Citrus Uses in the Food Industry

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Citrus fruits occupy an important position in the context of the fruit trade, considering that both fresh fruits and processed products are produced on a large scale. Citrus fruits are recognized as an essential component of the human diet, thanks to their high content of beneficial nutrients such as vitamins, minerals, terpenes, flavonoids, coumarins and dietary fibers. Among these, a wide range of positive biological activities are attributed to terpenes and flavonoids derivatives.

Keywords: Citrus ; orange peels ; nutraceutical values ; industrial applications

1. Introduction

Nowadays, consumers are more aware of the relationship between diet and health. In fact, the global consumption pattern has shifted toward foods that provide both nutritional value and health benefits. Therefore, the demand for functional foods has experienced an exponential growth in recent years [1].

Citrus fruits are excellent sources of natural bioactive compounds with well-known health-promoting properties. As previously described, the main citrus phytochemicals include polyphenols (mainly flavonoids), carotenoids, vitamins, organic acids, dietary fiber, and essential oils, which have promising biological activities due to their anti-oxidative, anti-inflammatory, and anti-carcinogenic properties [2]. These health benefits associated with citrus bioactive compounds are one of the main reasons the citrus-based food industry is expanding [1].

The mentioned citrus phytochemicals are mostly found in citrus-fruit wastes, namely, citrus peels, seeds, pomace, and pulp, which accounts for about 50 to 60% of the total weight of the fruit. Large amounts of processing waste are generated during the production of orange and other citrus juices, mainly the peel, cores, and segment membranes. OP is the main by-product, accounting for about half of the fruit mass. In fact, the citrus-processing industry generates more than 60 million tons of waste worldwide [3]. In the USA alone, juice processing of oranges and grapefruits generates more than 5 million tons of citrus waste [4] and 700,000 tons of OP annually [5]. In India, about 2.15 million tons of citrus peels out of 6.28 million tons of citrus fruits are generated from citrus juice processing annually.

When OP goes unused, they become waste and are a possible cause of environmental pollution. Therefore, it is urgent to investigate and find solutions to convert these wastes into economically valuable products. OP were traditionally dried and marketed as livestock feed or used in the food industry [6]. They have been used to produce animal feed [7][8], single-cell proteins, fibers, pectinase/cellulose [9], immobilization support [10], ethanol, and bio-sorbents for heavy-metal removal [11]. Currently, the extraction of phenolic compounds from OP has attracted considerable scientific interest for use as natural antioxidants, especially in foods to prevent rancidity and the oxidation of lipids [12][13][14].

2. Extraction of Bioactive Compounds for Food Applications

In recent years, considerable attention has been paid to the extraction of bioactive compounds from citrus waste for further use as food additives, encapsulants, nanoparticles, prebiotics, or as a source of pectin, essential oils, polyphenols, carotenoids, or dietary fibers [15][16][17][18].

The extraction, isolation, and characterization of the mentioned bioactive components from citrus waste represent crucial steps in their recovery. Appropriate extraction conditions must be tested, optimized, and used to ensure that no degradation or loss of the bioactive compounds occurs. In fact, the solvent selection and extraction technique are the most crucial steps to maximize the yield of bioactive compounds [1][2]. Recently, several authors have focused their efforts on describing, in detail, the extraction techniques as well as the conditions employed to recover the different bioactive compounds from citrus wastes to improve the extraction yield [3][19][20]. Traditionally, the most commonly applied techniques include maceration, hot-water extraction, solvent extraction, and alkaline extraction [17][20]. Dar et al. [21] employed a maceration method to recover bioactive compounds from citrus peels, testing different solvent compositions (ethanol, water and 50% aqueous ethanol) for 8 h at room temperature. These authors reported that the extraction yield was higher with aqueous ethanol, followed by ethanol and water (29.28, 23.65 and 6.53%, respectively). In another study [22], phenolic compounds, ascorbic acid and the free radical scavenging activity of peel and pulp from Orlando orange, Kinnow mandarin and Eureka lemon fruits were assessed applying a conventional extraction method (80% aqueous ethanol at 70 °C in a shaking water bath for 3 h). These authors reported different levels of compounds in the matrices studied, demonstrating the major advantage from conventional extraction methods versus the most recent ones, namely, their versatile application in different matrices as well as their easy operation and low cost of application. The work developed by Gómez-Mejía and co-workers [23] is also in-line with these achievements. These authors tested the extraction of bioactive polyphenols from different citrus peels by employing magnetic stirring, evaluating the influence of extraction time (X1; 10–15 min), ethanol–water ratio (X2; from 20:80 to 40:60 *v*/*v*), and extraction temperature (X3; from 62 to 90 °C). Depending on the type of citrus peel, different extraction conditions were validated, but these authors concluded that the proposed conventional extraction method uses a lower amount of ethanol, reduced extraction times and a lower sample-to-solvent ratio than the novel techniques. Nonetheless, significant efforts have been made to replace these traditional techniques with green extraction methods, such as ultrasound-assisted extraction (UAE), subcritical water extraction (SWE), supercritical fluid extraction, microwave-assisted extraction (MAE), and enzyme-assisted extraction, not only to improve the extraction efficiency of bioactive compounds from citrus waste but also to overcome some of the most common drawbacks of traditional extraction techniques [15][16]. Šafranko et al. [16] investigated a two-step green extraction technique to recover bioactive compounds from mandarin peel (*Citrus unshiu* Marc. Var. *Kuno*). Firstly, these authors employed a supercritical CO2 (SC-CO2) extraction to recover volatile compounds, reporting limonene as the dominant component (30.65% at 300 bar), followed by farnesene, linoleic and hexadecanoic acids. Afterwards, the residue from SC-CO2 treatment was subjected to different SWE conditions to obtain bioflavonoids. Hesperidin (0.16–15.07 mg/g) was the most abundant flavanone in mandarin peel, followed by narirutin, and rutin. However, these authors reported that for extraction temperatures higher than 160 °C, the possible formation of undesirable compounds, such as chlorogenic acid and 5-hidroxymethilfurfural, can represent a limitation for the large-scale exploitation of the SWE technique. Another environmentally friendly extraction technique widely used is UAE [18]. Ordóñez-Santos et al. [18] employed UAE for the recovery of carotenoid compounds from mandarin epicarp to be further used as a natural colorant in bakery products. These authors reported a total carotenoid amount of 140.70 ± 2.66 mg β-carotene/100 g of dry sample for an extraction performed at 60 °C for 60 min. Montero-Calderon et al. [24] also employed a UAE technique to extract bioactive compounds from orange (*Citrus sinensis*) peel. At the optimal UAE conditions (50% aqueous ethanol, 30 min, 400 W of power), a total carotenoid concentration of 0.63 mg ß-carotene/100 g, vitamin C concentration of 53.78 mg AA/100 g, a total phenolic content of 105.96 mg GAE/100 g, and a hesperidin maximum concentration of 113.03 ± 0.08 mg/100 g were obtained. Despite the advances in the recovery of bioactive compounds from citrus matrices, most of the applied environmentally friendly extraction methods still cause concerns about the health and safety of the produced bioactive extracts, and the possible degradation and/or formation of undesirable compounds due to high temperatures [20]. Indeed, Benassi et al. [25] tested three different techniques, namely, conventional hot-water extraction, rapid solid–liquid dynamic (RSLD) and MAE, to recover pectin from waste orange peel. These authors concluded that the “hot-water” extraction assisted with citric acid was the most sustainable extraction route, ensuring higher extraction yield (21%) as well as a high quality of the extracted pectin (degree of esterification of 82.5%). This fact evidences the main advantage of the traditional extraction method—namely, its simplicity of operation and/or equipment—over the novel UAE, MAE or SWE, especially at an industrial scale. Therefore, researchers need to increase their efforts to find more sustainable, economical, and rapid techniques to recover the different bioactive compounds from citrus waste and to enable their safe incorporation into food products.

3. Food Industrial Applications

3.1. Functional Food Ingredient

In recent years, consumer attention is moving towards consuming dietary-fiber-enriched foods. Considering that citrus fruits are excellent sources of antioxidants and dietary fibers, their inclusion in daily consumed foods such as baked goods, meat, and dairy products has become a hot topic of scientific research. Several authors have reported the potential of using citrus powder or flour in bakery and confectionery products as a functional ingredient [26][27][28]. For example, Caggia et al. [26] developed a low-fat bakery product (brioches) fortified with proportions (30, 50 and 70%) of debittered orange fibers, which improved the stability and nutritional properties of the developed product. The results obtained demonstrated that the addition of 50% debittered orange fiber resulted in a fat content of 4.5% in the products, in comparison to the 10% fat in the control sample.

Furthermore, due to the antimicrobial properties of these natural extracts, food product safety was also ensured. In another study [28], the positive effects of citrus albedo addition on bread shelf life due to the high pectin and fiber content was confirmed. The authors demonstrated that the partial replacement of wheat flour with dried fruit-peel powder provided a higher ability to bind large amounts of water. However, changes in the mechanism of staling, as well as structure modification as a consequence of fortification, should be further investigated. Iftikhar et al. [27] also demonstrated that *Citrus sinensis* (sweet orange) peel can be used to enhance the nutritional and functional properties of cakes due to their fiber and fat content. These authors concluded that the mixture of wheat flour with up to 3% citrus-peel flour is suitable for the development of cake with acceptable sensory attributes.

3.2. Food Additive

Food additives are responsible for the flavor, color, taste, and nutritional qualities of food products. In recent years, with the increasing consumption of organic foods, the replacement of synthetic food additives with natural ones represents a great advantage in the field of food-processing industry [2]. In this sense, another reported application for the bioactive compounds with antioxidant activity recovered from citrus waste is their use as food additives, especially in the preparation of candied products for confectionery/baking industry [29][30]. For example, Romero-Lopez et al. [31] prepared muffins enriched with different proportions of dietary-fiber-rich orange bagasse and reported that the prepared muffins (with 15% extract) had a high dietary fiber (15.3%) and low fat (15%) content compared to the control muffins. Furthermore, the addition of the dietary-fiber-rich orange-bagasse extract to the muffin reduced the predicted glycemic index, and no difference in sensory evaluation was observed between the control muffin and the muffin prepared with dietary-fiber-rich orange-bagasse extract. These results are of the greatest interest because the addition of dietary-fiber-rich orange-bagasse extract to bakery products may be an alternative for people who require foods with low glycemic index. In another work, Ojha and Thapa [32] also prepared biscuits by replacing the wheat flour with mandarin-peel powder (3, 6, and 9%). They reported that biscuits formulated with 6% of mandarin-peel powder were comparable to the control biscuits with no substitution; the content of fiber, ash, ascorbic acid, carotenoids, polyphenol and antioxidant activity improved, and the reported values were 0.85%, 1.32%, 1.5 mg/100 g, 69 μg/g, 2150 μg gallic-acid equivalents/g and 24.5%, respectively.

Regarding the application of citrus extracts as food additives in another type of product, the research from Nishad et al. [29] should be highlighted, which investigated the potential of using citrus-peel extracts in the maintenance of oxidative stability of meat balls during frozen storage. The authors demonstrated that the natural antioxidant extracts from citrus peels can control lipid oxidation in meat products, by inhibiting enzymatic reactions responsible for oxidative damage. Moreover, the addition of citrus extract had a positive effect on the color, flavor, and overall sensory properties of the meat balls, indicating that it can be used as a natural preservative for foods rich in fatty acids. Younis et al. [33] also incorporated the mosambi peel, a by-product of the juice industry, in sausages and patties and reported an enhancement in fiber content as well as in fat and moisture content. In addition, the addition of up to 6% of mosambi-peel extract improved storage stability, demonstrating the potential of using citrus waste as a food additive in meat products.

3.3. Food Colorant

The peels of citrus fruit are described as an excellent source of carotenoids. Not only do they impart color to fruits, these compounds also promote health benefits, which has attracted the attention of food industry as a solution to replace harmful synthetic colorants [1][17]. Barman et al. [34] used orange-peel waste to extract β-carotene, which was used to develop a stable nanoemulsion to be further employed as a natural colorant in food products. These authors reported that the addition of the nanoemulsion to fruit juice significantly enhances its color, thus providing an alternative to the use of synthetic colorants. Ordóñez-Santos et al. [18] optimized the process of ultrasound-assisted extraction of total carotenoids from the mandarin epicarp and demonstrated its potential to reduce the use of tartrazine in bakery products, such as cakes and bread, and the potential of its further use as a natural coloring additive. Ciriminna et al. [11] also investigated the technical and economic possibilities of using lemon-peel waste to produce water-soluble yellow colorant limocitrol 3-O-6″-[3-hydroxyl-3-methylglutaryl)])-d-glucopyranoside as a substitute for tartrazine. The authors demonstrated that this natural colorant can be easily obtained by simple solid–liquid extraction in aqueous ethanol or via hydrodynamic cavitation of lemon-peel waste in water.

Moreover, the obtained results on the chemical and physical stability of this natural colorant open the possibility to explore the industrialization of this new bioeconomy production. Despite these promising results, further research is needed to overcome the main limitations, such as the high cost of using natural biocolorants in industrial food applications [1].

3.4. Flavoring Agent

Synthetic flavors are still widely used in the food industry; however, the use of citrus essential oils as flavoring agents is gaining increasing attention. Essential oils, mostly recovered from citrus peel, are prominent sources of terpenoids, which are widely used as flavoring agents in foods, and also have antibacterial, antifungal, and insecticidal properties [35]. Most studies have focused on determining the volatile profiles of different citrus species [36], and only few applications of citrus in the food industry as flavoring agents have been found. Bergamot oils, a rich source of linalool and linalyl acetate with promising flavor characteristics, have been used in some flour-based confectionery in recipes to replace bergamot peels [37]. Recently, Matsuo et al. [38] studied the effects of adding *Citrus natsudaidai* (CN) peel extracts to aqueous solutions and reported that the solutions flavored with CN extracts exhibited preferential odor over the commercial citrus-flavored beverages, which were classified in the same group as commercial citrus juices by the electronic nose test.

In addition, the solutions flavored with CN extracts exhibited sourness, bitterness, and an orange-like taste, and the overall acceptability was not significantly different from commercial citrus-flavored beverages. The use of citrus essential oils in ice cream, marmalade, and jam-like food products has also been widely described by other authors [36].

3.5. Thickening Agent

As previously reported, citrus wastes, especially citrus peel, are an excellent source of pectin, which is extensively used in jams, jellies, marmalades, milk, and confectionery products due to their gelling and stabilizing properties [19]. Many studies have focused on finding more environmentally friendly extraction techniques to recover pectin from citrus peels [25][39], and very few papers reported results on its incorporation in food products. For example, Mann et al. [40] reported the production of ice cream using frozen Kinnow peel; both unblanched and blanched, at three levels (1, 3 and 5%). The addition of Kinnow peel improved the appearance, flavor, and overall acceptability of the ice-cream samples. The authors reported that the content of ascorbic acid and flavonoids (namely, naringin) in the ice-cream samples increased with the addition of Kinnow peel, showing that the best levels of frozen Kinnow peel, based on sensory evaluation, were unblanched—3% and blanched—5%. Mohamed et al. [41] reported the extraction of pectin from white and red Sudanese-grapefruit peel and confirmed that the gel-forming quality of the extracted pectin was similar to that of commercial pectin.

Jellies prepared with both types of grapefruit peel pectin set within the 10–25 min, indicating them to be rapid-set pectin and demonstrating their potential to be used as a stabilizer/thickening agent in different food products. In another study [42], jams were also prepared and their physicochemical and sensory properties analyzed. The authors extracted, characterized, and applied pectin recovered from grapefruit peel from Duncan cultivar to jam formulations and observed a significant effect on the texture of the final product.

4. Limitations of Applying Citrus Wastes in Food Industry

Several studies have demonstrated the promising potential of incorporating citrus-waste extracts in food-industry products. However, some parameters of citrus extracts, such as their low stability and water solubility, limit their further use at a larger scale. Since most of the bioactive compounds present in citrus extracts have poor bioavailability and increased sensitivity to different environmental conditions, such as pH, heat, and oxidation, their protection is a major challenge for the food industry in commercial applications.

In addition to these limitations, the conversion of citrus wastes into value-added food products raises concerns about the safety and toxicity of the citrus-waste extracts used [43]. In general, the potential of citrus wastes to be used as novel functional ingredients with a specific function is well-described; however, the evaluation of their safety has not yet been established. Nevertheless, the use of citrus wastes in food products must comply with current legislation and a risk assessment must be performed to assess their safeness, and very few studies have been conducted recently to address these issues [26][44]. Therefore, a holistic research approach is needed to integrate the value-addition strategy with risk analysis and to apply forecasting and optimization studies to the whole supply chain.

Furthermore, industrial-scale studies on the use of citrus food are still very limited, although they are also extremely necessary to define the barriers to a large-scale application. Therefore, collaboration between academic and industrial partners may be the key to increase the value of citrus-processing industries by converting their wastes into functional food products.

5. Application in Food Packaging

According to definition reported in the EC Regulation No 450/2009, "active materials and articles means materials and articles that are intended to extend the shelf-life or to maintain or improve the condition of packaged food; they are designed to deliberately incorporate components that would release or absorb substances into or from the packaged food or the environment surrounding the food".

Active packaging technology provides several advantages over the direct addition of active compounds to the packed food, such as the lower amounts of active substances required, the localization of activity at the surface, migration from the film into the food matrix, controlled release systems, and the elimination of additional steps within a standard process intended for introducing the active compounds at the industrial processing level, such as mixing, immersion, or spraying. Controlled-release systems are of industrial importance as they can prevent sensory or toxicological problems or inefficiencies of the system caused by too-high or too-low concentrations of the delivered substance [45].

In the review by Han et al. [46], the problems related to development of antioxidant and antimicrobial active packaging are well-defined, making it quite difficult to set specific targets for the selection of the natural extracts to be used due to the absence of reference benchmarking products. Recent research trends have focused on the development of active food packaging by adding antioxidants into packaging materials to extend the shelf life of the product. The most commonly used synthetic antioxidants in the food industry are butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT) and propyl gallate in U.S.A., especially for packaging cereals and snacks [47]. However, synthetic antioxidants can also be carcinogenic and harmful to consumers. This must be considered in active packaging, as migration from food contact materials is not negligible and is, indeed, a desired phenomenon. With increasing health awareness and consumer's demand for reduced use of chemicals in food packaging, more attention has been paid to finding naturally occurring, safe substances that can act as alternative antimicrobials and antioxidants. The use of natural antioxidants derived from plant extracts in food packaging is becoming increasingly popular.

Extrusion is the most popular technique to include natural extracts into the final formulation [45]. In this technique, bioactive compounds are incorporated before extrusion, so that the high temperatures of extrusion (the exact values depend on the melting temperature of the processed polymer) allow their effective and homogeneous distribution in the film, although, at the same time, they are responsible for the thermal degradation of the bioactives' activity. For example, Ha et al. [48] used a high-temperature profile (160–190 °C) to extrude an antimicrobial LLDPE-based film, which resulted in a high loss of functionality of the grapefruit-seed extract (GSE) and loss of antimicrobial activity. For this reason, heat-sensitive bioactive agents (i.e., natural extracts) should preferably be incorporated into the packaging using non-heating methods (e.g., electrospinning and surface coating). Among these methods, surface coating, in particular, is a simple process that relies on low temperatures but may suffer from poor adhesion to plastics and needs to be designed to be in direct contact with the food in cases where active packaging is the final objective of material production.

Natural extracts are already produced and commercialized by different companies, mainly for direct use in food or for the cosmetic and pharmaceutical industries. There are numerous scientific articles on the incorporation of such natural extracts to extend the shelf life of food products. However, large-scale demonstration is still pending [49], especially in relation to their use as packaging materials. While synthetic antioxidants are generally added to improve the properties of the materials during processing, natural antioxidants suffer from the major drawback of thermal degradation at the typical working temperature of the extrusion processes [50]. Therefore, encapsulation of antioxidants can improve their thermal resistance so that they can be incorporated directly into the plastic pellets before extrusion.

Green tea, rosemary extracts, essential oils and various fruit extracts are the most-used antimicrobial and antioxidant products investigated in the literature for packaging applications [51][52][53][54][55]. In 2015, Goglio Spa (Italy) won the Packaging Oscar for its product GTea®, an active packaging with a green-tea extract.

As the importance of environmental sustainability and circular economy is increasingly recognized, it would be better to use extracts obtained from agri-food residues, such as from orange peels.

5.1. Natural Extracts Requirements for Incorporation into Packaging Material

A key point in selecting the extract to be incorporated in active packaging is obviously the food-grade characteristic. It would be the best to use food-grade extracts since there will be no problem with migration restrictions, especially if active packaging is planned with the expected release of extracts into food. On the other hand, if the extract is not food-grade, it will have to be exploited to absorb substances from the packaged food or the environment surrounding the food and then incorporated only into the external coating, or into an intermediate layer (i.e., by incorporation into an adhesive if lamination is used to manufacture the multilayer film) with a barrier layer which prevents migration into the food.

If the aim is to provide antioxidant and/or antimicrobial activities, these properties are crucial for selection and it is necessary to verify the maintenance of the property after incorporation and over time. Furthermore, depending on the selected target food to be packed and the coloring power of the extract, this property could be incompatible with obtaining a suitable transparent packaging. However, it should be noted that many natural extracts, such as those from citrus species, have antioxidant/antimicrobial activity due to the presence of phenolic compounds, which are often colored. At the same time, the presence of phenolic compounds and carotenoids with the ability to absorb light in the 200–800 nm range could be of interest, as UV-Vis light can catalyze many degradation reactions in food products.

5.2. Need for Encapsulation

As explained earlier, natural extracts are sensitive molecules that can be denatured under harsh conditions. Encapsulation may be necessary to provide suitable solubility in the coating medium (when incorporated into the packaging material via a coating application), thermal stability at processing temperatures (when incorporated into a plastic polymer prior extrusion step), and/or light stability.

The thermal stability of natural additives is the main problem in cases of the direct incorporation of the extract for compounding a functionalized polymer masterbatch. The working temperature during extrusion to form the plastic film is the most challenging point, since it can exceed 150–200 °C depending on the processed polymer. For this reason, direct incorporation prior to film extrusion is often discarded. Spray-drying encapsulation can be applied to increase the thermal stability of the extracts. Thermal stability may also be required for some specific uses of packaging in the food industry, such as hot filling and thermal treatment after packing. In addition, other encapsulation technologies may be considered: extrusion with vibrating nozzles, jet cutter, coacervation and others.

Information on the maximum temperature that could be reached during the coating preparation or during the melting/extrusion process of the plastic material (in the case of direct incorporation of the extract into the polymer), or, eventually, by the final food industry end user, is, therefore, key to defining thermal-stability requirements.

Solubility in water or in another solvent is necessary if the extracts are to be incorporated via a coating application, depending on the solvent on which the coating is based. Furthermore, in Europe, the legislation for plastic food-contact materials (FCM) reports the use of different simulants to simulate the different ranges of food products (ethanol 10%, 20%, 50%; acetic acid 3%; vegetable oil with less than 1% unsaponifiable content, and simulant E for dry foods). Evaluation of solubility in these simulants is important in terms of desired or undesired release in the packaged food.

Encapsulation may affect and eventually improve thermal stability and solvent solubility, depending on the carrier materials used, but it is also important to check the potential effect of the encapsulation process on the antioxidant and antimicrobial properties of the original extract.

5.3. Literature Examples of Citrus Extracts Use to Develop Antimicrobial/Antioxidant Packaging

Plant/fruit extracts or essential oils are known for their potential antioxidant and antimicrobial properties and have been widely investigated in the literature for these properties as well as for their use in active food packaging [56]. Many of the extracts studied are obtained from fresh plants, fruits and herbs, but not from processing residues.

The antibacterial capacity of 32 essential oils against five foodborne (*L. monocytogenes*, *S. aureus*, *E. coli*, *S. Typhimurium*, *P. aeruginosa*) and spoilage bacteria in liquid phase (as minimum inhibitory concentration, MIC, values) was evaluated by Ghabraie et al. [57]. Among the oils tested, Chinese cinnamon, cinnamon bark and wild-bergamot essential oils were the only ones that exhibited inhibitory activity against all five pathogenic microorganisms tested.

However, these essential oils were not produced from residues and by-products, like in the case of essential oils from fruit peels. In addition, essential oils have a typical strong flavor that may interfere with their use in food packaging, as they could have a strong impact on the sensory profile of the packed foods.

Grapefruit-seed extract is made from the seeds and pulp of grapefruit and it contains tocopherol, citric and ascorbic acids [58]. The antioxidant and antimicrobial effects of this extract have been reported in different products such as ground beef [58]. There are several studies in the literature in which grapefruit-seed extracts were incorporated into bio-packaging [52][59], which showed good antimicrobial activity against *L. monocytogenes* and *E. coli*.

Kanmani and Rhim [60] incorporated a GSE at different concentrations (from 0.6 to 13.3 μg/mL) into an agar-based film through a casting technique and the obtained films were evaluated for their antimicrobial activity against *L. monocytogenes*, *Bacillus cereus*, and *E. coli*. Only the enriched films showed antimicrobial activity, with an extract-dose correlation, and better performance against *L. monocytogenes* (Gram +) than against *E. coli* (Gram -), probably due to the specific outer membrane which inhibits the diffusion of the active compounds through the lipopolysaccharide layer.

Extracts can be obtained from citrus peel, which is a residue of citrus-juice production. Jridi et al. [61] incorporated phenolic extracts from red-orange peels (*C. sinensis*) in both dried and fresh forms at concentrations of 5 and 10 mg/mL into a fish gelatin-based film using the casting technique. The films were tested for their antimicrobial activity against *Micrococcus luteus*, *Staphylococcus aureus*, *Bacillus cereus*, *Pseudomonas aeruginosa*, *Salmonella enterica*, *Listeria monocytogenes* and *Enterobacter* sp. Some antimicrobial activity was exhibited against all the tested microorganisms, with *S. aureus* being the most sensitive. In general, the fresh extract was more effective than the dry extracts, showing a reduction in activity after drying.

The focus of the study of Bassani et al. [62] was the development of an innovative, biodegradable, and sustainable PLA-based active-packaging solution incorporating an antioxidant extract from orange peel. The extract was first obtained using hydro-alcoholic extraction and then purified by a resin absorption process (which was required to remove sugars and organic acids and increase antimicrobial activity). The extract contained up to 50% total phenols on dry matter and it was either freeze-dried or spray-dried with pectin or ß-cyclodextrins as a carrier material to obtain powder formulations. Encapsulation could improve thermal stability compared to the freeze-dried extract, particularly when cyclodextrins were used. The three powder extracts were incorporated into commercial PLA at different concentrations (0.25, 0.50, 1.50, 2.0 wt %.) and film samples were obtained by hot pressing. The films were assayed to evaluate the influence of the extract addition on the thermal stability of the polymer, color, and mechanical properties of the films under accelerated light storage conditions (in a Suntest XXL+ aging chamber, for 500 h). Positively, extract addition preserved the transparency of the bioplastic and did not modify the degradation temperature profile of the PLA film. However, the extracts resulted in a yellowish coloration that increased with the amount added and achieved an unacceptable browning at a dosage of 2%. The accelerated light storage test highlighted that encapsulation improved color stability and that the film performed worse in terms of mechanical properties (Young module, tensile strength and elongation at break) when it was enriched with the extracts (in this case, the freeze-dried formulation was preferable to the encapsulated ones).

In the work of Fiorentini et al. [63], different commercial citrus-peel extracts were investigated for their thermal stability, which was then improved by a spray-drying encapsulation process with beta-cyclodextrins. The study revealed that the antioxidant capacity was retained after the encapsulation process, with an apparent 20–25% reduction in the total phenolic content of the original extract. In addition, the antimicrobial activity against *S. aureus* was almost unaffected by spray drying, with MICs ranging from 5–0.625 mg/mL to 5–1.25 mg/mL. The encapsulated extract with the best antioxidant and antimicrobial activity was incorporated into a polylactic acid/polyhydroxy butyrate (PLA/PHB) film produced on an industrial scale by cast extrusion. The obtained extract-enriched film was proven to be compliant with European regulations for food-contact materials in relation to overall migration in contact with acidic, hydrophilic and fat-containing food categories. The authors also evaluated the migration of active compounds and observed a potential release of 13.41% in hydrophilic food products and 11.02% in acidic products (pH < 4.5). The film showed growth inhibition by 30 and 60% against *E. coli* and *S. aureus*, respectively.

Colon and Nerin [64] tested a grapefruit dried hydroalcoholic extract for incorporation into a coating applied on a PET film. The system was prepared according to EU patent EP1477519-A1.41 and the coating was applied at room temperature reaching a maximum of 40 °C using hot air to eliminate the solvent. Active films were prepared with different weight percentages of the active agent/active layer, which varied from 0.7 g active/m^2 film to 3.0 g active/m^2 film. The obtained films were subjected to a free radical gas stream to increase oxidation and the antioxidant capacity was then determined based on the oxygen radical absorbance capacity (ORAC) assay. The grapefruit extracts revealed a lower antioxidant activity (based on ORAC assay) compared to green-tea or green-coffee extracts.

A commercial grapefruit-seed extract (GSE) was also used by Wang et al. [59] to develop active films based on poly(lactide) (PLA) or antimicrobial low-density polyethylene (LDPE). It was prepared by mixing corn starch with 20% wt glycerol (as plasticizer) and 40% of GSE and heating at 120 °C for 30 min in an autoclave. The plasticized starch mixture was then cooled to room temperature and pulverized using a blender. The obtained powder was mixed with the plastic resins (PLA and LDPE) at the ratio of 1:10 *w*/*w* to obtain the masterbatches, which were cast and blow extruded, respectively. The addition of the extract decreased the lightness of the films and increased the color (increase in the chromatic coordinates a* and b*) due to both the dextrinization of the starch and the phenolic content of GSE. The starch blend addition increased the LDPE thickness but not the PLA film. This is due to the fact that the hydrophilic thermoplastic starch is more compatible with more hydrophilic polymers, such as PLA. The antimicrobial activity of the films was tested against *L. monocytogenes* and *E. coli* and was high for the enriched PLA film, but not for the LDPE. Finally, the capacity of the films to act as active food packaging was tested on a fish paste, where the activity of the GSE-PLA film was confirmed, while the GSE-LDPE showed inhibitory activity only after 6 days of contact. This was consistent with the results of the migration tests, which showed a slower release of the GSE components from LDPE than from PLA.

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