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Wheat gluten–based coatings and films: Preparation, properties, and applications

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Abstract
Effective food packaging that can protect foodstuffs from physical, chemical, and biological damage and maintain freshness and quality is essential to the food industry. Wheat gluten shows promise as food packaging materials due to its edibility, biodegradability, wide availability, low cost, film-forming potential, and high resistance to oxygen. The low mechanical properties and poor water permeability of wheat gluten coatings and films limit their wide applications; however, some inferior properties can be improved through various solutions. This work presents a comprehensive review about wheat gluten–based coatings and films, including their formulation, processing methods, properties, functions, and applications. The mechanical and water resistance properties of coatings and films can be reinforced through wheat gluten modification, combinations of different processing methods, and the incorporation of reinforcing macromolecules, antioxidants, and nanofillers. Antioxidants and antimicrobial agents added to wheat gluten can inhibit microbial growth on foodstuffs, maintain food quality, and extend shelf life. Performances of wheat gluten–based coatings and films can be further improved to expand their applications in food packaging. Current research gaps are identified. Future research is needed to examine the optimal formulation and processing of wheat gluten–based coatings and films and their performance.

KEYWORDS
antimicrobial effect, edible coatings and films, mechanical property, packaging materials, water resistance, wheat gluten

1 | INTRODUCTION

Effective food packaging is critical to the food industry. The packaging can protect foodstuffs from physical, chemical, and biological contamination; maintain their safety, stability, and quality; and extend their shelf life during transportation, distribution, and storage (Kalpana et al., 2019). Food packaging conventionally uses petroleum-based polymers. In recent decades, biodegradable polymers are gaining increasing popularity to reduce environmental pollution. Synthetic polymers, such as polylactic acid, polybutylene succinate, and polyvinyl chloride, have been extensively investigated and applied in food packaging, as are microbial polymers produced by natural or genetically modified microorganisms such as polyhydroxyalkanoates and poly β-hydroxybutyrate (Assad et al., 2020; Kumar et al., 2021; Xu et al., 2019; 2020). Unlike traditional food packaging materials, edible coatings or films are thin layers coated or wrapped on the surface of foodstuffs, aimed to prevent the movement of moisture, gas, and solute between foodstuffs and the environment (Maan et al., 2021). Coatings are the edible thin layers coated on the foodstuffs, whereas films are usually molded into sheets and then wrapped on foodstuffs. Edible coatings or films can also be utilized in the form of soft gel capsules, microcapsules, soluble strips, and flexible pouches (Han, 2014). Edible coatings and films are required to be biocompatible and are commonly made from biopolymers, such as polysaccharides (starch, cellulose, pectin, gum, chitin, and dextran), proteins (wheat gluten, corn zein, whey protein, and soy protein), lipids (waxes, shellac resins, fatty acids, and acylglycerols), or their combinations (Amin et al., 2021; Mohamed et al., 2020).

Protein-based edible coatings and films have attracted increasing attention due to their low cost, wide availability, high nutritional profile, and improved gas barrier and mechanical properties compared to polysaccharides—or lipids-based coatings and films (Bourtoom, 2008; Zhang & Mittal, 2010). In addition, proteins' amphiphilic nature, electrostatic charges, and conformational denaturation allow them to undergo physical, chemical, and enzymatic treatments to make more desirable coatings and films (Khashayary & Aarabi, 2021). Proteins have also been reported in the literature for film and coating uses. Corn protein contains nearly the same amount of hydrophobic and hydrophilic amino acids (Rajpurohit & Li, 2022). Corn zein, the major protein in corn, is an amphiphilic protein and insoluble in water (Ge et al., 2022), whereas it is soluble in 70%–85% aqueous ethanol. Corn zein films possess good water barrier properties, but poor mechanical properties (Sun et al., 2018). Due to the high molecular weight (HMW) of soy protein and with a hydrophobic/hydrophilic amino acid ratio of 0.64 (Rajpurohit & Li, 2022), soy protein film has been reported to have low mechanical strength, high brittleness, and be sensitive to moisture (Jin et al., 2020). Whey proteins are naturally composed of β-lactoglobulin, α-lactalbumin, and bovine serum albumin, which exist as globular proteins. It has been reported that whey protein can form transparent and flavorless films; however, the functionality of film is highly dependent on the whey protein composition, processing temperature, and pH (Fematt-flores et al., 2022). Another major type of protein, wheat gluten, has been widely used to make coatings and films because it is naturally abundant, biodegradable, cheap, edible, transparent, and oxygen-resistant (Kaushik et al., 2015; Mojumdar et al., 2011). It has been used in food packaging for strawberry and cheese to improve the quality of foodstuffs during storage (Tanada-Palmu & Grosso, 2005; Türe et al., 2010). In addition, antioxidants and/or nanofillers have been incorporated into wheat gluten–based films to improve their antioxidant, flavonoid, and montmorillonite nanoclay components (Cortés-Triviño & Martínez, 2018; El-Wakil et al., 2015; Salgado et al., 2015).

Lagrain et al. (2010) reviewed how processing methods (e.g., dry processing and wet processing) affect the thermal, mechanical, and barrier properties of wheat gluten-based films. Mojumdar et al. (2011) reviewed the properties and potential applications of solvent cast wheat gluten-based films. Many additional studies on gluten films and coatings have been published during the last decade. To our knowledge, there is no up-to-date and comprehensive review about wheat gluten-based coatings and films' formulation and processing methods, properties, antimicrobial functions, and applications. Therefore, in this review, we focus on the formulation of wheat gluten–based coatings and films using different methods, such as solvent casting, compression, molding, extrusion, electrospinning, or a combination of these methods. We also review the characteristics of the resultant coatings and films, including their thermal, mechanical, and barrier properties, their antimicrobial functions, and their applications in food packaging. Current research limitations and future promising topics are also identified.

2 | GLUTEN COMPOSITION AND PHYSICOCHEMICAL PROPERTIES

Wheat gluten is known as the storage protein in the wheat grain endosperm and is responsible for dough formation and structural integrity in wheat-based food products. It is the mass residue when wheat starch is washed away from wheat flour (Cook & Rose, 1934). Wheat gluten is composed of two distinct protein fractions: gliadin and glutenin (Cook & Rose, 1934). Gliadins are soluble in 70% aqueous alcohol, but glutenin is insoluble in most common solvents due to its large size and cross-linked polymeric structure. Gliadins are composed of single-chain proteins, which contain α-gliadins, β-gliadins, γ-gliadins, and ω−gliadins. Glutenin proteins are composed of polypeptide chains linked by interchain disulfide bonds (S–S). Glutenin proteins can be separated by sodium dodecyl sulphate polyacrylamide gel electrophoresis (SDS-PAGE) into two groups under reduced conditions: HMW glutenin subunits and low molecular weight (LMW) glutenin subunits (Day et al., 2006; Lagrain et al., 2010). LMW subunits can be further subclassified into B-type LMW subunits and C-type and D-type LMW subunits (Shewry, 2019, 2002). According to Shewry and Halford (2002), wheat gluten proteins can also be classified into three groups: HMW prolamins with HMW subunits; S-rich prolamins possessing α-gliadins, γ-gliadins, B-type, and C-type LMW subunits; and S-poor prolamins possessing ω−gliadins and D-type LMW subunits.

Glutenin proteins contribute to the cohesiveness and elasticity of dough, which are believed to be mediated by noncovalent bonds such as hydrogen bonds and stabilized by other noncovalent and covalent interactions (Delcour et al., 2012). The viscoelasticity of wheat gluten allows the formation of three-dimensional networks in wheat-based foodstuffs such as bread loaves. Factors reported to affect gluten functionality include temperature, pH, chemical additives such as oxidizing or reducing agents, and enzymes such as peptidases and transglutaminase (Delcour et al., 2012). In addition, covalent disulfide bonds in glutenin play an important role in gluten functionality.

Wheat gluten possesses a glass transition from the glassy state to the rubbery state. Glass transition temperatures for gluten, gliadin, and glutenin were reported to be 130–190, 120–170, and 140–180°C, respectively (Delcour et al., 2012). The incorporation of water into wheat gluten can lower its glass transition temperature (Singh & MacRitchie, 2000). Guerrieri et al. (1996) demonstrated that cross-linking of wheat glutens was formed above 50°C, and gluten hydrophobicity increased at approximately 45°C, which suggests conformational changes of gliadin and glutenin. The temperature of cross-linking reaction for glutenin was reported to be above 60–70°C, whereas the cross-linking temperature for gliadin was above 120°C (Angellier-Coussy et al., 2008; Domenek et al., 2002). Glutenin mobility decreased due to the heat-induced cross-linking reaction with gliadins (Redl et al., 1999). The unique compositions and physicochemical properties of gluten proteins enable their applications in edible coatings and films.

3 | FORMULATION AND PROCESSING OF WHEAT GLUTEN-BASED COATINGS AND FILMS

Wheat gluten–based coatings and films are usually prepared via solvent casting, compression, molding, extrusion, electrospinning, or a combination of these methods, as illustrated in Figure 1 (Ansorena et al., 2016; Gutiérrez et al., 2021; Zhang et al., 2022). Biodegradable and edible macromolecules, such as other proteins (Dong et al., 2022; Zhang et al., 2022), polysaccharides (Chen et al., 2014; Dong et al., 2022), lipids (Ansorena et al., 2016; Fakhouri et al., 2018; Rocca-Smith et al., 2016), acids (Dong et al., 2022; Nataraj et al., 2018), and antioxidants such as rosemary extracts and tannins (Girard et al., 2019; Türe et al., 2008), can be added to improve water resistance, mechanical properties, and antimicrobial performance of the films and coatings (Figure 2). Formulation and processing of wheat gluten coatings and films are shown in Table 1. During these processes, wheat gluten's intermolecular bonds (noncovalent and covalent bonds) break, mobile polymer chains are rearranged in a desired conformation, and new intermolecular bonds and interactions are formed to stabilize the three-dimensional network (Figure 3) (Lagrain et al., 2010). Plasticizers, such as glycerol, sorbitol, diethanolamine, and triethanolamine, can also be added to improve film flexibility, although the addition of plasticizers decreased film's tensile strength and water vapor permeability (Gontard et al., 1993; Irissin-Mangata et al., 2001; Zubeldía et al., 2015).

With the solvent casting method, variables, such as the pH, solvent type, heat treatment, and solvent evaporation, are shown to influence the property and quality of wheat gluten–based films (Gennadios et al., 1993; Kayserilioğlu et al., 2003; Lens et al., 2003). Large wheat gluten particles were found in acidic pH solutions, which affected surface morphology and the appearance of film (Marcuzzo et al., 2010). Moisture content affected wheat gluten's glass transition temperature and further affected films' elongation at break and brittleness (Lens et al., 2003). Kayserilioğlu et al. (2003) reported that at 35% relative humidity (RH), the increase of drying temperature from 20 to 80°C improved the tensile strength of glycerol-plasticized wheat gluten film, whereas at 70% RH, the increase of drying temperature from 20 to 80°C reduced the film's tensile strength. This indicates that moisture content affected the tensile strength of wheat gluten–based film (Kayserilioğlu et al., 2003).

Compression and molding are widely used for making wheat gluten–based films, especially films that contain hydrophobic substances due to low compatibility in aqueous solutions (Wu et al., 2017). Coatings and films made from compression or molding can be either monolayers (Ansorena et, al., 2016; Girard et al., 2019) or multilayers (Fabra et al., 2015; Wang et al., 2022). Compared to solvent cast films, films made from compression showed higher film stress but lower elongation at break (Mangavel et al., 2004). This probably resulted from gliadin polymerization through disulfide bonds, which was caused by the high temperature (120–150°C) during compression (Mangavel et al., 2004). However, Zuo et al. (2009) demonstrated that in the presence of methylcellulose, mechanical properties of wheat gluten/methylcellulose/glycerol made from casting were superior to those of films made from compression. The dispersion of methylcellulose and the degree of methylcellulose/wheat gluten mixing were different between solvent casting and compression methods, which led to the difference in mechanical properties (Zuo et al., 2009).

Compression and molding temperature and time can influence film properties due to the effect of heat and temperature on wheat gluten's glass transition temperature, especially with the incorporation of water or plasticizers (Angellier-Coussy et al., 2011). That said, the compression or molding temperature has more effect than the

TABLE 1 Formulation, processing methods, and characterization of wheat gluten–based coatings and films

Formulation	Processing methods	Film properties and functions	Reference
2.5 g of glycerol, 10 g of wheat gluten, 40 g of deionized water, 50 g of absolute ethanol and hydrochloric acid, and 2.5 g lipid	Solvent casting	Water sorption, water affinity, and water transfer decreased; extensibility increased	Rocca-Smith et al. (2016)
3 g of wheat gluten dissolved in 100 ml of 0.5% tartaric acid solution (w/v), and 30% glycerol (dry basis)	Solvent casting	Water resistance and UV resistance increased	Dong et al. (2022)
3 g wheat gluten, 1% (w/w) carboxymethyl cellulose, and 30% glycerol (dry basis)	Solvent casting	Thermal stability increased	Dong et al. (2022)
3 g wheat gluten, 1% (w/w) egg white protein, and 30% glycerol (dry basis)	Solvent casting	Physical strength and elongation at break increased	Dong et al. (2022)
3 g wheat gluten, 1% (w/w) apple pectin, and 30% glycerol (dry basis)	Solvent casting	Smooth and homogeneous film	Dong et al. (2022)
3 g of wheat gluten in 100 ml deionized water, and 30% glycerol (dry basis)	Solvent casting	Elongation at break increased	Dong et al. (2022)
Banana fiber–wheat gluten film (ratio of 30/70, 50/50, 70/30, w/w%) and glutaraldehyde (5%, 10%, and 20%, on the basis of film)	Solvent casting combined with molding	Glutaraldehyde addition reduced water sorption of film, increased film strength and thermal stability	Nataraj et al. (2018)
Banana fiber–wheat gluten film (ratio of 30/70, 50/50, 70/30, w/w%) and citric acid (5%, 10%, and 20%, on the basis of film)	Solvent casting combined with molding	Citric acid addition increased film strength and thermal stability	Nataraj et al. (2018)
1–2.5 g of wheat gluten, 0.5–1 g of carboxymethyl cellulose, 0%–10% cellulose nanofiber (solid matter basis), and 30% plasticizer (biopolymer basis)	Solvent casting	The optimal formulation includes 1% gluten, 0.868% carboxymethyl cellulose, and 8.549% cellulose nanofillers according to the evaluation of microstructure, surface topography, and distribution of nanocellulose	Bagheri et al. (2019)
Wheat gluten and methylcellulose (MC) dissolved in ammonia at 5 and 2.5 wt%, respectively, and glycerol 25 wt% (on the basis of wheat gluten and MC)	Molding	The addition of MC from 0.2% to 1.0% increased Young's modulus from 60.5 to 609.9 MPa, tensile strength from 1.7 to 44 MPa, and strain at break from 19.2% to 41% at 100°C, respectively. The addition of MC from 0.2% to 1.0% increased Young's modulus from 187 to 680 MPa, tensile strength from 5.2 to 45.9 MPa, and strain at break from 10.2% to 39.3% at 125°C, respectively	Zuo et al. (2009)
Wheat gluten and glycerol (15, 20, and 25 wt%)	Solvent casting combined with compression	The addition of glycerol from 15% to 25% (wt) decreased tensile strength from 325.7 to 22.6 MPa but increased elongation at break from 21.2% to 39.8%	Zubeldía et al. (2015)

(Continues)

TABLE 1 (Continued)

Formulation	Processing methods	Film properties and functions	Reference
Wheat gluten, methylcellulose microfibers dissolved in ammonia at 5 and 2.5 wt%, respectively, and glycerol 25 wt% (on the basis of wheat gluten and methylcellulose microfibers)	Molding	The addition of methylcellulose microfibers increased elongation at break and tensile strength	Song and Zheng (2009)
Solutions prepared by 25% (w/v) gluten in 50% acetic aqueous solution without or with addition of 4%, 8%, 12%, 16% glycerol monolaurate (gluten basis)	Electrospinning	Glycerol monolaurate addition resulted in more hydrophilic film and enhanced water stability and antimicrobial activities against *Staphylococcus aureus* and *Escherichia coli*	Zhang et al. (2020)
Wheat gluten/glycerol/distilled water (55/22.5/22.5) with or without 1% montmorillonite, or 1% sodium hydroxide, or 1% sodium hydroxide and montmorillonite; wheat gluten/glycerol/distilled water (65/17.5/17.5) with or without 1% sulfuric acid, or 1% sulfuric acid and montmorillonite	Extrusion and compression	The addition of montmorillonite improved the water absorption, rheological, and mechanical properties of resultant films	Cortés-Triviño and Martínez (2018)

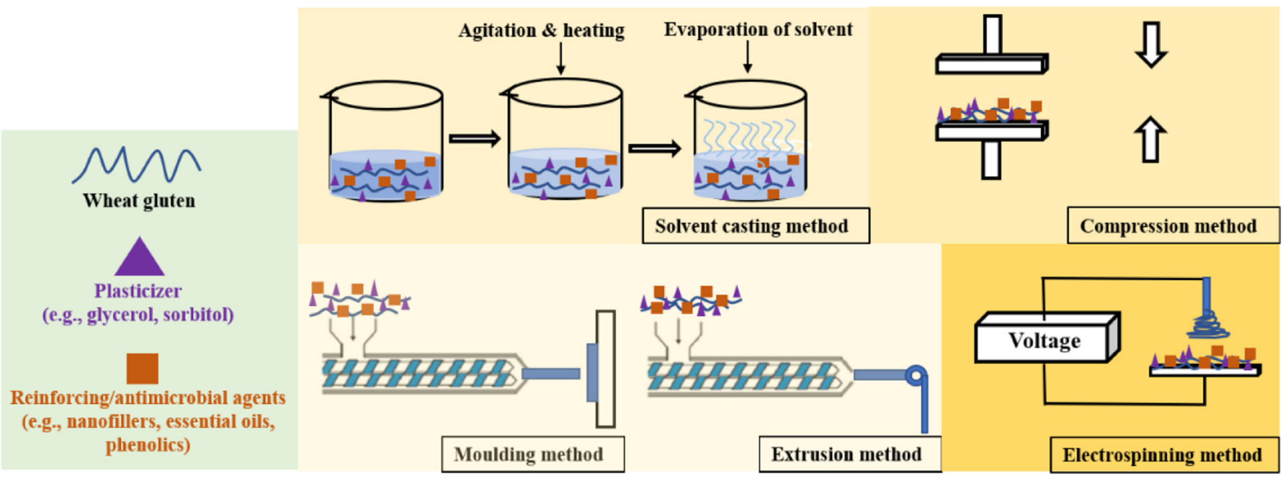

FIGURE 1 Different methods for producing wheat gluten–based coatings and films

compression or molding time. This is because the formation of gluten coatings and films depends on the breakdown of disulfide bonds and the formation of new disulfide bonds during heat treatment, and temperature influences the degree of final cross-linking in gluten networks (Gällstedt et al., 2004; Zubeldía et al., 2015). In addition, the transition from the rubbery state to the glassy state will influence polymer chain mobility, cross-linking, and aggregation of wheat gluten, thus affecting thermal degradation and mechanical properties (Lagrain et al., 2010).

Extrusion processing involves heat, shear, and pressure, which strongly influence the molecular mobility of wheat gluten (Redl et al., 2003). Heat and shear during extrusion processing affect wheat gluten denaturation and cause the breakage of native hydrophobic and disulfide groups, thus influencing the thermal and mechanical properties of films (Asgher et al., 2020).

Electrospinning processing is a widely used method to synthesize nanofibrous scaffolds and has been used to process wheat gluten films incorporated with nanofillers for food packaging. During electrospinning, the polymer solution is charged with high voltage through the tip of a needle. Then, the electric field overcomes the surface tension, and the charged solution evaporates. Finally, extended polymer solution droplets form nanofibers on the

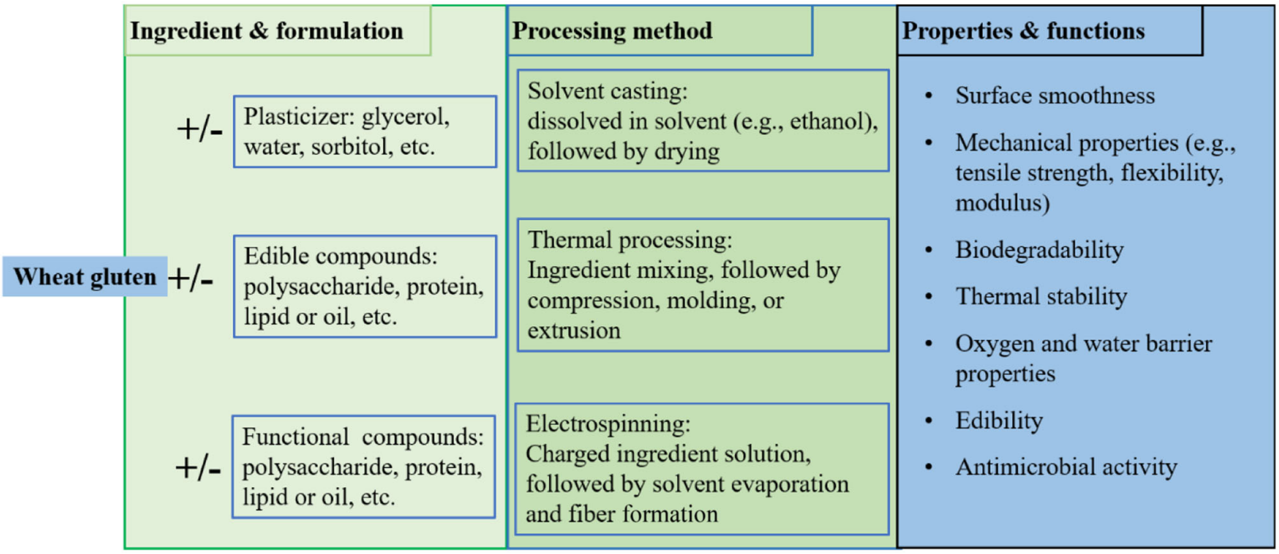

FIGURE 2 Preparation of wheat gluten–based coatings and films

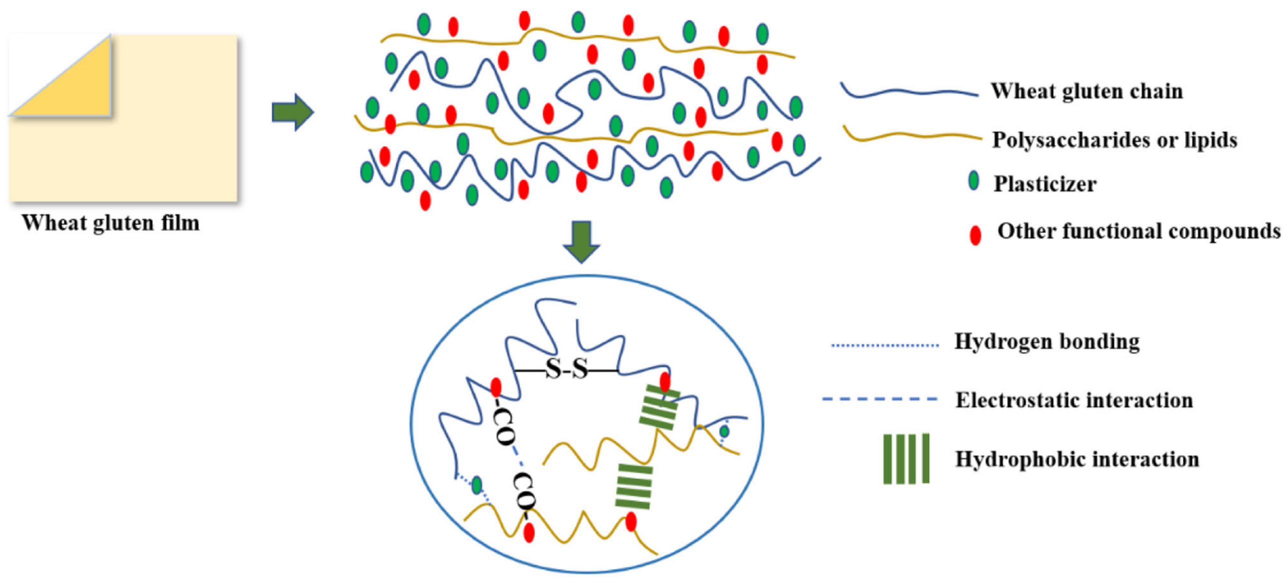

FIGURE 3 Interactions between wheat gluten and polysaccharide, protein, lipid, and bioactive compounds

collector (Fabra et al., 2015; Senthil Muthu Kumar et al., 2019). Zhang et al. (2020) incorporated glycerol monolaurate into wheat gluten–based film through electrospinning and found that the addition improved the film's water stability and antimicrobial activity against *Staphylococcus aureus* and *Escherichia coli*.

Cold-plasma treatment and glycation have been shown to enhance the properties and functions of wheat gluten–based films (Moosavi et al., 2020; Zhang et al., 2022). Cold-plasma treatment enhanced film's tensile strength and surface smoothness and reduced its oxygen transmission rate (Moosavi et al., 2020). In addition, cold-plasma treatment did not significantly affect film's water solubility and permeability. Wheat gluten film with zein nanofiber that was glycated with xylose through the Maillard reaction has been reported to have higher thermal stability, water vapor resistance, water stability, and elasticity (Zhang et al., 2022).

In summary, solvent casting, compression, molding, extrusion processing, and electrospinning are the major methods to process wheat gluten–based coatings and films. Solvent casting method usually resulted in high-elongation films, whereas compression or molding method usually resulted in rupture-resistant films (Mangavel et al., 2004).

Extrusion processing is usually operated at high temperature that may largely alter the structure and properties of wheat gluten, thus further affecting resultant films. The glass transition properties of wheat gluten are the main factors that influence the thermal and mechanical properties of coatings and films. Variables in processing, including the addition of plasticizers and other biodegradable and edible compounds, affect wheat gluten's phase transition properties, therefore changing the thermal and mechanical attributes of the resultant coatings and films. However, little is known about the effect of adding these additives on wheat gluten glass transition properties and underlying mechanisms. More research on this topic is warranted.

4 | PROPERTY ENHANCEMENT OF WHEAT GLUTEN-BASED COATINGS AND FILMS

Wheat gluten–based coatings and films possess good oxygen barrier properties due to their high resistance to nonpolar substances, such as O_2, CO_2, and lipid (Lin & Zhao, 2007). However, due to their hydrophilic nature, they have poor mechanical strength (including tensile strength and elongation at break), weak water resistance, and surface cracks. This section focuses on ways to improve the thermal, mechanical, and barrier properties of wheat gluten coatings and films by incorporating other proteins, polysaccharides, lipids, and other additives.

Adding polysaccharides, such as cellulose, wheat bran, methylcellulose, and chitosan-to-wheat gluten–based coatings, and films can enhance their properties due to the interaction between polysaccharides and wheat gluten (Bagheri et al., 2019). Mastromatteo et al. (2008), Chen et al. (2014), Mastromatteo et al. (2008), Song and Zheng (2009), and Zuo et al. (2009) studied the effect of adding glycerol, spelt, and wheat bran to wheat gluten films and found that the presence of glycerol increased the films' water vapor permeability, whereas the addition of bran reduced the water vapor permeability. In addition, the higher content of bran increased the film's elastic modulus, whereas the higher content of glycerol decreased the elastic modulus (Mastromatteo et al., 2008). Maximum addition of glycerol with minimum spelt and wheat bran resulted in the highest value of elongation at break, whereas minimum glycerol and maximum spelt and wheat bran led to the lowest value of elongation at break. Incorporating methylcellulose microfiber (0%–42.8%, wt) into glycerol-plasticized wheat gluten–based film significantly increased the glass transition temperature, the film's elongation at break, and its tensile strength (Song & Zheng, 2009). The addition of chitosan to wheat gluten was found to significantly increase the extensibility, toughness, and moisture solubility of film (Chen et al., 2014). Sartori et al. (2018) reported that films made from pectin–wheat gluten at 1:1 or 3.75:1 ratio showed greater elongation at break compared to the film made from pectin–wheat gluten at 1:3.75 ratio, whereas the latter exhibited higher water vapor barrier property and lower solubility (Sartori et al., 2018). In addition, film made from pectin–wheat gluten showed a greater UV/light barrier property compared to synthetic polyethylene film (Sartori et al., 2018).

Nanofillers possess a high geometrical aspect ratio and have the potential to reinforce the mechanical and barrier properties of nanocomposite film (Xu et al., 2019, 2020). Rafieian et al. (2014) optimized wheat gluten film formulation with cellulose nanofibrils, glycerol, and SDS through response surface methodology. The condition of 11.129/100 g cellulose nanofibrils, 35.440/100 g glycerol concentration, and 6.259/100 g SDS concentration resulted in the highest tensile strength and elongation at break, which were 3.63 MPa and 86.03%, respectively (Rafieian et al., 2014). There was no significant difference in the glass transition temperature and degradation behavior between films created under this condition and the control film, indicating that the addition of cellulose nanofibrils, glycerol, and SDS did not affect the thermal properties of wheat gluten–based films (Rafieian et al., 2014). The optimal formulation for wheat gluten–based films with cellulose nanofiller (CNF) and carboxymethyl cellulose (CMC) was also determined: Overall, 1% wheat gluten, 0.686% CMC, and 8.549% CNF resulted in desirable water vapor permeability and mechanical properties (Bagheri et al., 2019). Cortés-Triviño and Martínez (2018) reported that pH has a significant effect on the mechanical properties of wheat gluten–based films. Films made at pH 6 exhibited the highest value for Young's modulus and tensile strength compared to films made at pH 5.4 and 10.8. The strain at break, however, was not significantly affected by the pH. In addition, the addition of montmorillonite nanoclays (MMT) (1%, wt) to wheat gluten improved films' mechanical and rheological properties at all pH conditions due to the exfoliation of MMT in the wheat gluten matrix (Cortés-Triviño & Martínez, 2018).

Hydrophobic substances, such as lipid and oil, were also added to wheat gluten to improve coatings and films' water resistance and antimicrobial and antioxidant activities. For example, the addition of lipid (25%, wt, db) (a blend of beeswax, mono- and di-glycerides, and glycerol monostearate) to wheat gluten has been shown to significantly reduce films' water sorption, water affinity, and water transfer (Rocca-Smith et al., 2016). The addition of lipid to wheat gluten altered the glass transition temperature at the 0.11–0.75 water activity range. Correspondingly, the addition of lipid to wheat gluten reduced the network structuration, which was reflected in a decrease in Young's modulus and tensile strength at the 0.11–0.84 water activity range. The addition of lipid to wheat gluten increased the elongation at break at water activity of 0.75 and 0.84, whereas it decreased the elongation at break at water activity from 0.11 to 0.65 (Rocca-Smith et al., 2016). Ansorena et al. (2016) reported that the addition of thyme essential oil (0%–15%, wt) to wheat gluten significantly decreased tensile strength, storage modulus, and moisture sorption of glycerol-plasticized (20%, wt) films. The addition also improved the film's deformability but did not significantly affect its water vapor permeability and total soluble matter. The addition of thyme essential oil (0%–15%, wt) to wheat gluten improved film's in vitro antioxidant and antimicrobial properties.

Zhang et al. (2020) added glycerol monolaurate (4%, 8%, 12%, and 16%, wheat gluten basis) to wheat gluten and found that the addition increased film's tensile strength and decreased the elongation at break. This was probably due to the brittle network formed between gluten and glycerol monolaurate. More addition of glycerol monolaurate to wheat gluten increased the film's swelling ratio but reduced its weight loss (Zhang et al., 2020). Nataraj et al. (2018) investigated the effect of glutaraldehyde (5%, 10%, and 20%) and citric acid (5%, 10%, and 20%) on the mechanical, thermal, and water barrier properties of banana fiber/wheat gluten film (30/70, 50/50, 70/30, w/w). The separate addition of glutaraldehyde and citric acid was found to reduce film's water sorption. This probably resulted from wheat gluten and the amine and hydroxyl groups on the fiber forming cross-links. The cross-links reduced the number of hydroxyl groups on the surface to form hydrogen bonds with water, which in turn affected water sorption (Nataraj et al., 2018). The addition of glutaraldehyde or citric acid also increased film's tensile strength compared to that of the control film (Nataraj et al., 2018). Khashayary and Aarabi (2021) found that the addition of vanillin (0.92%), salicylic acid (0.94%), and montmorillonite cloisite (1.92%, w/w) to wheat gluten created wheat gluten–based film with maximum mechanical properties and considerable water resistance. These additions improved film's tensile strength due to the formation of covalent bonds between wheat gluten's amino acid groups and aldehydes; they increased film's water solubility due to the addition of hydrophilic substances (Khashayary & Aarabi, 2021).

Phenolic compounds were also incorporated into wheat gluten to improve film properties. Girard et al. (2019) studied the effect of condensed tannins (proanthocyanidins, PA), hydrolysable tannins (tannic acid), and catechin on surface morphology and tensile strength. Scanning electron microscope analysis showed that the addition of catechin (10 mg/g) to wheat gluten resulted in a smooth film, whereas the addition of PA (10 mg/g) and tannic acid (10 mg/g) reduced the cracks of film compared to pure wheat gluten film (Girard et al., 2019). The addition of PA to wheat gluten significantly improved tensile strength, whereas the addition of tannic acid and catechin did not (Girard et al., 2019). Film's elongation at break was not significantly influenced by the addition of catechin, PA, or tannic acid. The in vitro digestibility and solubility of film with added catechin, PA, and tannic acid were tested at pH 2 and 7 to mimic digestion in the stomach and intestine, respectively (Girard et al., 2019). Results showed that at pH 2, PA significantly reduced film's digestibility and solubility. The cross-linking between PA and wheat gluten formed insoluble complexation, which lowered digestibility and solubility in acidic pH. At pH 2, catechin and tannic acid did not significantly affect digestibility and solubility compared to the control film. At pH 7, there is no significant difference in digestibility and solubility in any films.

Taken together, the addition of edible macromolecules, such as polysaccharides, proteins, lipids, and oils to wheat gluten, has been found to enhance the mechanical properties, water vapor permeability, and antimicrobial activity of gluten-based coatings and films. The compatibility and surface morphology of wheat gluten–based films can also be affected by factors, such as hydrophobic interaction and pH. Most studies focused on the mechanical, water barrier, and antimicrobial properties of wheat gluten–based films incorporated with edible compounds, but little is known about the solubility and digestibility of resultant films. This will be a useful area for future research.

5 | ANTIMICROBIAL FUNCTIONS AND APPLICATIONS OF WHEAT GLUTEN-BASED COATINGS AND FILMS

There is a growing demand for antimicrobial food packaging for inhibiting microorganism growth, maintaining food quality, and extending the shelf life of packaged foodstuffs. Incorporation of antioxidants and antimicrobial agents can improve the antimicrobial activities of wheat gluten coatings and films (Zhu et al., 2022). For example, the addition of food preservative and essential oil was reported to enhance the antimicrobial activity. When 10% and 2.5% (wt) potassium sorbate was added to glycerol-plasticized wheat gluten film, the film inhibited the growth of *Aspergillus niger* and *Fusarium incarnatum*, respectively (Türe et al., 2012). The inhibitory effect of thyme essential oil was measured by the size of the "clear" inhibition zone in the presence of bacteria. Results showed that the addition of thyme essential oil (3.5%, 10%, and 15%, wt) to wheat gluten significantly increased the "clear" zones in the film in the case of native microflora from lettuce and broccoli, *E. coli* and *Pseudomonas aeruginosa*, *Listeria* *innocua*, and *S. aureus* (Ansorena et al., 2016). The addition of glycerol monolaurate to wheat gluten–based film was also reported to significantly suppress the growth of *S. aureus* and *E. coli* because glycerol monolaurate could disrupt the phospholipid membranes of microorganisms, alter membrane permeability, and result in ion leakage (Zhang et al., 2020).

Antioxidative plant phenolic extracts have also been incorporated into wheat gluten–based coatings and films to improve antimicrobial function (Girard et al., 2019; Khashayary & Aarabi, 2021). Türe et al. (2008) added natamycin, rosemary extract, and a combination of natamycin and rosemary extract, respectively, to wheat gluten/methyl cellulose composite to determine the antimicrobial effect against *A. niger* and *Penicillium roqueforti* on potato dextrose agar. Natamycin addition at 2 and 1 mg/10 g inhibited the growth of *A. niger* and *P. roqueforti*, respectively (Türe et al., 2008). The addition of rosemary extracts alone did not show any antimicrobial effect, whereas the combination of natamycin and rosemary extract inhibited the growth of *A. niger* (Türe et al., 2008). The addition of vanillin and salicylic acid to wheat gluten montmorillonite nanoclay film also showed inhibitory effect against *A. niger* and *Alternaria alternate PTCC5224* strains (Khashayary & Aarabi, 2021).

The antimicrobial effect of wheat gluten–based film on the quality of packaged foodstuffs has also been reported. Tanada-Palmu & Grosso (2005) investigated the effect of bilayer biocomposites of wheat gluten and lipids (beeswax and static and palmitic acids) on the quality of refrigerated strawberries. Reduced numbers of infected strawberries and lower weight loss were observed for strawberries covered with coating or film during 16 days of storage (Tanada-Palmu & Grosso, 2005). In addition, film was more effective in maintaining strawberry quality than coating. Strawberries covered with coating or film showed better color and firmness retention than the control strawberries (Tanada-Palmu & Grosso, 2005). Strawberries covered with coating showed better physicochemical attributes, including titratable acidity, total soluble solids, and reducing sugar than strawberries covered with film and the control strawberries (Tanada-Palmu & Grosso, 2005). Sensory analysis showed that strawberries covered with coating were acceptable to consumers.

Chen et al. (2022) reported that edible coatings and films based on wheat gluten and lignocellulose nanofibers showed strong antimicrobial effect against *E. Coli*, UV blocking property, water barrier property of 10.3 g mm^{-1} m^{-2} day^{-1}, and gas barrier property of 15.9 cm^3 μm m^{-2} day^{-1} kPa^{-1}. The preservation effect of wheat gluten/lignocellulose nanofiber coating on bananas, grapes, and persimmons was studied, and results showed that the coating retained more than 90% of the fruits' original weight over 7 days. The coating also reduced the decaying and enzymatic browning of bananas compared to uncoated bananas. Fruits, including cherry, litchi, and waxberry, stored in sterilized boxes and covered with wheat gluten lignocellulose nanofiber film showed the most edible rate and lowest weight loss compared to uncovered fruits and fruits covered with plastic wrap (Chen et al., 2022).

The quality of cheese covered with wheat gluten–based film was investigated. Türe et al. (2010) reported that the addition of natamycin (5–20 mg per 10 g) to wheat gluten/methyl cellulose film significantly reduced the 2-log growth of *A. niger* in spore counts, and the addition of natamycin (2 mg per 10 g) to wheat gluten–based film also significantly inhibited the growth of *A. niger* on cheese surface (Türe et al., 2010). By contrast, the addition of natamycin to wheat gluten/methyl cellulose film did not significantly reduce the growth of *P. roqueforti* in either spore counts or no cheese surface (Türe et al., 2010). Özkök and Caba (2019) compared the quality of cheese coated with protein (wheat gluten, soy protein, and whey protein) and red grape juice during storage. Wheat gluten coating was more flexible and adhesive compared to soy protein and whey protein coatings (Özkök & Caba, 2019). However, cheese covered with wheat gluten coating experienced more moisture loss compared to soy protein and whey protein coatings. The inhibitory effect of red grape juice against yeast growth was observed in cheese covered with soy protein and whey protein coatings; no yeast content was observed in cheese covered with wheat gluten coasting, which is probably due to the inhibitory effect of the ethanol added during coating manufacturing (Özkök & Caba, 2019).

Perishable vegetables and fruits are the main agricultural products that require protection from coatings and films to maintain or improve quality, extend shelf life, and inhibit microbial growth. However, so far, only few fruits covered with wheat gluten–based coatings or films during storage have been studied for quality and physicochemical properties. The coating and film's underlying mechanism for food preservation such as their effect on respiration rate and polyphenol oxidase activity is not well studied. More studies are needed to better understand the effect of wheat gluten–based coatings and films on the biological activity, quality, and preservation of postharvest agricultural products.

6 | CONCERNS WITH WHEAT GLUTEN IN COATINGS AND FILMS

Although wheat gluten has been extensively utilized in food products and edible coatings and films, it can trigger immune disorders in celiac disease populations and cause intestinal lining damage, diarrhea, fatigue, weight loss, bloating, and anemia (Gujral et al., 2012). Thermal processing of wheat gluten can alter its antigenicity through conformational changes and/or digestion (Pasini et al., 2001). Rahaman et al. (2016) and Rumbo et al. (1996) studied the effect of processing conditions, including pH (3, 5, and 7), temperature (80, 90, and 100°C), and shear (500, 1000, and 1500 s^{-1}) on wheat gluten's antigenicity. The antigenicity was reduced at pH 3 and 90°C, pH 5 or 7 and 100°C. The pH and temperature were reported to contribute more to the reduction of antigenicity than shear (Rahaman et al., 2016). It is believed that pH and temperature affected the burial of some antigenic hydrophobic residues, modified thiol content, or destroyed/masked some epitopes (Rahaman et al., 2016).

To prevent gluten-related disorders, wheat gluten is not allowed for dietary intake among celiac disease populations. However, to date, there is no report yet about allergy associated with wheat gluten–based coatings or films. More research is warranted to study the safety of these coatings and films, such as their safe content limit.

7 | CONCLUSION AND OUTLOOK

This article comprehensively reviewed wheat gluten–based coatings and films for food packaging. Topics reviewed include coating and film formulation, production, properties, functions, and applications. Wheat gluten–based coatings and films are primarily produced through solvent casting, compression, molding, extrusion, and electrospinning. The coatings and films are highly resistant to oxygen and thus possess good oxygen barrier properties. The improvements of their mechanical properties and water vapor property have been extensively studied through methods such as wheat gluten modification or the incorporation of macromolecules or antioxidants. Different processing methods and the addition of other biomacromolecules or compounds can alter wheat gluten's glass transition property, which further alters the coatings and films' thermal, mechanical, and barrier properties. However, systematic studies regarding the relationship between coating and film formulation, processing methods, and properties are still lacking. In addition, more suitable processing methods for wheat gluten–based edible film are still warranted for optimizing the quality with reasonable cost.

The antimicrobial activity of wheat gluten–based films has attracted increasing interest due to the promise to inhibit microbial growth, extend shelf life, and improve the qualities of packaged foodstuffs. However, existing studies on the incorporation of antimicrobial agents into wheat gluten–based coatings and films are few, and only a few antioxidants have been studied. The addition of antimicrobial agents resulted in structural complexity of wheat gluten–based edible film; thus, more attention is required. More research is needed on the effect of antimicrobial agents on microbial growth and qualities of packaged foodstuffs during storage. In addition, there are still some challenges with the antimicrobial film, such as industrial-scale production, consumer acceptance, improved sustainability, and practical applications in various types of foods, which require more research in the future.

AUTHOR CONTRIBUTIONS

Jingwen Xu: Conceptualization; methodology; formal analysis; investigation; resources; writing – original draft; writing – review and editing; project administration. **Yonghui Li**: Conceptualization; methodology; formal analysis; investigation; resources; writing – review and editing; project administration.

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CONFLICTS OF INTEREST

The authors declare that they have no known conflict of interests.

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