish and Garbary, 2010). The most known antioxidants of macroalgal origin are phlorotannins mostly present in brown macroalgae. Fucus vesiculosus has been found as the best source of phlorotannins and antioxidant compounds and because of that it is applied as functional ingredient in different food matrices to prevent spoilage and oxidative deterioration (Afonso et al., 2019). It was shown that the addition of F. vesiculosus commercial extract can be exploited as a food antioxidant agent due to the inhibition of formation of primary and secondary oxidation products, as well as the maintenance of the colour of pork liver pâté (Agregan et al., 2018). Carotenoids are as well added into the food products as antioxidants, but they have one more important role. They can serve as natural color enhancers and replace synthetically obtained which are known to cause liver and renal toxicity, as well as promote carcinogenesis. β-Carotene is the most applicable macroalgal derived pigment which has multiple activities, from color enhancer to anticancer agent that is absorbed 10 times more easily by the body than the synthetic one (Christaki et al., 2012). All the compounds discussed above with their properties when they are added into the food products are summarized in Table 1.

Table 1. Macroalgal compounds of interest in the food industry and their properties in the food products

| Macroalgal compounds | Properties in the food products | |
|---|---|--|
| Carotenoids Phlorotannins | Replacers of synthetically obtained antioxidants, Extension of shelf-life | |
| Carotenoids | Colour enhancer | |
| Phlorotannins | Prevention of spoilage and oxidative deterioration | |
| Proteins | Nutritional value, Higher content than in legumes | |
| Minerals | Higher content than in plants, Salt replacers, Food supplements for reaching the recommended daily intake | |
| ω-3 and ω-6 Polyunsaturated fatty acids | Cardiovascular and coronary protection, Improvement of infant cognitive performances | |

Apart from the functional properties, macroalgal compounds can influence product texture and consistency. When 4 % of *Fucus vesiculosus* powder was added during bread preparation, it was observed that dough had increased viscosity and consistency, while porosity decreased that led to higher density and crumb firmness of the bread (Arufe et al., 2018). Also, better quality was achieved for pasta when two algal powders, Sargassum marginatum and Undaria pinnatifida, were added. Not only the fortified pasta exhibited better cooking and sensory characteristics, but also its nutritional quality was enhanced due to the higher protein content (Prabhasankar et al., 2009a; Prabhasankar et al., 2009b). When algal powders of Undaria pinnatifida and Laminaria japonica were added in the production of cottage cheese, its consistency and firmness were better (Lalić and Berković, 2005). The food products containing macroalgal derived compounds are listed in Table 2.

Table 2. Food products containing valuable macroalgal derived compounds

| Food product | Macroalgal derived compounds or extracts | References | |
|--------------------|---|---|--|
| Pork liver pâté | Fucus vesiculosus commercial extract | Agregan et al., 2018 | |
| Pork patties | F. vesiculosus ethanol extract | Agregan et al., 2019 | |
| Snack bars | Docosahexaenoic acid (DHA) | Arterburn et al., 2007 | |
| Bread | Powder of Fucus vesiculosus | Arufe et al., 2018 | |
| Ice-cream | ω-3 Fatty acids | Chee et al., 2007 | |
| Meat products | Powder of Himanthalia elongata | Cofrades et al., 2017 | |
| Infant food | Docosahexaenoic acid (DHA) | Jensen et al., 2005 | |
| Cottage cheese | Powder of <i>Undaria</i> pinnatifida and Laminaria japonica | Lalić and Berković, 2005 | |
| Milk Yogurt | F. vesiculosus ethanol extract | O'Sullivan et al., 2014; 2016 | |
| Pasta | Powder of Sargassum marginatum and Undaria pinnatifida | Prabhasankar et al., 2009a; Prabhasankar et al., 2009b | |

Macroalgae can be commercialized and/or consumed in many forms; fresh, dried, as powder or incorporated in the food products that are called high added-value products (Leandro et al., 2020). They are already commercialized as healthier and natural substitutes of pasta (*Himanthalia elongata* as spaghetti) or bacon (*Palmaria palmata* as sea bacon). The red algae of genus *Porphyra* are used for the preparation of sushi rolls and crispy snacks, while wraps are made of *Undaria pinnatifida* (wakame) and *H. elongata*. The macroalgal products available on the market are labeled as "fat-free", "gluten-free", "mineral rich", "low carbohydrates" and "low calories". Seamore

company is one of the most popular manufacturer of macroalgae products including wraps, bacon, pasta and bread (Seamore, 2020).

Challenges that need to be overcome when the algae are used as food ingredients go from the amount of needed biomass to sustain the market development, to the consistent research of their physiochemical characteristics and the impacts which they have on the food products when used as ingredients. The incorporation of macroalgae into the food usually leads to a deterioration of the sensory properties of the products. This can be overcome if more attention is paid to research into alternative approaches, such as the use of algae extracts consisting of whole algae or the encapsulation of algae or their extracts and the use of food flavourings to camouflage the undesirable flavourings. There is also a lack of information on the bioavailability of nutrients and/or phytochemicals, which is the decisive factor when macroalgae are presented in terms of their nutritional value. This will require more attention in future studies, as well as the potential functionality of macroalgae enriched products (Afonso et al., 2019).

4. Macroalgal polysaccharides as hidden food components

Polysaccharides isolated from macroalgae are the most implemented macroalgal derived compounds in food products. They are valuable additives such as stabilisers, thickening agents, texture modifiers and binders of ingredients thanks to their functions including water-binding capacity, gelation and formation of emulsions and foams. They have the ability to control starch retrogradation, replace fat, enhance flavour and improve fibre content when added in the food products (Menon, 2012). Macroalgal polysaccharides which are used in various food products are summarized in **Table 3**.

Carragenan is one of the main additives used in the food industry, from various dairy products (ice creams, milkshakes, yoghurts) to meat products (hams) as thickening and stabilizing agent or emulsifier (Pereira and van de Velde, 2011; Armisen, 1995). The molecular weight of the carragenan used in food products is regulated by Food and Drug Administration (FDA) where the minimum value of 100 kDa was established but the usual range is 400-600 kDa. There are three types of carragenan which can be used in food preparations differing in sulfation degree where kappa (κ) carragenan has one sulfate group per disaccharide, iota (1) has two sulfates, while lambda (λ) has three sulfate groups. The used carragenan depend on the desired properties, 1- and κ -carragenans have gelling properties, while λ -carragenan is a thickener/viscosifier (Pereira et al., 2013).

Agar is frequently used as a thickener in food products, as well as a vegetarian substitute for gelatine. The difference between agar and carrageenan is that carrageenan gels by Vol. 15(3) 2020 — 17

Table 3. Polysaccharides utilized in food products and its properties

| Compound | Macroalgae species | Application | Properties | Food additive code | References |
|------------|--|---|--|---|---|
| | | Oc | hrophyta, Phaeophycae | ea | |
| Alginate | Laminaria hyperborea, Macrocystis pyrifera, Laminaria digitate, Ascophyllum nodosum and Laminaria japonica | ice-cream, beer and soft drinks, meat products | emulsifier, stabiliser, foaming agent, thickener/ viscosifier, binder and filler | sodium alginate E-401, potassium alginate E- 402, ammonium alginate E-403, calcium alginate E-404, propylene glycol alginate E-405 | Alba and Kontogiorgos, 2019; Sartal et al., 2012; Cofrades et al., 2012 |
| | | | Rhodophyta | | |
| Carragenan | Kappaphycus alvarezii and Eucheuma denticulatum | dairy products: ice creams, yoghurt, cheese and milk, low-fat meat products, clarifying beer and fruit juices | gelling, stabilising and thickening agent | E-407 | Sartal et al., 2012 |
| Agar | Gracilaria and Gelidium species | pie filling, ice-creams, dairy products, wines, low fat products, icings and bakery glazes | thickener, stabiliser, clarification of wines, binder, crystallisa-tion prevention | E-406 | Sartal et al., 2012 |

both ionic and hydrogen bonding, while agar gels only by hydrogen bonding (Kilinc et al., 2013; Pereira 2016).

Alginate is, next to cellulose, the most abundant biopolymer in the world. It acts as thickener, emulsifier and it needs no heat to gel. The spherification process, a technique used in molecular cooking, includes the addition of agar in combination with calcium lactate of calcium chloride (Leandro et al., 2020). Alginates are responsible for the interaction with cholesterol and facilitate its excretion (Cofrades et al. 2012).

Apart from commercially available macroalgae polysaccharides, scientists studied the addition of polysaccharides on the quality of foods such as bread, where Guarda et al (2004) added carragenan and sodium alginate to prevent dehydration of fresh bread during storage. When the powder of Undaria pinnatifida was added to meat products such as patties, less thawing and cooking losses and softer texture were observed when compared to the samples without the addition of macroalgae. It was explained by the microstructural changes caused by the formation of alginate chains (Lopez-Lopez et al., 2011). The firmer and chewier structure was accomplished due to better water and fat-binding properties of macroalgae that can be explained by the presence of polysaccharides (Cofrades et al., 2008; Cofrades et al., 2012). With the addition of Laminaria digitata extract (0.5 % w/w) containing polysaccharides to pork patties, the quality of the products in terms of oxidative stability increased by the reduction of lipid oxidation in the cooked samples, but

the products were not very well accepted in the sensory analysis (Moroney et al., 2013). Similar observations occurred when laminaran and fucoidan from *L. digitata* were added to pork homogenates (Moroney et al., 2015).

Conclusions

Macroalgae as a source of bioactive compounds are widely applied in functional products so their positive influence on human health is inevitable. Antioxidants and pigments are important macroalgae constituents in the industrial range, but even more important are polysaccharides such as alginate, agar and carrageenan, which are already used in several food industries. With the exception of their functional properties, macroalgal polysaccharides have a significant influence on the texture of food products acting as emulsifiers, thickening and gelling agents. Most people are not aware that they consume macroalgae but many products, such as meat and dairy products, we consume on daily basis contain macroalgae derived compounds or their extracts. Also, macroalgae can be eaten as whole foods including more than 600 species known to have nutritional value higher than some plants and vegetables.

This review provided information that may help in the development of novel food products, but further research on the nutritional and functional value of macroalgae is still needed.

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Application of 3D Food Printing in Food Industry Development

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Summary

Food industry is craving to develop new technologies and processes which will satisfy customers' ambiguous wishes but also specific dietary needs. In 21st century, food allergies or food intolerances are affecting population from nurseries up to elderly stages while causing people, not only health issues but also making their nutrition plans and grocery shopping challenging and expensive. Three dimensional (3DP) food printing is a potential solution to advance current food processing techniques, but also to integrate 3DP and digital gastronomy technique to customize food products. If applied in domestic cooking or catering services, 3DP can provide an engineering solution for customized food design and personalized nutrition control, but also has the potential to redefine food supply chains. Still, people might be doubtful to eat 3D printed food due to their perception that it does not taste as good as traditional food and it appeals almost over-processed and artificial. This paper gives the short overview on 3DP techniques, devices and ingredients used for 3DP, as well as the possible future application in food manufacturing.

Keywords: 3D food printing, additive technology, food manufacturing, ingredients for 3DP

Introduction

People are now following trends more than ever, so even in food consumption there are leading trends of coming back to organic and wholesome food, but also trying new techniques and food processing techniques. So where does 3D printing (3DP) belong in food processing? By definition additive manufacturing is a new technology focused on the computer navigated layer-by-layer deposition of materials to build up 3D objects (Hao, 2010). All these requirements make food more and more expensive and exclusive. However, an important advantage of 3DP includes minimal manual labor and the capacity to manufacture extremely complex shapes, making it appropriate for a wide range of applications, especially in a fast-increasing consumer goods industry such as the food industry (Drury, 2003; Galantucci, 2009; Godoi, 2016; Li, 2015; Tumbleston, 2015; Khalil, 2007; Sun, 2015; Zoran, 2011).

Currently food manufacturing is oriented to serial production which is not only convenient but also considerably cheaper per product unit compared to manually manufactured products. 3D food printing revolution can also radically change the way we think about food manufacturing and preparation. This process can eliminate grocery shopping for ingredients to preparing the ingredients and cooking, and lead to an instant readymade meal (Tran, 2011). But like any other novelty, it takes time to be accepted by wider consumers, but it can also find a niche for people with specific dietary needs.

3D food printing - 3DP

Up to now there are several food printing techniques (Table 1.), such as selective laser sintering/hot air

sintering, hot-melt extrusion/room temperature extrusion, binder jetting, and inkjet printing, but the extrusion-based 3D food printing is the most commonly implemented method (Sun, 2015). This is a digitally controlled, robot-assisted manufacturing process that allows complex 3D food products (made of liquid/semi-solid/solid material) to be built layer by layer (Huang, 2013).

a) Hot-melt extrusion – FDM

This process originates from fused deposition modelling (FDM), and describes the extrusion process of melted semi-solid thermoplastic material from stationary FDM head and deposited onto a substrate. In food processing it is mostly used for 3D chocolate products which are slightly heated above its melting point and solidified almost immediately after extrusion (Hao, 2010). FDM food printer includes compact size and low cost of maintenance, but cons such as long production time and delays caused by temperature fluctuation.

b) Sintering technology

Also known as powder bed binder jetting, each powder layer is distributed evenly across the fabrication plat- form, and liquid binder sprays to bind two consecutive layers of powder (Sachs, 1990). The powder material is usually stabilized by spraying water mist to minimize the disturbance caused by a binder dispensing. Sugars and starch mixtures can be used as a powder material and binders could be liquids mostly based on high sugar content, which also presents cons of this technique as well as high machine costs, but this technology offers faster fabrication and the possibility to build complex structures.

Table 1. Comparison of 3DP technologies in food printing (Sun, 2015)

| | Hot-melt extrusion | Sintering technology | Inkjet powder printing | Inkjet printing |
|---------------------|---|---|--|---|
| Materials | Food polymers such as chocolate | Low melting powder such as sugar, NesQuik, or fat | Powder such as sugars, starch, corn flour, flavours, and liquid binder | Low viscosity materials such as paste or puree |
| Viscosity | $10^3 \sim 10^5 \text{ cP}$ | Not applicable | 1~10 cP (binder) | 5×10 ² ~5×10 ³ cP |
| Platform | Motorized stage Heating unit Extrusion device | Motorized stage Sintering source (laser or hot air) Powder bed | Motorized stage Powder bed Inkjet printhead for binder printing | Motorized stage Inkjet printhead Thermal control unit |
| Printing Resolution | Nozzle diameter: 0.5~1.5 mm | powder size:100 μm | nozzle diameter ≤ 50 μm Powder particle ≤100 μm | nozzle diameter ≤ 50 μm |
| Fabricated Products | Customized chocolates | Food-grade art objects, toffee shapes | Sugar cube in full color | Customized cookies, Bench-top food paste shaping |
| Pros | Cost effective Fast fabrication | Better printing quality Complex design | More material choices Better printing quality Full colour potential Complex design | Better printing quality |
| Cons | Low printing quality | Expensive platform High power consumption Limited materials | Slow fabrication Expensive platform | Slow fabrication Expensive printhead Expensive platform Limited materials |
| Machine & Company | Choc Creator by Choc Edge | Food Jetting Printer by TNO | Chefjet by 3D Systems | Foodjet by De Grood Innovations |

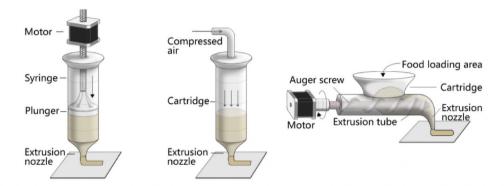
c) Inkjet printing

Inkjet food printing releases multi-material streams/droplets from a syringe-type multi-channel print head in a drop-on-demand way under gravity and creates 3D edible food products such as cookies, cakes or pastries. This process can be used to drop on-demand materials onto pizza bases, biscuits, and cupcakes and dry them by solvent evaporation.

d) Extrusion-based 3D printing (E3DP)

E3DP is an additive manufacturing technique, where the material is pushed in a melted, slurry or paste form,

through a nozzle to build up an object layer by layer (Wolfs, 2019). In the pre-extrusion phase, as shown in Fig.1, the food formulation must remain fluid which is achieved by ensuring that the used food materials have a small particle size. The result of extrusion-based 3D food printing is achieving a comparable geometric product obtained through digitalized design while allowing personalized nutritional control (Sun, 2018). Single or multi nozzles can be used one by one or simultaneously to print products according to consumer demands producing unique tastes, and customized nutritional profiles. This 3D printing technique is suitable for producing carbohydrate based food, such as bread, pizza and cookies, etc, due to the potential of high precision and industrialization in carbohydrate based 3D food.



(a) Syringe-Based Extrusion (b) Air Pressure Driven Extrusion (c) Screw-Based Extrusion

Fig. 1. Extrusion mechanisms a) syringe-based; b) air pressure driven; c) screw-based (Sun, 2018)

Ingredient formulations

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3D printing trials were conducted with ingredients commonly found in foods, which contained components from all main groups of nutrients, i.e. carbohydrates, protein, fat and dietary fiber, either alone or in combination with other ingredients (Lille, 2018).

Various food formulations and food recipes can be tested in order to maintain not only nutritional value but also to preserve the sensorial characteristic like texture in cookies. This simple but widely appreciated treat contains fat to provide lubricity or mouth coating, starch to provide viscosity and proteins to provide a range of textures and nutrition. Acknowledging the major health problems of modern society, obesity and chronic diseases digitized food printing offers the key to upgrade the recipe of existing food products' nutritional profile by reducing or eliminating undesirable ingredients, or replacing them with healthy ingredients (i.e. fiber and micronutrients such as vitamins and minerals) (Wolfs, 2019). The most successful materials for 3D food printing are dough (with or without additives) due to its consistency, rheology and ability to solidify after printing. Additives like starches, polysaccharides or proteins have a positive effect on melting behaviour, glassy state and plasticization of 3D pastes during liquid- and powder-based 3D printing (Bhandari, 1999). The flow behaviour and viscous modulus of a dough affect its extruding action, while its elastic modulus, gel strength, rupture strength and adhesiveness influence its ability to support the 3D structures. The dough can be easily extruded because of its pseudoplastic behavior with its viscosity decreasing as the shear rate increases. Various starch types have specific shear-stability and a yield stress which helps in retaining the shape after deposition. But starch paste concentration will define printing technology, mostly the extrusion of the material through the syringe tip and final shape, precision and stability of 3D printed objects. The starch is used in 3D paste as a thickening agent due to its ability to gelatinise by heat in the aqueous suspension.

Apart from some gum-like protein (e.g. gelatin), most proteins cannot be used directly as a 3D printing raw material. Denaturation of proteins under external stress (temperature or mechanical strength) or compounds (e.g. strong acid or base) is a good way to prepare raw material adapted to 3D printing. Proteins can be deposited with polysaccharide materials (e.g. alginate) in a layered structure under the defined conditions. Regarding proteins isoelectric points and possible aggregation, protein based gelation and hydrogel forming mechanism can be widely used in 3D food printing. Due to various external effects like, pH and temperature variations, proteins can be transformed in easy to print pastes in mixture with fibres and various sources of carbohydrates. Plant proteins and soluble fibres (from oats and barley) can be used as flour replacements, sugar can be replaced by natural sweeteners, fats could be enriched trough addition of Chia and flax seeds or its oils rich in omega-3 fatty acids. 3DP offers great insight to every ingredient segment which also offers possibility to develop recipes for people with severe allergies, like dairy products, nut products, gluten intolerance, etc.

But what about minor ingredients? How can we add vitamins, natural colorants and bioactive compounds like anthocyanins, carotenoids, betanidins, plant polyphenolic compounds, or essential fatty acids?

By combining established techniques like electrospinning and microencapsulation in addition to 3D printing technologies. These technologies can be directly integrated into the food printing process through multi-print head platform, to control ingredients dispensing. Micro- encapsulation can pack minerals, vitamins, flavors and essential oils within another material for the purpose of protecting active ingredients from the close environment. Electrospinning can produce food materials with preferred size and structure (lower fat and lower salt) with appropriate sensory properties and novel ingredi- ents (Neethirajan, 2011).

Chemical and physical changes that are developed during food processing may not occur during the 3D printing

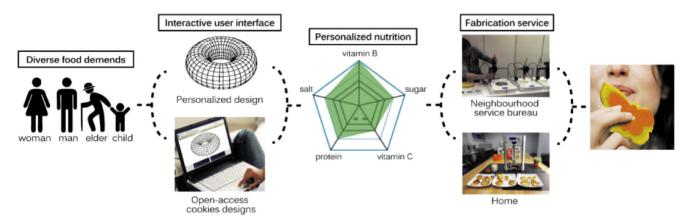


Fig. 8. Schematic diagram of food design and fabrication service.

Fig.2 Schematic diagram of food design and fabrication process

process and this is due to ingredients and their interactions, structure, texture, and taste, but also 3DP machines. The crucial factor here is to adapt the recipe design and printing process to achieve the planned contents on the sensorial and nutrition profiles (Fig. 2). 3DP pre-processing includes the adaptation of ingredients (e. g. gluten formation and leavening), but also post-processing (e. g. forming and baking, cooking, drying).

Future of 3DP

Food printing has demonstrated its ability structure personalized chocolates and produce simple homogenous snacks, but these applications are still primitive with limited internal structures or monotonous textures. Food is the most complex material to shape, and it is necessary to study printing materials, platform designs and printing technologies thoroughly. There are several EU-funded projects that support the development of food printing to produce tailor-made soft foods with tailored nutrient contents for older people with swallowing or chewing difficulties, which provides a great insight into the future importance of 3DP foods. However, in order to obtain better and more accurate data on people's nutritional needs and energy intake, data modelling through algorithms will allow us to obtain accurate food intake based on the height, weight, gender and health needs of each patient. To obtain similar foods as conventionally produced, detailed studies on ingredient effects on the texture and its interaction, as well as the accurate effect of cooking-related chemical reactions should also be modelled. The future for 3DP will bring a tailor-made food supply chain where consumers will be able to produce physical products that are not needed at home but in nearby local restaurants, resulting in lower distribution costs, simplified food supply through a mobile and web application and satisfied customers3DP offers a zero waste cycle of food waste in any household or restaurant kitchen, but also ensures the development of novel and functional foods with different textures and flavours. Fine dining restaurants could reduce prices of chefs' high end meals by using 3DP final decoration steps which could replace manual decoration as well to precisely calculate almost each drop of ingredients on the serving plate. These are just a few possible examples of the use of 3DP in the food sector, and there are certainly many more that we may not yet be aware of.

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Activities of the Croatian Academy of Engineering (HATZ) in 2020

Auspices, Organization/Coorganization of Conferences

Auspices

- Professional conference "nZEB in practice Designing buildings according to nZEB standards", February 20, 2020, Faculty of Architecture of Zagreb
- Faculty of Chemical Engineering, 13th meeting of young chemical engineers, February 20, 2020, Zagreb University of Zagreb Faculty of Textile Technology, 13th
- International Scientific and Professional Conference "Textile Science and Economy", Faculty of Textile Technology, Zagreb, April 24, 2020
- Conference "Baška GNSS Conference Technologies, Techniques and Applications Across PNT", May 17-21, 2020, Baška, Island of Krk
- Faculty of Civil Engineering, 6th International Conference on Road and Rail Infrastructure - CETRA 2020, Pula, May 20-22, 2020
- 2nd International Students' GREEN Conference, May 21-22, 2020, Faculty of Agrobiotechnical Sciences Osijek, Osijek
- 13th International Conference "International Conference of the Croatian Nuclear Society - Nuclear Option for CO2 Free Energy Generation", Croatian Nuclear Society, May 31 to June 03, 2020, Zadar
- Conference "The 8th International Symposium on Applied Electromagnetics", June 28 – July 01, 2020, Beli Manastir 18th Ružička Days "TODAY SCIENCE – INDUSTRY
- TOMORROW", September 16-18, 2020, Vukovar
- 18th International Scientific-Professional Conference ,,The Science and Development of Transport ZIRP 2020", September 29-30, 2020, Šibenik
- International Conference on Smart System and Technologies 2020 (SST 2020), October 14-16, 2020, Osijek

Organization/Coorganization of Conferences

· Croatian Engineer's Day, March 2, 2020, Zagreb

Participation at Meetings of Public Interest

Department of Chemical Engineering of the Croatian Academy of Engineering in cooperation with the Faculty of Chemical Engineering of the University of Zagreb, Lecture "Plasmonic phenomena and photoelectron generation in Au/TiO, nanorod arrays for visible light harvesting" and lecture "Formation of a junction between TiO_2 and β - Bi_2O_3 to enable efficient visible-light harvesting in advanced oxidation processes for waste water treatment", January 20, 2020

- Faculty of Textile Technology, Faculty Day, January 24, 2020, Zagreb
- Faculty of Chemical Engineering, presentation of the book "Modeling in Chemical Engineering" February 04, 2020, Zagreb. Authors Prof. Emeritus Zoran Gomzi and retired Prof.dr.sc. Zelimir Kurtanjek, both in emeritus status within the Department of Chemical Engineering of the Croatian Academy of Engineering
- Presentation of opportunities for Inclusion of the Croatian Economy and Science in EUMETSAT Projects, February 13, 2020, Croatian Chamber of Economy, Zagreb
- Croatian Wood Cluster, 10th International Conference on Biomass and Renewable Energy, February 24, 2020, Zagreb
- Presentation of the new visual identity of Faculty of Electrical Engineering and Computing, March 13, 2020, FER, Zagreb, cancelled
- Two lectures in co-organization of the Department of communication systems ("CubeSats: Toy or Tool? -Commercial & Scientific Use Case Examples", "Cubasat development in a University lab – lessons learned"), March 2020, 16-17, Faculty of Electrical Engineering and Computing, Zagreb
- E-infrastructure Days, SRECE DEI 2020 and Conference of the hr-Zoo project, April 7-8, 2020, Zagreb
- Department of Chemical Engineering of the Croatian Academy of Engineering, lecture ,,The Resource Gateway: Microfluidics and Requirements Engineering for Sustainable Space Materials Processing Systems", June 03, 2020, Faculty of Chemical Engineering, Zagreb
- Conference "Climate and clean water", June 26, 2020, Zagreb
- Day of Energy Institute Hrvoje Požar, July 03, 2020, MSU Zagreb
- 43rd International Conference on Information, Communication and Electronic Technology, MIPRO 2020, September 28 to October 02, 2020

Note: Some of the above mentioned HATZ activities were not carried out due to COVID-19 disease, but based on information from the organizers, they will take place later in some form or another, about which the HATZ members will be informed in due course.

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