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Abstract	driven the food industry to find altern the consumers with disorders related deals with the descriptions, character friendly and edible films, coatings at bakery products. Particular requirement to gluten free bakery foods are remark different components, preparation and methodology to determine films and addition, the main applications of edible and extend the shelf-life of bakery pro- the overall importance of edible matri- protect, retain or control the release of attention was paid to the potential of to incorporate probiotics, bioactive formulation of gluten free baked pro- highlight the relevance of edible cover	in relation to gluten intolerance has natives to provide adequate products to to gluten intake. The present chapter risation and main applications of eco- nd toppings used in the production of ents that must be fulfilled to be applied ed. A description and guidelines of the application techniques, as well as, the coating properties, are summarised. In le matrices to improve the global quality oducts are listed. It was also highlighted ices to constitute systems that allow to f different functional additives. Special f edible films and coating as vehicles compounds and/or nutrients into the oducts. Hence, this chapter pretends to ings as a useful and feasible strategy for attractive and nutritious foods products.
Keywords (separated by " - ")	Edible coverings components - Topping - Functional bakery products - Active	s constitution - Physical characterisation films and coatings

Chapter 7 Gluten Free Edible Films, Coatings and Toppings

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Abbreviations

AFM	Atomic force microscopy	8
ASTM	American society for testing materials	9
Tg	Glass transition temperature	10
GF	Gluten free	11
HPMC	Hidroxyporpyl methylcellulose	12
LDPE	Low density polyethylene	13
MC	Methylcellulose	14
OPP	Oriented polypropylene	15
SEM	Scanning electron microscopy	16

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S. Flores $(\boxtimes) \cdot L$. Gerschenson

17	a_w	Water activity
18	WVP	Water vapour permeability

19 7.1 Introduction

The gluten intolerances have determined diet changes based on the elimination of 20 ingredients that contain prolamins and glutenin from wheat, rye and barley being 21 replaced, in part, for alternative grains and tubers that do not induce the disease, for 22 instance, rice, corn, sorghum, and millet (Lebwohl and Green 2021). This has led to 23 an important challenge for the food industry due to the need of developing formula-24 tion strategies, generally known as "gluten free" (GF) ones, that include the use of 25 suitable additives linked to this dietary modification, while helping to produce 26 safety and organoleptically adequate food products (Zoumpopoulou and Tsakalidou 27 2019). According to the Food and Drug Administration (U.S.A.), the GF food is 28 defined as the food that does not contain gluten, or its presence should be lower than 29 20 ppm (McCabe 2010). 30

Bread and sweet baked goods (cakes, biscuits, doughnuts, etc.) are an essential 31 constituent of the human daily diet, representing the most important basic food 32 worldwide (Nils-Gerrit Wunsch 2020; Xu et al. 2020). There is a wide assortment 33 of such products and, a possible classification is the one proposed by Smith et al. 34 (2004) who grouped them as follow: unsweetened (bread, rolls, buns, crumpets, 35 muffins, and bagels), sweet (pancakes, doughnuts, waffles, and cookies) and filled 36 (fruit and meat pies, sausage rolls, pastries, sandwiches, cream cakes, pizza, and 37 quiche) goods. In their formulation, these products include complex carbohydrates 38 (mainly wheat flour), proteins, lipids, vitamins, and minerals (Soukoulis 39 et al. 2014). 40

Another classification proposed is based on the water activity (a_w), one of the 41 most important product properties affecting the physical and microbial deterioration 42 of bakery products. Smith and Simpson (1995) classified bakery products as follow: 43 (a) low moisture bakery products (cookies and crackers, $a_w < 0.6$) in which micro-44 biological spoilage is not a problem, (b) intermediate moisture products (chocolate 45 coated, doughnuts, Danish pastries, cream-filled cake, soft cookies, a_w 0.6-0.85) 46 where osmophilic yeasts and moulds are the predominant spoilage microorganisms, 47 and (c) high moisture products (bread, pita bread, fruit pies, carrot cake, cheese-48 cake, pizza crust, pizza, $a_w > 0.85$ and generally 0.94–0.99), where almost all bacte-49 ria, yeasts, and moulds are capable of growth (Smith et al. 2004). 50

51 When no preservative additives are added, bread and bakery products are charac-52 terised by their limited shelf-life reaching a maximum of 3–5 days at room tempera-53 ture. After this time, physical, chemical and microbiological changes are produced, 54 resulting in the loss of freshness, texture, taste and microbial spoilage (growth of 55 bacteria, yeast and mould) causing consumer's rejection (Melini and Melini 2018). 56 Those alterations can cause not only economic losses, but also threaten human 57 health. Therefore, to extend bread and bakery products shelf-life and to assure their quality and safety properties, preservation techniques such as the use of preservatives or adequate packaging materials and the application of innovative processing technologies are proposed (Mitelut et al. 2021; Qian et al. 2021).

Over time, one of the most conventional technique applied to extended freshness61quality was the use of chemical additives as was previously detailed in Chap. 4. The62bakery industry is looking for novel alternatives including the use of antioxidant and63antimicrobial compounds obtained from natural sources, new packaging technolo-64gies, application of functional coatings, etc. (Klinmalai et al. 2021; Silva et al. 2021;65Nallan Chakravartula et al. 2019a).66

Traditionally, to select a suitable packaging material for bakery products, the 67 most important properties usually sought are gases and water vapour barrier, UV 68 barrier, thermal stability, mechanical resistance (Roy and Rhim 2020). The most 69 used packaging materials to preserve bread are different types of paper, such as 70 waxed paper or the glazed imitation parchment which is strong and has grease resis-71 tance. It is usually impregnated on both sides with paraffin wax containing low 72 density polyethylene (LDPE) and other additives (Martins et al. 2021). One alterna-73 tive is LDPE bags with a strip of adhesive tape at the end to be twisted and sealed. 74 Cakes and pastry products, which are more susceptible to crushing damage, are 75 usually packed in grease-resistant paperboard bags with transparent cellophane 76 windows and wrap, such as cling film, plastic nests or aluminium foil base plates 77 and double plastic film layers. For long shelf-life products (biscuits and other), cel-78 lulose films coated with LDPE are generally used (De Pilli 2020; Galić et al. 2009) 79 or other multi-layered films such as aluminium-coated LDPE, oriented polypropyl-80 ene (OPP) or acrylic-coated OPP films which represent more effective barriers to 81 oxygen and water vapour. In the case of fresh baked stuff immediately consumed, it 82 is commonly packaged in bags made of polyolefin film, such as LDPE or polypro-83 pylene bags, normally micro-perforated to allow moisture to escape and avoid leath-84 ery consistency of the crust (Pasqualone 2019). 85

Regarding packaging methods, the application of new technologies such as vacuum packaging, nitrogen flushing, modified atmosphere, functional or active packaging (with antimicrobial activity) reduce the growth of spoilage microorganisms, extending bakery products shelf-life (Qian et al. 2021).

It is important to highlight that the plastic derived from fossil hydrocarbons comprise 46% of global plastic waste generation, producing a huge impact to the environment, which often end up in landfill sites or oceans, causing a significant pollution due to the poor infrastructure, the lack of recycling options and to the long periods of time required for their degradation (Tiseo 2021; Geyer et al. 2017). Thus, there is a wide interest in the development of new materials for substituting plastic packaging by using renewable resources to reduce polluting residues. 90

In this framework, biodegradable packaging has emerged as an innovative and 97 promising solution since they decompose after fulfilling their purpose (Chiralt et al. 98 2020; Tapia-Blácido et al. 2020). New biodegradable materials can be classified in 99 chemically synthesised polymers made from natural or petroleum-based molecules 100 (polylactic acid, polycaprolactone, polyvinyl alcohol, polyglycolic acid, polybutyl- 101 ene succinate, polybutylene adipate-co-terephthalate); directly extracted from 102

biomass (biopolymers such as cellulose, starches, chitosan, alginate, gelatine, collagen, etc.) and biosynthesized via microbial fermentation (polyhydroxyalkanoates,
bacterial cellulose) (Zhang et al. 2022; Kamarudin et al. 2022; Birania et al. 2022).
These have been used to develop new eco-friendly and active systems that could be
applied to protect or improve quality of GF bakery products. In the following sections of this chapter, a special description of biodegradable and edible matrices is
performed.

110 7.2 Edible Films, Coatings and Toppings

111 7.2.1 Edible Films and Coatings

The named edible films can be defined as standalone materials disposed as thin lay-112 ers based on eatable components (biopolymers, food additives, etc.) and are gener-113 ally used in the production of wraps, pouches, bags, capsules and casings. On the 114 contrary, coatings involve slurries that are directly applied (by deposition, adhesion 115 and drying) on the food surface and are considered as an integral part of the food 116 product. They are designed not to be removed from the food item. Usually, they can 117 be classified according to their formulation or the application method used, as will 118 be described in the methodology section. To obtain edible packaging the following 119 main technique stages must be performed: achieving the solubilisation of the bio-120 polymer in a suitable solvent to obtain the slurry (for films and coatings) or mixing 121 solid materials if it is used a thermomechanical process (without solvent addition, 122 for films), solvent evaporation when corresponding and film constitution and stabi-123 lisation. There are no differences in the material composition between coatings and 124 films but they are mainly different in relation to their thickness (Aguirre-Joya 125 et al. 2018). 126

Edible active packaging is an innovative solution due to its capability to carry preservatives compounds, which reduce the microorganism's growth and assure the safety and quality of foods extending their shelf-life (Jafarzadeh et al. 2020; Fang et al. 2017). Certain additives can be incorporated into the edible packaging formulation such as antimicrobials and antioxidants compounds (Qian et al. 2021; Dobrucka and Cierpiszewski 2014).

133 There are different methods to incorporate those compounds (Qian et al. 2021):

- (a) Direct incorporation of preservatives (thermally stable) into the packaging
 materials produced by solvent casting or extrusion technology (co-extrusion,
 extrusion or injection moulding);
- (b) Surface coating of packaging material with a film containing antimicrobial agents (essential oils derived from plants, such as cinnamon, clove, oregano, thyme, and lemon) entering the headspace through evaporation or migration to the food surface through diffusion (Mani-Lopez et al. 2018; Fang et al. 2017).
 This method illustrates an additional use of films;

7 Gluten Free Edible Films, Coatings and Toppings

- (c) Sachet/pad of antimicrobial packaging (non-volatile or volatile) are designed to hold by adsorbing or embedding the antimicrobial agents to be released inside the package continuously (Ju et al. 2019; Otoni et al. 2016). This type of packaging presents some limitations such as, the risk of accidental ingestion and additional operational steps to place them in each package;
 142
- (d) Stimuli-responsive antimicrobial packaging, in which responsive nano-carriers
 can encapsulate active compounds materials and release them on demand when
 an external stimulus (light, temperature and pressure) is applied (Qian
 et al. 2021).

Regarding the materials, as structural material or film matrix, biopolymers are gen-151 erally used. These might be: (a) hydrocolloids that includes proteins such as colla-152 gen, gelatine, mung bean protein, corn zein, whey protein, soy protein, casein and 153 others (Chen et al. 2019); and polysaccharides such as starch, cellulose and its 154 derivatives, pectin, chitosan, alginate, carrageenan, pullulan and gellan gum (Kouhi 155 et al. 2020); (b) lipid-based materials (bee wax, paraffin wax, carnauba wax, poly-156 ethylene wax, candelilla wax, rice bran wax, ouricury wax and jojoba oil); and (c) 157 blend of hydrocolloids and lipids (Jeva Jeevahan et al. 2020; Zhong et al. 2019). 158

Moreover, they have to be aligned with the consciousness growth on celiac disease and gluten intolerance, which represents one-third of the global food intolerance market, added to consumer's choice to follow a GF diet that has had an important impact in the growth of GF products in the last 10 years (Juhász et al. 2020). In this context, the use of ingredients that provide safety characteristics and additionally have favourable nutritional and mechanical profiles to be used as edible packaging materials has become a focus of interest (Vilpoux et al. 2019).

Other hydrocolloids are the most crucial ingredients in edible packaging for GF 166 baking products such as hydroxypropyl methylcellulose (HPMC) and carboxy-167 methylcellulose (CMC), present good barrier properties against oxygen and lipids 168 in film formulation (Roman et al. 2018; Anton and Artfield 2008). Likewise, 169 β -glucan, pectin, carrageenan, xanthan gum, guar gum, locust bean gum, tara gum 170 or agarose are applied in commercially available GF products (Vidaurre Ruiz et al. 171 2019). CMC, chitosan, ε -poly-L-lysine are natural polymers that present desirable 172 film-forming properties and also antimicrobial activity (Fang et al. 2017). 173

Regarding starch sources, wheat, rye, and barley are common cereals containing 174 gluten. Besides, contamination of oats with wheat, rye or barley can occur during 175 grain harvesting, transport, storage and processing (Xu et al. 2020). Previous 176 research has been extensively focused on GF edible packaging made from various 177 natural GF starches such as potato, sweet potato, cassava, rice, sorghum. Figure 7.1 178 summarises main materials used to formulate GF edible films and coatings. 179

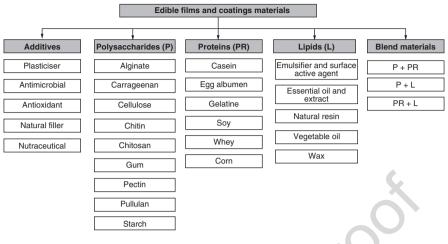


Fig. 7.1 Main materials used to formulate GF edible films and coatings

180 7.2.2 Toppings

To achieve a more attractive appearance and therefore a greater degree of accep-181 tance by consumers, toppings are incorporated into bakery products. They create a 182 decorative quality and provide additional flavour with the addition of small particles 183 (chunky, crisp or chewy bits) contributing different textures, flavours and colours, 184 widening the range of products considerably. Toppings are applied mainly for aes-185 thetic and decorative reasons, however, in some cases they positively contribute by 186 providing also desired technological effects, such as to promote the regulation of 187 water activity at the surface of the product or reduce the risk of microbial growth. 188 Additionally, the access of oxygen can be minimised, preventing the exposure to 189 light and protecting the product mechanically. By combining these positive effects, 190 it is possible to extend the shelf-life of bakery products (Tiefenbacher 2017). 191

Frequently, toppings consist of dry ingredients, such as seeds, grains, chopped 192 nuts, cereal crisps, fruit pieces (candied or dried), chocolate chips or sprinkles, 193 cocoa (nibs roasted and powder), cookie crumbles, puffed marshmallow, jelly bits, 194 flavoured bits, cheese, herbs, seasonings (spices), toffee (milk caramel) or fudge 195 bits, sugar (powder or crystals), or salt that are sprinkled superficially on the dough 196 or in the final product. When those ingredients in small discrete pieces are added 197 into filling creams, chocolate enrobing or bakery products are called inclusions 198 (Tiefenbacher 2017). 199

Moreover, wet toppings or wet ingredients can be applied on bakery products by frosting (thick and opaque mixture) covering the surface and sometimes filling the inside of cakes. Some examples are icing, mixture of confectioners powdered sugar and liquid, thin enough to be brushed on with a pastry brush or spread on pastries, rolls, and simple cakes; glaze or enrobing. Other examples are a mixture of sugar and liquid thin enough to be poured – about the consistency of thin corn syrup to coat fruit cakes, cupcakes and pieces of cake; and fillings like a thick mixture which is used between the layers of cake. It may be some of the frosting to which nuts, marshmallows or fruits are added. Whipped cream and custard mixtures are sometimes used for fillings (Tiefenbacher 2017) and natural or untreated cocoa is often used in frostings, icings, and fudge (Ortiz 2016). 210

In bakery products like muffins, toppings can be as simple as a cinnamon/sugar 211 blend or as complex as a nut streusel. Some toppings are also materials meant to 212 'sink in' to the muffin creating a 'filling' such as the addition of a sweet cream 213 cheese mixture, creating changes in texture and flavour as the consumer eats the 214 product. In bagels, toppings (seeds, finely chopped onion, or salt) are often coated 215 on the top after boiling and before baking (Ortiz 2016). Table 7.1 shows different 216 sources of ingredients used for bakery toppings. 217

Source	GF food matrix	May contain gluten trace	Gluten
Grains and alternatives	Amaranth, buckwheat, chestnut, corn (maize), millet, cornmeal, quinoa, rice, sago, sorghum, soya, tapioca, teff, uncontaminated oats	<u>8</u>	Barley, bulgur wheat, couscous, dinkel wheat, durum wheat, einkorn wheat, emmer wheat, Kamut, rye, semolina, spelt, triticale, barley, oats
Milk products, cheese and eggs	Cheese, eggs, and milk (liquid and dried) cream (single, double, whipping, clotted, soured), buttermilk, plain yoghourt	Some soft and spreadable cheeses, coffee and tea whiteners, fruit and flavoured yoghurts, and soya desserts	Yoghourt, or muesli with whole grains
Fruit, vegetables, nuts, seeds, and pulses	Fresh, frozen, canned, dried, baked, and boiled fruit and vegetables. Plain nuts, seeds, and pulses	Fruit pie fillings, processed vegetable products, deep-fried, microwave, and frozen chips, instant mash, potato waffles, roast potatoes. Roasted nuts and pulses in flavoured sauces (baked beans)	Fruit in batter and bread crumbs
Home baking	Arrowroot, artificial sweeteners, corn starch, cream of tartar, food colouring, gelatine, icing sugar, potato starch, ground almonds	Baking powder, cake decoration, marzipan, ready-to-use icing	Batter mixes, bread crumbs, stuffing mix
Confectionary, desserts, and savoury snacks	GF jelly, licorice root, seaside rock, homemade popcorn, plain rice cakes, and crackers	Chocolate, ice cream mousses, sweets, tapioca pudding. Flavoured popcorn, potato and vegetable crisps, flavoured rice cakes, and rice crackers	Made from wheat, rye, and barley, pretzels, wafers, licorice sweets, pudding made using semolina or wheat flour

 Table 7.1 Raw and partially processed gluten and GF sources used for bakery toppings

t1.1

7.3 Preparation and Characterization of Edible Films, Coatings and Toppings

Several technologies can be used to produce and study edible films, coatings and 220 toppings. Differences arise basically from food type and form of application on the 221 final product, the materials composition and properties requirements. For instance, 222 edible coatings are usually directly applied on the food product surface, while edi-223 ble film is separately produced and later used as packaging material. Various bio-224 polymers such as polysaccharides, proteins, lipids and their composites are used in 225 films and coatings formulations, most of which are GF, thus a wide spectrum of 226 properties and processing conditions are possible and need to be studied and opti-227 mised regarding the final product requirements. Besides, aiming to further extend 228 the food shelf-life span, many active films and coatings have been developed, most 229 of which contain essential oils (EOs) and other antioxidant or flavonoids rich com-230 pounds. These are usually volatile or thermosensitive compounds which limit the 231 film or coating preparation and application technologies. 232

233 7.3.1 Processing Technologies

Edible films can be manufactured by two techniques: a wet route based on a bio-234 polymer solution (usually water based) with further solvent evaporation, known as 235 casting; and a dry route in which polymers are processed in low moisture conditions 236 with the presence of plasticisers and other additives (e.g., compression moulding or 237 extrusion). The first is a batch process with some limitations, such as restricted 238 239 product size and yield, long production times, high energy demand (for solvent evaporation) and large volumes of solvent (since solids do not exceed 10-12% of 240 the suspension total mass). Therefore, it is mainly used at a laboratory scale, still 241 extensively studied for surface coating characterisations. Besides, semi-continuous 242 tape-casting and spread-coating techniques can also be used for large-scale manu-243 facture of biodegradable and edible films (Oliveira de Moraes et al. 2013). In both 244 techniques the film forming solution (or suspension) is spread over the laminated 245 material to be coated (paper for example) or directly on a non-adherent carrier-tape. 246 The film thickness is adjusted with micrometric screws that regulate the gap left by 247 the spreading blade and depends strongly on the solution's rheological behaviour 248 (Ortega et al. 2021). The suspension is later dried by heat conduction, circulation of 249 hot air (heat convection) or infrared heating, resulting in a bi-layer material (spread-250 coating) or film that can be easily removed from the tape-carrier surface. Later on, 251 and depending on the film characteristics, it can be rolled, cut, drilled, stamped or 252 laminated. Various GF edible films obtained by casting have been studied as biode-253 gradable or edible packaging for foods based on starches (Bertuzzi et al. 2007; 254 Flores et al. 2007; Müller et al. 2008; Oliveira de Moraes et al. 2013; Pérez-Vergara 255 et al. 2020; Mantovan et al. 2018; López and García 2012; Versino et al. 2016), 256 pectins (Troung and Kobayashi 2020; Nallan Chakravartula et al. 2019b; Fishman 257

et al. 2000; Sucheta et al. 2019; Gouveia et al. 2019), gelatine (Fakhouri et al. 2015; 258 Musso et al. 2017; Wang et al. 2021), chitosan and other marine derived hydrocol-259 loids (Senturk Parreidt et al. 2018a, b; López et al. 2015; Pranoto et al. 2005; Tan 260 et al. 2020; Fu et al. 2021; Morales-Jiménez et al. 2020), soy, whey and pea proteins 261 (Seung and Rhee 2004; Denavi et al. 2009; Nallan Chakravartula et al. 2019b; 262 Seydim et al. 2020; Huntrakul et al. 2020; Sun et al. 2013), among others. Drying 263 conditions are strongly dependent on the biopolymer and solvent used, temperature 264 typically ranges from room temperature to 60 °C with drying times of 5 to over 265 48 h. In casting method films thickness can be adjusted by controlling the ratio of 266 film suspension weight to plate area. Drying conditions (rate and temperature) 267 determine film characteristics (e.g., water content, crystallinity, etc.), affecting its 268 microstructure and properties (Bader and Göritz 1994) 269

Larger scale production technologies are needed to produce cost-effective, bio-270 based edible materials as food packaging. Thus, existing technology for synthetic 271 materials are also used as continuous technologies like extrusion followed by blown, 272 injection or thermo-compression (Mohammadi Nafchi et al. 2013; Flores et al. 273 2010; Garrido et al. 2016; Huntrakul et al. 2020; Fakhouri et al. 2013). Melt pro-274 cessing requires high temperatures and shear to disrupt the biopolymers' original 275 structure, plasticising it. However, additives, such as plasticisers and antioxidants, 276 are needed to thermally plasticise the polymer mix avoiding its degradation (Ortega 277 et al. 2021). 278

Thermoplastic starch (TPS) based films are obtained by melt-mixing, though it 279 is highly sensitive to moisture and present stickiness during processing (López et al. 280 2013b). Therefore, blending the starch with other polymers improves film formabil-281 ity, and mechanical, barrier and thermal properties as has been extensively reported 282 in literature (Dang and Yoksan 2015; Pelissari et al. 2012; Fakhouri et al. 2013; 283 Huntrakul et al. 2020; Ochoa-Yepes et al. 2019; Jebalia et al. 2019; Ferreira et al. 284 2021; Fishman et al. 2000; Flores et al. 2010). Extrusion can be single, twin or mul-285 tiple screwed co-rotating or counter rotating (i.e., screws rotate in the same or oppo-286 site directions respect to the feed and product flow) or a mixture of both in the case 287 of a multiple screw extruder. Temperature profiles through the extruder are impor-288 tant parameters for polymer processing that facilitate commercial processability 289 and condition the materials properties. 290

The selected film processing technology influences the film formation mecha-291 nism and therefore the resulting physical properties of the material. Casting creates 292 films stabilised largely by non-covalent interactions (hydrogen bonds, hydrophobic 293 and electrostatic interactions) while extrusion and compression moulding may 294 induce covalent interactions among the matrix compounds. Some investigation 295 comparing these technologies indicate that extruded materials result in greater 296 toughness and stiffness while casting films are more flexible, especially when cross 297 linkage was evidenced in thermoformed or extruded materials (Ciannamea et al. 298 2014; Versino et al. 2016; Ochoa-Yepes et al. 2019). 299

Active packaging technologies offer new or extra functions such as gases scavengers (O_2 , CO_2 , and ethylene), moisture regulation, flavours emission control and preservation, microorganism growth prevention, among others, that are aimed to extend the shelf-life of foods maintaining their nutritional quality and safety 303

(Kechichian et al. 2010; Jamróz and Pavel 2020; Remya et al. 2017). Active films 304 are usually prepared with the same methods previously described, though alterna-305 tives for protection and migration control are usually needed when EOs are used. 306 Encapsulation and electrospinning have been reported as successful methods to pre-307 serve and modulate the EOs antimicrobial or antioxidant properties (Scaffaro et al. 308 2020; Varghese et al. 2020; Sharifi and Pirsa 2021; Atarés and Chiralt 2016). 309 Moreover, Oriani et al. (2014) stated that a maximum of 0.1% of EOs into an edible 310 coating minimises their sensory impact; encapsulation can also be used in this regard. 311

Edible coating can be applied on a food product by four different techniques: 312 dipping, spraving, fluidized-bed, and panning (Senturk Parreidt et al. 2018a, b; 313 Suhag et al. 2020). Its efficiency is strongly dependent on the selected application 314 procedure, regarding the nature of food that should be coated, such as shape and 315 size, their surface characteristics and the desired coating thickness and the coating 316 material properties such as surface tension, density, and viscosity (Andrade et al. 317 2012). Dipping is the most widely used method to apply edible coatings on fresh 318 products, particularly in ready to eat fruits and vegetables. In general, they are sub-319 merged for 5-30 s in the formulation of edible coatings which commonly can 320 include antimicrobials and/or antioxidant to extend product's shelf-life (Suhag et al. 321 2020; Guerreiro et al. 2015; Senturk Parreidt et al. 2018a, b). It is a simple and low-322 cost technique commonly used at laboratory scale. The process consists of three 323 steps: immersion, deposition, and evaporation of solvents (Andrade et al. 2012; 324 Costa et al. 2014). Adhesion of the coating solution relies on the interaction with the 325 food surface. For instance, smooth and uniform adhesion on hydrophobic rough 326 surfaces can be very difficult due to the low surface free energy (Senturk Parreidt 327 et al. 2018a, b). Meanwhile, when the coating affinity for the product surface is 328 high, the time required will be minimal, allowing the coating solution to be applied 329 spontaneously (Park and Seo 2011). Multilayer or layer-by-layer coating techniques 330 are often needed in fresh cut products to achieve good adhesion on the highly hydro-331 philic surface. In this regard, Guerreiro et al. (2015) applied an edible coating to 332 raspberries by first immersion into alginate or pectin coating solution followed by 333 the immersion in a calcium chloride solution, allowing to form the typical egg-box 334 gel due to the chemical gelation of the hydrocolloid in presence of a bivalent cat-335 ion salt. 336

On the other hand, the spraying method is frequently used for industrial applica-337 tions, in this technique the coating solution is distributed through the formation of 338 droplets over the targeted food surface area with the help of nozzles (Suhag et al. 339 2020). One of the advantages of the spraying technique is that it needs less amount 340 of coating material to effectively coat the surface product due to the high spraying 341 pressure used (60-80 psi) (Andrade et al. 2012). Additionally, the thickness control 342 as well as the possibility of multilayer applications are also valued characteristics. 343 Spray-flow characteristics are dependent on liquid properties (density, viscosity and 344 surface tension), operating conditions (mainly flow rate and air pressure), and sys-345 tem conditions (nozzle design, spray angle, etc.). Three types of spraying techniques 346 have been used: (i) air spray atomization, where air is used for fine spraying of the 347 droplet on food products; it is a cost effective method used on food products (Valdés 348 et al. 2017), (ii) air assisted airless atomization, when high-viscosity and high-solids 349

coatings formulations are used (Peretto et al. 2017), and (iii) pressure atomization, 350 in this technique, the edible coating is applied to food products by passing it through 351 small size nozzles (Andrade et al. 2012). 352

Likewise, edible coatings can serve as adhesive for decorative toppings, which 353 are commonly included in bakery products (typically between 12% and 22% vol-354 ume percentage of the total product) to enhance their attractive sensory characteris-355 tics (Tiefenbacher 2017). In this sense, small particulate inclusions or decorative 356 toppings of different texture, flavour and colour can be used. Examples of currently 357 used toppings in bakery products are chopped nuts, cereal crisps, candied fruit or 358 toffee pieces, chocolate chips, cookie crumbles, flavoured bits, sugar or chocolate 359 sprinkles, coarse sugar or spices. These are processed by conventional food process-360 ing technologies, such as air drying, extrusion cooking, melt mixing, etc. and are 361 usually GF if cleaning protocols are carefully managed. 362

Recently, 3D printing of edible inks and pastes have been studied and commercialised, especially as food toppings. This technology expands the food processing alternatives for customizable nutrient content food products (Dankar et al. 2018). Current 3D food printing techniques include paste extrusion, ink-jet printing, powder binding deposition, sheet lamination, melt extrusion of chocolate, and bioprinting (Rowat et al. 2021).

7.3.2 Characterisation

The performance of edible films and coatings in extending food products shelf-life 370 depends on the materials light, water vapour and gas permeability and their mechan-371 ical properties to resist transport and different ambient conditions. Coating integrity 372 is a critical factor that depends on matrix flexibility, surface tension and adhesion to 373 the food product. Besides, rheological characteristics and surface adherence are of 374 particular importance for edible coating formulations, due to their direct impact on 375 surface covering and durability. Similarly, controlled or slow-release kinetics deter-376 mine its performance as active material and therefore its effectiveness on the prod-377 uct preservation. Finally, such properties need to be preserved by the film or coating 378 until the food that it wraps is consumed or, in the worst case, disposed of. 379 Consequently, all properties should be evaluated through time, ideally simulating 380 the storage conditions and average time until consumption of the packed or coated 381 food product. 382

7.3.2.1 Rheological Behaviour and Surface Properties

Rheological properties of film suspension should be tailored to fit the coating process: spraying requires low viscosity while higher viscosity is needed for immersion coating. The thin film formed on food surfaces depends on the viscosity of the coating solutions and can be well controlled with a specific spray-gun application 387

369

(Andrade et al. 2012; Suhag et al. 2020). This technology offers consistent coating
with uniform thickness, and the possibility for multilayer applications (MartínBelloso et al. 2009; Ustunol 2009). The highly viscous solution cannot be sprayed
very easily on the food products so that only dipping methods can be adapted which
results in the higher thickness of the coating material on the surface of food products (Andrade et al. 2012).

Likewise, considering the scale-up of the coating or film production the rheological behaviour of filmogenic solutions or suspensions is critical and conditions the processing operations involved, such as the pumping machine capacity. In general, starch-based filmogenic suspensions, which are widely used in coating formulations, exhibit a pseudoplastic behaviour while other formulations based on polysaccharides such as cellulose derivatives (like methylcellulose (MC) or HPMC) presented a Newtonian behaviour at low concentrations (1%).

On the other hand, coating formulation and mainly, its surface tension as well as 401 food product characteristics determine the adhesion to food substrate and successful 402 coating application. Products with smooth and soft surfaces (such as some vegeta-403 bles like tomato) require formulations with low surface tensions to ensure coating 404 adherence and uniformity. For products with irregular and rough surfaces (such as 405 strawberries) the formulation must include plasticisers to prevent the appearance of 406 cracks or pores in the coating. An alternative to enhance coating adhesion is the 407 addition of surfactants and lipids to filmogenic suspensions, reducing their surface 408 tension. The more similar the surface tensions of the product surface and that of the 409 coating formulation, the greater the compatibility and the better the adhesion of the 410 coating. Coating compatibility also is related to the hydrophilic-hydrophobic char-411 acteristic of both the surface and the formulation, which could be evaluated through 412 contact angle measurements (Ramírez et al. 2012; Rossi et al. 2019). 413

Film or coating superficial appearance depends on their formulation, since matri-414 ces without plasticisers are brittle and rigid due to the strong interactions between 415 the polymer chains that can also lead to aggregate formation. Besides, these struc-416 tures are incompatible with irregular product surfaces leading to cracks and pores 417 and conditioning coating integrity. The presence of these defects also limits barrier 418 and mechanical properties of films and coatings. Plasticiser addition in the formula-419 tions can solve this problem by improving the coating flexibility (García et al. 1998; 420 López et al. 2010). Likewise, the plasticiser/polymer ratio should be optimised 421 since high plasticiser concentrations reduce barrier properties and may cause segre-422 gation from the matrix. In starch-based formulations glycerol or sorbitol are com-423 monly used as plasticisers in concentrations between 5 and 50 g/L, depending on 424 starch concentration (García et al. 2009; Versino et al. 2016). 425

426 7.3.2.2 Mechanical Properties

Uniaxial tensile tests are usually performed to assess the film's mechanical resistance. From these tests, strain-stress curves are obtained and the Elastic Young
Modulus, maximum tensile strength and elongation at break are assessed. The

mechanical properties depend on additives-matrix interactions that can also be strongly affected by physical, chemical, and environmental conditions, which influence the material stability and flexibility. Plasticisers are often needed to enhance the materials flexibility, especially in starch-based films and coatings (Versino et al. 2016). The addition of lipids, including essential oils has also been reported to increase extensibility of biopolymers-based materials (Bof et al. 2021; García et al. 2001; Jamróz and Pavel 2020). 430

On the other hand, dynamic mechanical analysis is a useful tool to study relax-437 ation processes associated with glass transition temperatures (Tg). The Tg corre-438 sponds to the temperature at which $tan\delta$ and E" (loss moduli) curves presented a 439 maximum peak while E' (storage moduli) curve shows an abrupt fall. It has been 440 widely used especially in starch-based formulations. The knowledge of the Tg is 441 crucial since it is strongly related to mechanical film properties and also, modifica-442 tions on both film formulations and storage conditioning may affect Tg and conse-443 quently the mechanical resistance and flexibility of developed films. In this sense, 444 the inclusion of plasticisers decreases the intermolecular forces between polymer 445 chains and consequently reduces Tg values. Thus, being water the most ubiquitous 446 plasticiser of hydrophilic films and coatings, film water content and relative humid-447 ity of storage should be carefully controlled and monitored. 448

7.3.2.3 Barrier Properties

Water and gas barrier properties are key for food products preservation. Gas perme-450 ability is usually tested on films or the coating standing by its own (also as a film). 451 Water vapour permeability (WVP) is often determined gravimetrically according 452 to American Society for Testing Materials (ASTM) standard test method, ASTM 453 E96/E96M or a modification of this norm, thus various relative humidity conditions 454 have been reported (Huntrakul et al. 2020; Nallan Chakravartula et al. 2019b; Musso 455 et al. 2017; Ochoa-Yepes et al. 2019; Fakhouri et al. 2015). The WVP indicates the 456 ability of the film or coating to protect the food from moisture migration from or 457 towards the product. For instance, to prevent pastry from drying which would not be 458 desirable for texture acceptance by the costumer or to prevent an increase in mois-459 ture content and water activity, which can promote mould growth and a faster deg-460 radation of the product. Unplasticised films often yield significantly higher WVP 461 values than plasticised ones, due to the presence of pores and cracks (Versino et al. 462 2016). Even though plasticisers used in edible films and coatings are generally 463 hydrophilic, its inclusion generates structural modifications on the biopolymer net-464 work leading to a less ordered and compact structure. In general, starch-based films 465 exhibit lower WVP values compared to both protein films and other polysaccharide-466 based films (García et al. 2001, 2004; Versino and García 2014; Rivero et al. 2010; 467 Tavassoli-Kafrani et al. 2016). 468

Considering that hydrocolloid-based films are very sensitive to relative humidity, 469 physicochemical characterisation generally includes water sorption isotherms 470 determination, useful for the estimation of film stability under different ambient 471

472 conditions. In general, sorption isotherms are obtained, being experimental data
473 satisfactorily fitted by GAB model, estimating the monolayer water content values
474 (Mali et al. 2002; Müller et al. 2009).

Regarding film gas permeability most methods use the same principle: a mea-475 surement of the gas transmission rate through an edible film located between two 476 compartments. One side of the film is exposed to the gas being studied, and a detec-477 tor is placed in the other compartment, which is initially free of the permeated 478 compound (Sánchez-Tamayo et al. 2020). These methods were developed for syn-479 thetic materials, described in the ASTM procedures, and adapted to edible films and 480 coatings (ASTM D3985, F1927). The manometric and the volumetric methods 481 measure the difference in absolute pressure, and the continuous-flow or isostatic 482 method uses a stream flux of the gas to be measured on one side of the film and a 483 nitrogen stream on the other side to carry the gas to the analyser. Coulometric sen-484 sors, infrared sensors, or gas chromatography may be used for gas concentrations 485 analysis. In the case of O₂ permeability the use of specific equipment, the Mocon 486 Oxtran 2/21, is widely used due to its precision and simplicity. Although for CO₂, 487 N_2 and ethylene the determination requires the use of an especially designed cell 488 and in general chromatography measurements. It is well known that polysaccha-489 rides films such as starch-based ones exhibit a highly selective gas permeability 490 ratio (CO_2/O_2) compared with conventional synthetic materials. The modified atmo-491 sphere created by the coating generates a physical capture of CO₂ inside the fruit or 492 vegetable and partial sealing of the pores, reducing the gas exchange and gas trans-493 fer rates; and this is monitored through respiration activity measurements. This 494 selective gas permeability can be attributed to a higher solubility of CO_2 than O_2 in 495 the film matrix. Development of composite edible films and coatings with selective 496 gas permeability could be promising for controlling respiratory exchange and 497 improving the conservation of food products. 498

499 7.3.2.4 Microstructure

The materials microstructure is generally studied by microscopy techniques, mainly scanning electron microscopy (SEM) or atomic force microscopy (AFM). Compact and homogeneous matrix of films is an indicator of structural integrity and, consequently, good mechanical properties are expected. To evaluate this, SEM observations of both the surface and the cross-sections are carried out. Figure 7.2 shows the microstructure of an edible coating based on HPMC applied on pumping vegetable tissue.

Topography and roughness of starch films surface can be evaluated by AFM. In
the case of nanocomposite films transmission electron microscopy is also conducted
to evaluate the nanometric size particles included in the matrices.

Interactions among film-formulation components as well as their compatibility are commonly studied by Fourier transform infrared spectroscopy, being this technique combined with chemometric analysis. In order to evaluate the crystallinity degree of edible films and coatings X-ray diffraction is commonly performed. Film crystallinity depends on the biopolymer source and plasticisers, film drying

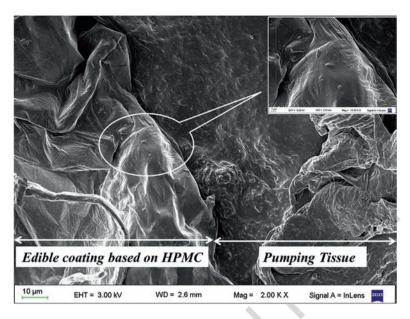


Fig. 7.2 Microstructure of an edible coating based on HPMC supporting *L. casei* cells and applied on pumping vegetable tissue

conditions and their final moisture content (García et al. 2009). Evolution of film matrix crystalline structure during storage can also be evaluated by differential scanning calorimetry, which allows to determine the Tg, being in this case the modulated technique more appropriate since it discriminates between the total heat flux, and the reversible and non-reversible contribution.

7.3.3 Characterisation After Application on Food or Simulated 520 Food Systems 521

In the case of edible coatings and toppings a complete sensorial analysis is manda-522 tory, showing that the coatings did not influence consumer acceptance, especially 523 when taste is evaluated. In general, sensory panels are performed with both trained 524 and untrained panellists on the basis of a hedonic scale. The selection of panellists 525 includes men and women covering a wide age range to simulate the spectrum of 526 potential consumers of the product being evaluated. In this sense, adhesion proper-527 ties evaluation should be taken into account, especially for products destined for 528 distant markets or long term storage, since it conditions the coatings' performance 529 (Ncama et al. 2018). 530

Likewise, the microbiological tests should be conducted to assure the product 531 safety. In this sense, the absence of microorganisms that cause foodborne illness as well as the counts of aerobic mesophilic and psychrophilic bacteria and mould and 533

veasts are commonly evaluated following standard procedures. This is of particular 534 interest when active films and coatings are applied to extend food shelf-life. In this 535 sense, López et al. (2013a, b) developed films based on blends of native and acety-536 lated corn starches containing glycerol as plasticiser, potassium sorbate and citric 537 acid as antimicrobial agents. The developed active starch films were able to inhibit 538 Candida spp., Penicillium spp., S. aureus and Salmonella spp. growth, which are 539 responsible for some foodborne diseases. These active films were effective to extend 540 cheese shelf-life from 14 to 17 days at 4 °C. Besides, sorbate controlled release 541 from the polymeric matrix was studied and diffusion coefficients in aqueous and 542 semisolid media were also determined (López et al. 2013a). Rivero et al. (2013) also 543 studied the controlled release of propionic acid from chitosan films to dough. 544

7.4 Application as Shelf-Life Improver and as Carriers of Bioactive Compounds, Vitamins or Minerals

Actual challenges faced by the food industry involve the extension of food shelf-life 547 without impairing the nutritious properties of food products and through the use of 548 sustainable techniques (Díaz-Montes and Castro-Muñoz 2021). The food products 549 available in the market for celiac population, particularly bakery stuffs, might have 550 reduced nutritional and organoleptic quality due to the necessary change in formu-551 lation. Such nutrient deficiencies promote an unbalanced diet for GF consumers, 552 especially regarding fibre, bioactive compounds, vitamins and minerals (Capriles 553 et al. 2016). As a consequence, important efforts have been focused in diversifying 554 and improving the offer of GF items with better nutritional characteristics (Genevois 555 et al. 2020). 556

With the purpose of contributing to the development of healthy foods with long 557 shelf-life, many emergent technologies have been explored in the last years. As was 558 previously mentioned, edible films and coatings are one of the hurdles that have 559 been explored. They are produced using polysaccharides, proteins, and lipids, and 560 besides, they can support antioxidants, antimicrobials, vitamins, probiotics, miner-561 als, flavouring, and colouring agents. The use of antioxidants, antimicrobials and 562 other nutraceuticals supported in these edibles layers allows to control their loca-563 tion, their release and the matrix also exerts a physical protection that slows down 564 their destruction. As a consequence, films and coatings can help to improve shelf-565 life and/or food quality (Gerschenson et al. 2018; Alzate et al. 2021). In particular, 566 in the case of considering the probability of consumer's gluten intolerances, the use 567 of edibles films must be adapted to this restriction, ensuring the absence of harmful 568 proteins in the formulation. In this case, alternative proteins, starches, rice by-569 products or different hydrocolloids like alginate that have algal origin, can be used 570 for films and coatings production (Senturk Parreidt et al. 2018a, b). In addition, 571 edible films and coatings can contribute to enhance the nutritional properties of 572 foods through the support of vitamins, probiotics, and other compounds. 573

In the next items and Table 7.2, there will be described some matrices mainly based on polysaccharides that were developed in the last years and that can be used

t2.1	Table 7.2 Di	Table 7.2Different GF edible films and coatings used in bakery products	oatings used in bake	ry products			
t2.2 t2.3	Application	GF matrix	Active agent	Bakery stuff	Main effects	Shelf-life (t, T)	References
t2.4 t2.5	Coating	Groundnut oil	TN	GF flatbread	Slowing of staling rate	8 days 2 °C	Patil et al. (2019)
t2.6 t2.7	Coating	Corn starch, MC, soybean oil, glycerol	.I.N	Crackers	Reduction of the hydration kinetic in a high a _w environment	20 days 25 °C	Bravin et al. (2006)
t2.8 t2.9 t2.10	Coating	Egg protein, hydrocolloid component, vegetable oil	N.I.	Sweet baked goods	Reduction of water migration rate between components	85 days 25 °C	De Pilli (2020)
t2.11 t2.12	Coating	Lepidium sativum seed gum	.I.N	Sorghum GF bread	Improvement of crust and overall quality in comparison with normal glazes	N.R.	Sahraiyan et al. (2020)
t2.13 t2.14	Coating	Okra mucilage	N.I.	Soft dough biscuits	Improvement of the crispiness	6 days 25 °C	Senanayake et al. (2021)
t2.15 t2.16 t2.17	Coating	Candelilla wax (in sunflower oil), beeswax (in sunflower oil), HPMC	N.I.	Bread	Reductions of bread weight loss and the crumb firmness	14 days 25 °C	Chen et al. (2021)
t2.18 t2.19	Coating	Pectin, alginate and whey protein	N.I.	Mini-buns	Reduction of moisture loss and textural changes	N.R.	Nallan Chakravartula et al. (2019c)
t2.20 t2.21 t2.22 t2.23	Coating	Sodium alginate, whey, glycerol	Lactic acid bacteria	Bread	Reduction of mesophilic and facultative aerobic bacteria count protection against mycelium fungi of genera <i>Aspergillus</i> and <i>Penicillium</i>	5 days 28 °C	Gregirchak et al. (2020)
t2.24 t2.25 t2.26	Coating	Egg white protein	Carvacrol, thymol, <i>trans</i> - cinnamaldehyde	Bread	High antifungal efficacy of coatings supporting thymol and carvacrol nanocomplexes	7 days 25 °C	Deseta et al. (2021)
t2.27 t2.28 t2.29	Coating	Cassava starch, inverted sugar, sucrose	Soluble coffee, cocoa powder or propolis extract	Muffins	High antimicrobial action against mould and yeast maintenance of the global quality	87 days 25 °C	de Oliveira Melo Naponucena et al. (2019)

(continued)

				Bakery		Shelf-life	
	Application	GF matrix	Active agent	stuff	Main effects	(t, T)	References
t2.30 t2.31 t2.32 t2.33	Coating	Sodium alginate or blends sodium alginate and whey protein concentrate	Lactobacillus rhannosus GG	Pan bread	Improvement of the viability of <i>L</i> . <i>rhamnosus</i> GG No modification of textural, flavour and thermophysical properties of crust	7 days 25 °C	Soukoulis et al. (2014)
t2.34 t2.35	Coating	Potato starch, inverted sugar, sucrose	Potassium sorbate and/or citric acid	Mini panettone	Inhibition of mould/yeast grown	48 days 35 °C	Ferreira Saraiva et al. (2016)
t2.36 t2.37	Coating	Sodium alginate, whey protein, glycerol	L. brevis	GF cookies	Improvement nutritional quality without modifying physical and sensorial properties	30 days 35 °C	Chávez et al. (2022)
t2.38 t2.39 t2.40 t2.41	Coating	Mung bean starch, guar gum, sunflower seed oil	Grapefruit seed extract	Non- glutinous rice flour cakes	Improvement stability by retarding starch retrogradation and inhibiting <i>B. cereus</i> and <i>P. citrinum</i> growth	N.R.	Lee et al. (2020)
t2.42 t2.43 t2.44 t2.45	Film	Starch-based (tapioca, potato, corm), cellulose nanofiber.	TN	Muffin	Good performance as a liner to hold the batter and protect the muffins from sticking to the pan during baking. Film could be consumed	N.R.	Shih and Zhao (2021)
t2.46 t2.47 t2.48 t2.49	Film	MC, polyethylene glycol	Clove bud or oregano essential oil (Tween 80 addition)	Bread slices	Reduction of yeasts and moulds counts	15 days 25 °C	Otoni et al. (2014)
t2.50 t2.51 t2.52 t2.53	Film	Cellulose-derivative polymer	Cinnamaldehyde	Pastry dough (P d) and bread (B)	Inhibition of aerobic mesophilic, yeast and mould growth.	P d: 30 days 8 °C. B: 12 days 23 °C	P d: 30 days Lopes et al. (2013) 8 °C. B: 12 days 23 °C
t2.54 t2.55	Film	Chitosan	Apricot kernel essential oil	Bread	Inhibition of fungal growth	10 days 25 °C	Priyadarshi et al. (2018)
t2.56 t2.57	Film and coating	Chitosan-carboxymethyl cellulose-oleic acid	Zinc oxide nanoparticles	Sliced bread	Reduction of fungal growth and retard the staling rate	35 days 25 °C	Noshirvani et al. (2017)
t2.58	NI not incorp	NI not incorporated, NR not reported					

 Table 7.2 (continued)

in the development of food products that need GF formulation. Many advances have 576 been reported in relation to improve global quality of diverse traditional bakery 577 products based on wheat using active edible films and coatings through the exten-578 sion of microbial stability during storage (de Oliveira et al. 2019; Oian et al. 2021; 579 Axel et al. 2017; Noshirvani et al. 2017), the maintenance of the crispiness and 580 textural characteristic by reducing staling (Senanayake et al. 2021; Nallan 581 Chakravartula et al. 2019c; Chen et al. 2021) and the increase of nutritional or func-582 tional properties, through probiotics or prebiotics incorporation (Zoghi et al. 2020; 583 Fernándes et al. 2020). On the contrary, few studies were performed combining GF 584 edible films or coatings on GF bakery products. Recently, Chavez et al. (2022) cov-585 ered GF cookies with an edible coating based on sodium alginate (1% w/w), whey 586 of milk protein (2% w/w) and glycerol (5% w/w) water solution supporting probiot-587 ics, L. brevis strain, improving functional value without affecting sensorial and 588 physical properties. Another research (Lee et al. 2020), reported that cakes made 589 with non-glutinous rice flour and coated with mung bean starch and guar gum slurry 590 containing sunflower seed oil, decreased the hardness by 29% and the crystallisa-591 tion rate by 24% compared with those of uncoated samples along storage at 592 25 °C. The authors concluded that edible coating retard the starch retrogradation in 593 coated cakes. In the same study, the addition of 0.8% (w/w) grapefruit seed extract 594 to coating exerted an effective antimicrobial activity against B. cereus and P. citri-595 num during rice cake storage. Similarly, Patil et al. (2019) demonstrated that staling 596 rate was successfully retarded with the help of a groundnut oil coating on the sur-597 face of the GF flatbread during storage at 4 °C. Finally, Sahraiyan et al. (2020) 598 analysed the effects of traditional glazes (oil, cheese powder, xanthan gum) on the 599 physicochemical and sensory parameters of sorghum GF bread and were compared 600 with Lepidium sativum seed gum coating. Results showed that application of novel 601 glaze was better than the usual glazes to improve the crust and overall quality of 602 GF bread. 603

7.4.1 Films and Coatings and WVP Control

Bravin et al. (2006) studied the development of emulsified edible films constituted 605 by corn starch, MC and soybean oil. The techniques explored for deposition of the 606 film forming solution were spreading or spraying. The presence of oil depressed the 607 WVP. With this formulation, both techniques produced, in general, similar 608 WVP. Atomization pressure of 2 bar and film thickness of 30 µm were identified as 609 optimum for the application of edible coating to bakery products. Edible coating of 610 previously described characteristics was applied for controlling moisture uptake in 611 crackers submitted to RH of 65-85%. Crackers coated with this formulation showed 612 a longer shelf-life due to the control of moisture transfer exerted by the film, con-613 firming its potential for slowing the hydration rate in the RH range studied. In 614 another study, Shih et al. (2011) developed edible films using various ratios of 615

pullulan and rice wax up to 46.4% (w/w). Authors reported that water vapour barrier 616 increased and hydration capacity decreased with a higher addition of rice wax help-617 ing to lengthen the shelf-life of food products. Several bio-polymeric matrices were 618 analysed by Cando et al. (2017), who studied the production by casting technique of 619 biodegradable films based on dispersions of a mix (50:50) of cassava, rice or potato 620 starch and bovine gelatine. The total solid concentration of the dispersions was 2% 621 (w/w) and glycerol was used as plasticiser. The films with cassava starch showed the 622 lowest WVP. Nisar et al. (2018) developed antimicrobial films based on citrus pec-623 tin with the incorporation of different levels of clove bud essential oil. The inclusion 624 of oil diminished the WVP and increased the deformability and heat stability of 625 the films. 626

7.4.2 Films and Coatings. Weight Loss and Antimicrobial/ Antioxidant Effect

Sadygova and Kozlov (2015) developed an edible foam for coating bakery products, 629 with gram flour (5-15%), ashberry powder (5-10%), table salt (1-3%) and water up 630 to 100%. The film was produced through drying at 55-65 °C for reducing the mois-631 ture content to 5-10%. This coating lengthens the shelf-life of bakery products 632 helping to decrease microbiological count. Ferreira Saraiva et al. (2016) studied the 633 application of edible coatings based on potato starch (46 g/kg), inverted sugar (14 g/ 634 kg) and sucrose (7 g/kg), with the purpose of reducing the preservatives added to 635 mini panettones. The preservatives added to coating formulations were potassium 636 sorbate (1 g/kg), citric acid (10 g/kg) and both additives (1 g/kg sorbate and 10 g/kg 637 citric acid or 0.5 g/kg sorbate and 5 g/kg citric acid). Panettones without coating and 638 additives showed the growth of mould and yeasts after 24 days of storage. On the 639 contrary, the presence of films with both additives showed fungal growth only after 640 40 days. The authors concluded that the use as coatings of films with additives in 641 concentrations lower than those normally used for these foods, increased their 642 shelf-life. 643

Regarding active films added with natural preservatives, Nisar et al. (2018) 644 showed that antimicrobial films based on citrus pectin with the incorporation of dif-645 ferent levels of clove bud essential oil were effective against S. aureus, E. coli and 646 L. monocytogenes when evaluated through the diffusion tests, showing a diameter 647 increase from 18.50 to 30.27 mm, 12.53 to 21.20 mm and 14.67 to 26.43 mm, 648 respectively, with the increase of oil concentration from 0.5% to 1.5%. The most 649 sensible bacteria was S. aureus. According to Alzate et al. (2017) the addition of 650 carvacrol, the main component of the oregano essential oil, to edible film formula-651 tion based on cassava starch and HPMC, highly improved the antimicrobial barrier 652 action against Z. bailii, L. plantarum, and P. fluorescens in comparison with films 653 containing only potassium sorbate. Recently, Mahcene et al. (2020) studied the use 654 of sodium alginate to constitute films incorporated with essential oils of some 655 medicinal plants (R. officinalis L, A. herba alba Asso, O. basilicum L and M. 656

pulegium L). The films showed a strong antibacterial effect against Staphylococcus 657 aureus (ATCC 43300), Escherichia coli (ATCC 25922), Salmonella enterica (ATCC 658 14028), Enterococcus faecium (ATCC 35667), Klebsiella pneumoniae (ATCC 659 70060) and Enterococcus faecalis (ATCC 29212). The antioxidant capacity, 660 reported as DPPH inhibition %, of the different films showed values of 4% for 661 M. pulegium's oil film to 23% for O. basilicum oil film in comparison with the con-662 trol film which revealed no radical scavenging activity. The authors attributed these 663 low values to the destruction of the active principles during film production and/or 664 to the reaction of active principles with alginate. 665

Utama-ang et al. (2021) studied the microwave-assisted extraction (400 W, 1 min) 666 of phenolic compounds from dried ginger and its incorporation (3.2% w/v) in a rice-667 based edible film. This edible film showed antioxidant activity due to the presence of 668 6-gingerol, 6-shogaol, paradol and zingerone. The incorporation to the formulation 669 of 3.2% (w/v) ginger extract determined that the film showed antimicrobial activity 670 against S. mutans DMST 18777. Yerramathi et al. (2021) developed a film based on 671 alginate crosslinked with ferulic acid, for extending the shelf-life of different foods. 672 The edible films developed showed an increasing antioxidant effect with the concen-673 tration of ferulic acid. Table 7.2 summarises the different GF active edible packaging 674 used to improve the quality or extend the shelf-life of bakery products. 675

The effectiveness of the methods described in Table 7.2 depend on the food product characteristics (shape and size), the composition of the edible films and coatings, the physical properties (surface tension, density, and viscosity) (Andrade et al. 2012; Debeaufort and Voilley 2009), the processing method used and the sensory compatibility with the food (Restrepo et al. 2018).

7.4.3 Films and Coatings for Supporting Micronutrients/ Probiotics

In particular, edible films have been proposed for the support and protection of pro-683 biotics. Probiotics are defined as nonpathogenic living microorganisms that have 684 beneficial effects on host health and disease prevention when administered in ade-685 quate amounts (Kunes and Kvetina 2016). Soukoulis et al. (2014) studied the appli-686 cation of film forming solutions based on 1% w/w sodium alginate or in blends of 687 0.5% w/w sodium alginate and 2% whey protein concentrate, to bread. These solu-688 tions contained the probiotic Lactobacillus rhamnosus GG, and after its application 689 on the surface of the bread, a drying step (60 °C, 10 min or 180 °C, 2 min) gave 690 origin to a film that did not affect visually the crust and did not affect bread staling. 691 The presence of whey protein concentrate improved the viability of the lactobacilli 692 during drying and also during bread storage. The authors reported that films based 693 exclusively on sodium alginate increased the viability of lactobacilli under simu-694 lated gastro-intestinal and concluded that a slice of bread (3-40 g) can provide 695 approximately 7 log CFU of lactobacilli, contributing to the development of more 696 healthy foods. In another study, Altamirano-Fortoul et al. (2012) developed 697

functional bread combining *L. acidophilus* encapsulation and starch based edible coating. The results demonstrated the ability of the coating to protect the probiotics during baking, allowing their survival. However, significant physicochemical changes were observed on the crust. Many other GF bio-polymeric matrices have been assays as carriers of probiotics to broaden the offer of functional foods and to improve probiotic viability during processing and storage (Zoghi et al. 2020).

Likewise, edible coatings may also act as carriers of nutrients. Caseins and whey 704 proteins based matrices have been used to control the release or support of calcium, 705 iron, vitamin D, A, E and folic acid in several food stuffs (Daniloski et al. 2021; Mei 706 and Zhao 2003). Genevois et al. (2016) developed a refrigerated ready-to-eat food 707 based on pumpkin and fortified with iron and ascorbic acid supported in a coating 708 matrix, which helped to enhance their bioaccessibility at *in vitro* simulated lumen 709 conditions. Edible folic acid-nanolaminates were obtained by the laver-by-laver 710 technique using alginate and chitosan biopolymers while the vitamin was incorpo-711 rated by post-diffusion (Acevedo-Fani et al. 2018). These systems were able to pro-712 tect folic acid from degradation by UV irradiation whereas the release profiles were 713 affected by pH level, being faster in simulated conditions of the small intestine 714 (pH 7). In another work, Behjati and Yazdanpanah (2021) elaborated a nano-715 emulsion vitamin D₃ fortified edible film based on guince seed gum which pro-716 moted high stability of the vitamin. Therefore, the authors concluded that fortified 717 edible films can be used in ready-to-eat food products to improve vitamin intake. 718 Moreover, Vitamin C (L-(+)-ascorbic acid), with important antioxidant activity, 719 could be retained and protected when was supported in an alginate edible coating 720 (De'Nobili et al. 2017) or a high methoxyl pectin edible film (Pérez et al. 2009), 721 constituting an effective strategy for reduce its degradation. The authors demon-722 strated that the immobilisation of water in the film network modulated vitamin loss. 723

724 **7.5 Conclusions**

GF edible films, coatings and toppings applied to bakery products provide protec-725 tion against external factors that can alter their quality. Toppings are essential for 726 flavour and determine the appearance of the product, providing certain short-term 727 barrier protection. On the other hand, edible coatings and films are a great barrier 728 that focuses on extending long-term protection and provide a way to incorporate 729 active components, such as antimicrobials or antioxidants to prevent microbial and 730 oxidative impairment. In addition, edible films and coatings also can improve the 731 nutritional quality of GF bakeries, since they can support vitamins, minerals, probi-732 otics, etc. Therefore, actual GF available products can become functional foods 733 offering to consumers with disorders related to gluten intake, higher quality and 734 more diversified foods. Thus, improved GF bakery could help to avoid the risk of a 735 nutritional deficiency and to exert a positive effect on health beyond basic nutrition. 736 Future research must be focused on studying the effects of different GF formula-737 tions and processing of edible films, coatings and toppings on the physicochemical, 738

sensorial and nutritional properties of GF bakery products. In addition, the influence
of the application methodology on such characteristics should be further explored in
order to help the achievement of a greater insertion of these technologies in the food
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industry while contributing to the generation of healthy foods.

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