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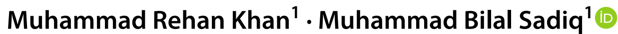

REVIEW PAPER

Importance of gelatin, nanoparticles and their interactions in the formulation of biodegradable composite films: a review

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Abstract
This study reviews the importance of gelatin, nanoparticles and their interactions in the formulation of biodegradable composite films. Gelatin is widely used in packaging because of its unique characteristics, i.e., film-forming ability and barrier properties. However, high moisture sensitivity and hydrophilic nature of gelatin restrict its application in food packaging. Therefore, gelatin is used in the formulation of composite films with better functional and barrier properties for food packing. The incorporation of nanoparticles (NPs) in gelatin film-forming solution improves the mechanical, thermal, barrier and optical characteristics of gelatin composite films. Furthermore, this review compiles the functional properties of gelatin extracted from different sources and functional characteristics of gelatin composite films incorporated with different NPs.

Keywords Gelatin composite films · Nanoparticles · Biodegradable packaging

Introduction

Plastic products of petrochemical origin are widely used in food packaging industry because of their excellent structural, barrier properties, cheap cost and aesthetic qualities. However, there is a growing concern about the associated negative attributes of these materials as they are not derived from sustainable sources. As a result, most of the conventional foods packaging materials due to their non-biodegradable nature are sent to recovery sites for incineration or to landfill for dumping [1].

Biodegradable films made from biopolymers play an important role in reducing the environmental impact of conventional plastic packaging materials [2]. Proteins and polysaccharides are the main biopolymers used in the development of biodegradable

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packaging material [1]. Commonly studied proteins for biodegradable films are milk proteins such as whey proteins [3], gelatin [4, 5] and soy protein [6]. Among polysaccharides carboxymethyl cellulose [7], starch [2] and chitosan [8] are mostly used for food-based biodegradable packaging.

The annual world production of gelatin is approximately 326,000 tonnes [9]. While the most important resources of gelatin production are pigskin (46%), cattle skin (29%), cattle and pig bones (23%) and other sources (2%) [10], gelatin has been extensively studied for its functional properties, as a barrier to protect food from drying, exposure to light and oxygen [8]. Gelatin is produced by the hydrolysis of collagen from bones, skin and connective tissues which are generated as a by-product/waste during slaughtering and processing [9]. The diverse functionalities of gelatin as a gelling agent, film-forming material, generally recognized as safe (GRAS) food additive and biodegradable polymer, have increased its utilization in food industry [11]. The gelatin films exhibit satisfactory mechanical properties, intermediate relative humidity and excellent oxygen barrier properties, but these characteristics are impaired by high moisture sensitivity and hydrophilic nature of gelatin, thus limiting its broader applications [12]. Therefore, the current trends in designing gelatin-based biodegradable packaging materials are based on optimization of functional properties of packaging material by studying the effects of different nanoparticles (NPs) (1–100 nm), i.e., silver, gold, titanium dioxide (TiO_2), zinc oxide (ZnO), montmorillonite (MMT) and copper, etc. [13–21]. Various researchers reported improved barrier and mechanical properties of gelatin nanocomposite films as compared to gelatin films [13, 18, 22]. Metal oxide NPs such as TiO_2 have also gained much attention in recent years because of its cheaper price, non-toxic and photostable properties [23]. The addition of TiO_2 NPs into gelatin films significantly improved tensile strength (TS) and elongation at break (EAB) of the films and reduced the water vapor permeability (WVP) due to creation of twisted pathways across film [23]. However, a higher concentration of NPs is disadvantageous for performance enhancement due to aggregation of NPs in film matrix, thus leading to reduced mechanical strength [16]. Hosseini et al. [24] reported an increase in TS from 7.44 to 11.28 MPa, and EAB was decreased from 102.04 to 32.73% with increasing chitosan NP content from 0 to 8% (w/w) due to reinforcement effect of NPs and strong hydrogen bonding between NPs and gelatin matrix. Furthermore, several NPs (i.e., silver, copper, MMT and gold) have been reported to impart their antimicrobial activity to gelatin-based films against food-borne pathogens [13–21].

Due to growing interest in biodegradable gelatin composite films, this review aims to explain the importance of gelatin sources, structure and functional properties in the formulation of composite films; moreover, the effect of NPs on mechanical, permeability, structural, antibacterial and biodegradable properties of gelatin films was also studied.

Components of biodegradable/edible films

At least one film-forming macromolecule (proteins, polysaccharides and/or lipids), a plasticizer, a solvent and a surfactant are involved in preparation of biodegradable or edible films [25]. An edible biodegradable film is produced by using dry or wet

manufacturing process, typically from food-derived ingredients. The resulting sheet should be a free-standing one that can be put on or between food components [26]. By controlling the transfer of oxygen, moisture, carbon dioxide, flavor and aroma of the food components, the biodegradable films offer a great potential to improve food quality and to extend the shelf life [1].

According to Suderman et al. [11], biopolymers used for the preparation of edible films can be classified into three different categories: polysaccharides, proteins (animal and plant based) and lipids. Before the formulation of biopolymer-based edible films, some important factors such as solubility, microbiological stability, wettability, transparency, cohesion, oil and grease resistance, permeability to water and gases, sensory and mechanical properties need to be considered [27]. In order to reduce surface tension and adhesion of plasticized films, surfactants are incorporated in film-forming solution (FFS) [28]. In the formulation of edible films, most widely investigated biomaterials are the hydrocolloids (proteins and polysaccharides) [11].

Among all the biopolymers, proteins, because of their good functional properties, are the most commonly used food packaging material [27]. Proteins typically occur either as globular or fibrous proteins and are polymers of proteinogenic amino acids linked through amide bonds with high intermolecular binding potential [29]. Globular proteins are folded into complex structures having covalent, ionic and hydrogen bonds, while fibrous proteins are responsible for making fibers as a result of close association with each other through hydrogen bonding [30]. Protein-based packaging films exhibit lower mechanical strength and poor water resistance when compared with synthetic packaging films. Still, polysaccharides are generally inferior as compared to the proteins in their film-forming abilities with inferior barrier and mechanical properties [31]. Compared to other protein-based biopolymers (i.e., soy protein, corn zein, wheat gluten and pea protein) having high melting temperatures, gelatin presents thermoreversible nature, low melting temperature with ability to melt-in-mouth [1].

Gelatin extraction

Gelatin is most commonly extracted from porcine, bovine, marine and poultry sources. Gelatin extracted from poultry and marine sources can serve as a better option as compared to bovine and porcine sources not only because of religious and social reasons, but also due to the fact that proteins derived from these sources (marine and poultry) have never had any association with bovine spongiform encephalopathy, which is a zoonotic degenerative neural disease reported from bovine sources [24, 32]. Various research studies reported the extraction of gelatin from fish and chicken mainly involving alkaline and acidic pretreatment followed by final extraction [4, 5, 33, 34].

Briefly, for extraction of gelatin, defatted freeze-dried skin samples are soaked in various concentrations of NaOH (0.01–0.2 M) at room temperature for 30–60 min followed by centrifugation at 4500–6500×*g* for 10 min or washed with distilled water depending upon sample source, respectively, i.e., chicken and/or fish [5, 35–37].

After alkaline pretreatment, samples are soaked in acetic acid (0.05–0.2 M) for 3–4 h at 4 °C or blend with 0.15% H_2SO_4 before citric acid (0.007–0.036 M) pretreatment for 30 min at 25 °C followed by washing with distilled water in order to remove residual matter [5, 35, 37–39].

Finally, pretreated samples are soaked in distilled water at 45–50 °C for 12–17 h to extract gelatin from skin. The solubilized gelatin solutions are filtered with cheese cloth or Whatman No. 4 filter paper and the filtrate is concentrated by using a vacuum rotary evaporator followed by freeze drying. The dry matter obtained is referred as gelatin powder [5, 38, 39].

The bones from slaughtered cattle are usually degreased, dried, sorted and crushed to a particle size of 1–2 cm. The bone pieces are washed with hydrochloric acid to remove minerals. The resulting spongy material is known as ossein. Cattle hides and ossein are treated with lime and water, liming usually takes around 8–12 weeks before final extraction with hot water. For the extraction and production of gelatin from porcine sources, pork skin is usually dehaired with hot dilute caustic soda solution followed by washing with cold water then soaked in cold dilute mineral acid (usually hydrochloric or sulfuric acid) for several hours and washed with cold water several times to be finally extracted with hot water. The number of extractions usually varies from 3 to 6. Initially temperature for extraction is 50–60 °C followed by temperature increase (5–10 °C) with subsequent extractions [40].

Gelatin structure and functional characteristics

Gelatin is derived from partial hydrolysis of collagen and contains high contents of amino acids such as glycine (Gly), proline (Pro) and hydroxyl proline (Hyp), which is a characteristic feature of gelatin. Gelatin also contains single or double unfolded chains of hydrophilic character, i.e., combination of α-chains (single chain/one polymer), β-chains (two α-chains cross-linked covalently) and γ-chains (covalently cross-linked three α-chains) [41]. It has been reported that amino acid compositions, relative content of α-chains, β- or γ-components, presence of protein fragments of lower molecular weight and higher molecular weight aggregates, had an effect on physical properties of gelatin. The presence of low molecular weight protein fragments was found to be responsible for lowering the gelling temperature of gelatin due to low degree of proline and lysine hydroxylation. However, the high gel strength of gelatin was mainly dependent on the high molecular weight protein fraction and higher content of α-chains [41, 42]. Gly has been reported to be the most abundant amino acid in gelatin, obtained from bovine skin and hide [43], porcine skin [44], fish skin [45] and chicken skin [46]. Gelatin also contains high content of alanine (Ala), glutamine/glutamic acid (Glu) and asparagine/aspartic acid (Asp) apart from Pro and Hyp [47]. The presence of cysteine (Cys) in amino acid composition of gelatin is an indication that a small quantity of stroma protein, which is highly insoluble and stable in salt such as elastin, might be present in gelatin [48]. Gelatin containing higher amount of Pro, Ala and Hyp exhibited the ability to develop triple-helix structures and considered to have higher viscoelastic properties, which are essential for gelatin gel structure stabilization as well as for gelatin-based films [42]. Amino acid

composition of gelatin extracted from various sources, i.e., fish [33, 42, 45, 49–58], porcine [38, 43, 44], bovine [43, 44, 46, 59, 60], squid [42, 52], duck [60, 61] and chicken [35, 46], is summarized in Table 1.

The most important physical properties of gelatin are gel strength and viscosity [62]. Bloom value which is another name for gel strength reflects the stiffness and strength of the gelatin and ranges from 30 to 300 bloom (220–300 is considered to be a high bloom, 150–220 a medium bloom and < 150 a low bloom) [46]. Greater gelatin strength is an indication of higher bloom value [63]. It is a well-established fact that imino acid content is responsible for stabilization of triple helix of collagen structure through hydrogen bonding between hydroxyl groups of Hyp in gelatin and free water molecules [64]. Hafidz et al. [43] reported that high amount of serine (Ser) and tyrosine (Tyr) in pigskin gelatin, having free hydroxyl group, lead to the formation of hydrogen bonds resulting into increased gel strength. The molecular weight and content of Hyp are the factors that might affect bloom strength [38]. According to Gòmez-Guillèn et al. [42] gelatin extracted from sole and megrim skin showed gel strength five times higher than cod gelatin, while squid gelatin showed the lowest gel strength (Table 2). The lowest gel strength showed by

Table 1 Amino acid composition of gelatin extracted from different sources

Amino acids	Composition of gelatin (g/100 g protein) from different sources					
	Fish	Porcine	Bovine	Squid	Duck	Chicken
Glycine	19.3–36.6	22.6–23.9	10.8–38.45	32.7–33.2	29.03–37.77	22.26–33.70
Proline	5.72–14.1	2.21–15.1	3.29–13.74	8.9–9.5	11.60–12.27	13.42–15.12
Glutamine	6.9–11.61	8.3–11.12	3.4–11.98	8.3–9.0	6.39–7.47	5.84–9.59
Hydroxyproline	5.0–9.6	9.32	10.67–11.28	7.4–8.0	10.28–11.54	11.36–12.13
Alanine	8.3–16.63	8.0–12.37	3.3–12.92	8.2–8.9	6.04–10.75	8.32–10.08
Aspartic acid/asparagine	4.4–7.2	3.49–6.76	1.7–7.46	6.1–6.5	3.18–4.12	2.11–5.91
Arginine	4.7–9.4	4.01–11.1	4.7–9.9	5.7–6.1	6.15–9.78	5.57–7.57
Serine	3.34–6.5	3.5–7.63	1.5–3.79	3.7–4.3	2.94–3.57	2.20–2.67
Leucine	1.9–2.83	2.54–3.05	1.2–3.14	2.7–3.2	3.01–3.17	2.63–3.25
Isoleucine	0.8–2.0	1.2–1.36	0.7–1.82	0.9–1.1	1.42–1.44	1.15–1.48
Lysine	1.2–4.43	2.46–3.5	1.1–4.86	1.2–1.3	2.11–2.71	3.21–4.66
Valine	1.1–3.12	2.45–4.84	1.0–2.55	2.1–3.7	1.97–2.24	1.94–2.07
Threonine	2.0–4.24	1.35–2.6	0.8–2.37	2.4–2.6	2.42–2.45	1.01–2.70
Tyrosine	0.2–0.86	0.52–0.7	0.2–1.16	0.6–0.8	0.6–0.93	0.82–1.22
Histidine	0.5–1.3	0.72–2.02	0.73–1.16	0.7–0.8	0.75–1.28	0.30–0.74
Phenylalanine	1.0–2.31	1.51–2.7	1.0–2.47	1.0	2.03–2.88	1.77–2.76
Methionine	0.7–2.06	0.61–1.0	0.22–1.69	1.0–1.3	1.05–1.92	0.07–1.12
Ammonium	–	9.60	0.59–7.68	–	–	–
Hydroxyl sine	0.5–1.43	0.19	–	0.5–1.5	–	–
Cysteine	0.1	–	0.47	–	0.16	0.16
Tryptophan	–	–	0.48	–	–	0.04

– not reported

squid skin gelatin might be due to the presence of protein fragment degradation as a result of high extraction temperature [47] that might reduce the ability of α-chains to anneal correctly by hindering the growth of existing nucleation sites during stabilization overnight. Sarbon et al. [46] reported a higher bloom value for chicken gelatin (355 ± 1.48 g) as compared to bovine gelatin (229 ± 0.71 g). Similarly, Rosli and Sarbon [65] reported that gel strength of eel skin gelatin (215.96 ± 9.62 g) was greater than bovine skin gelatin (181.28 ± 9.10 g), and this was due to intrinsic characteristic of its protein chain composition, amino acid content (especially Hyp and Pro), its collagenous properties, molecular weight distribution and the extraction methods [46]. The higher gel strength and melting temperature of eel skin gelatin are an indication of its good functional properties [65]. Chicken skin gelatin exhibited gel strength higher than other alternative sources (i.e., fish) due to high Hyp content [50]. Generally, chicken gelatin possesses a higher Pro and Hyp content as compared to the gelatins derived from any other source and thus exhibits better gel strength, melting and gelling temperatures.

Gelatin can be classified into two types: type A and type B gelatin based on the source of extraction (type A gelatin is derived from porcine sources and type B from bovine) [1, 78]. Cuttlefish skin gelatin had 19.4 g of imino acids (Pro and Hyp) per 100 g of extracted protein. The imino acid content for cuttlefish was greater than squid (17.5 g/100 g protein) and giant squid (16.3 g/100 g protein) [42, 52]. Squid gelatin showed significantly higher Hyp (8.0 g/100 g of extracted protein) and due to less hydrophobic character exhibited a significantly lower Lys (1.3 g/100 g protein) as compared to other marine sources which were in the range of 2.7–2.9 g/100 g of extracted protein [42]. The extraction yield and gel strength of skin gelatins from several marine species such as squid skin [42, 79], smooth hound [80], barbel [81], tuna [82], giant squid [52], bigeye snapper and brown stripe red snapper [33], cuttle fish [68, 83], silver carp [37] and warm water fish cod, haddock and Pollock [84] are presented in Table 2. The gelatin yields have been reported to vary among the fish species mainly due to differences in collagen amount and skin matrix [33]. Furthermore, a decrease in solubility of collagen due to high degree of cross-linking via covalent bonds might lead to reduced amount of extractable gelatin [33]. The higher temperatures used during extraction process were reported to stabilize the triple helix to a higher extent in collagen structure due to destruction of hydrogen bonds by means of higher energy, and as a result α- and β-chains were more released into the medium, ultimately leading to increased amount of gelatin obtained [55]. A lower yield (2.16% on wet weight basis) was obtained for chicken skin gelatin as compared to extracted fish gelatin (7.81–5.39% on wet weight basis) i.e., red and black tilapia, respectively [68], due to either incomplete hydrolysis of collagen or loss of collagen through leaching during series of washing steps [46].

Marine gelatins as compared to mammalian gelatins exhibit poor rheological properties, particularly gelatin from cold water fish species, such as salmon, cod and Alaska Pollock [58, 85]. This might be due to the lower number of imino acid-rich collagen regions, which lead to the formation of triple helical structures due to the formation of nucleation zones [47]. Although certain fish gelatins (tilapia, yellow fish tuna and catfish) might have similar levels of quality if not superior to mammalian gelatins, depending on processing conditions and on the species from which

Table 2 Extraction conditions, yield and gel strength of gelatin extracted from various sources

Source of gelatin	Extraction conditions	Yield (%)	Gel strength (g)	References
1-Fish and mollusca:				
Bigeye snapper skin	(1) Skin soaked in 0.2 M NaOH with skin/solution ratio of 1:10 (w/v) at 4 °C followed by washing to remove pigments and non-collagenous protein (2) Then alkali-treated skin was soaked in 0.05 M acetic acid with the same ratio at 25 °C for 3 h followed by washing (3) Soaked in distilled water with the same ratio at 45 °C for 12 h then filtered and freeze-dried	6.5 (W_w)	106	[33]
Brown stripe red snapper skin		9.4 (W_w)	219	
Lizardfish (*Saurida* spp) scales	(1) The stored frozen fish scales were thawed and treated with 2 volumes (v/w) of 0.51% NaOH solution at room temperature for 3 h to remove non-collagen protein and sub-cutaneous tissue followed by washing (2) Final extraction with distilled water at 78.5 °C for 3 h in a water bath without stirring (3) Filtration and freeze-dried	–	–	[66]
Jelly fish (*Lobonema smithii*) umbrella part	(1) Dried jelly fish soaked in HCl at 1 pH in the ratio of 1:15 (w/v) and stirred for 24 h at 4 °C and 150 rpm (2) At the end slurry was adjusted with 1 M KOH and kept in temperature controlled water bath at 45 °C for 12 h followed by filtration and drying	–	118	[34]
Cuttlefish (*Sepia pharaonis*) skin	(1) Skin soaked in 0.05 M NaOH by using sample/solution with a ratio of 1:10 (w/v) at 26–28 °C. Solution was changed to remove non-collagenous proteins then washed (2) Bleached with 5% H_2O_2 by using sample/solution with a ratio of 1:10 (w/v) for 48 h at 4 °C (3) Gelatin was extracted from bleached skin by distilled water for 12 h at 60 °C by using sample/solution with a ratio of 1:2 (w/v) (4) Extract was centrifuged at 8000*g* for 30 min at room temperature, and the supernatant was freeze-dried	13.21 (W_w)	–	[45, 67]

Table 2 (continued)

Source of gelatin	Extraction conditions	Yield (%)	Gel strength (g)	References
Sliver carp skin (*Hypophthalmichthys molitrix*)	(1) Fish skin soaked in 1/20 (w/v) 0.01 M NaOH containing 1% H_2O_2 for 24 h at 4 °C (2) Defatted with 10% isopropanol at 4 °C for 4 h. (3) Swelled with 0.05 M acetic acid for 4 h at 4 °C (4) Extracted with distilled water at 45 °C for 10 h (5) Centrifuged at 18,000*g* for 20 min at 10 °C obtained supernatant was then freeze-dried	–	–	[37]
Atlantic salmon (*Salmo salar*) skin	(1) The skin was washed with cold water then incubated twice for 30 min in cold 0.04 N NaOH (2) Followed by two successive acid incubations, firstly in 0.12 M H_2SO_4 then in 0.005 M citric acid each for 30 min (3) After final washing in cold water, gelatin was extracted by gentle stirring in water firstly at 56 °C then at 65 °C (4) Solubilized gelatin was separated, filtered, demineralized and concentrated	39.7 (D_w)	71	[50]
Atlantic cod (*Gadus morhua*) skin		44.8 (D_w)	108	
Cod (*Gadus morhua*)	(1) Skins were soaked in 0.05 M acetic acid (2) Gelatin was extracted in distilled water at 45 °C overnight, for squid a higher extraction temperature (80 °C) was used	7.2 (D_w)	72	[42]
Dover sole (*Solea vulgaris*)		8.3 (D_w)	341	
Megrim (*Lepidorhombus boscii*)		7.4 (D_w)	353	
Hake (*Merluccius merluccius*, L.)		6.5 (D_w)	103	
Squid (*Dosidicus gigas*) skins		2.6 (D_w)	10	
Giant squid (*Dosidicus gigas*) skin	(1) Skin samples were exposed to enzymatic digestion with pepsin at the rate of 1/8000 (w/w) during the swelling step in 0.5 M acetic acid for 72 h at 2 °C (2) Initial extraction was carried out in distilled water for 18 h at 60 °C, then by again soaking of collagenous residue in 0.5 M acetic acid followed by second extraction in distilled water	4–12 (D_w)	147	[52]

Table 2 (continued)

Source of gelatin	Extraction conditions	Yield (%)	Gel strength (g)	References
Black tilapia (*Oreochromis mossambicus*) Red Tilapia (*Oreochromis nilotica*)	(1) Fish skins were soaked in 0.2% (w/v) NaOH for 40 min (2) Followed by soaking in 0.2% H_2SO_4 and 1% citric acid (3) Final washing was carried out in distilled water for 12 h at 45 °C, filtered, collected and freeze-dried	5.31 (W_w) 7.81 (W_w)	181 128	[68]
Skipjack tuna (*Katsuwonus pelamis*) caudal fin	(1) Non-collagenous proteins of fins were removed by soaking the fins in 0.025 N NaOH by using a sample/solution ratio of 1/10 (w/v) for 1 h at room temperature followed by washing (2) The deproteinized samples were then demineralized with 0.6 N HCl for 5 d at room temperature followed by washing (3) Demineralized samples were then soaked in 0.2 M acetic acid for 3 h at room temperature (4) Final extraction was carried out in distilled water for 12 h at 50 °C, filtered and concentrated	1.99 (D_w)	126	[38]
Channel catfish (*Ictalurus punctatus*) skin	(1) Skin samples were soaked in 0.2 M NaOH with sample/solution ratio of 1/6 (w/v) for 84 min at 4 °C (2) Followed by soaking in 0.115 M acetic acid with 1/6 ratio (w/v) for 1 h followed by extraction with distilled water at 55 °C for 3 h	19.2 (W_w)	252	[69]
White-Cheek shark (*Carcharhinus dussumieri*) skin	(1) Skins were either cold soaked in 0.2 N HCl with sample/solution ratio of 1/3 (w/v) for 6 h or skin samples were soaked in 0.12 N NaOH (1/3, w/v) for 3 h followed by addition of 0.2 N HCl (1/3, v/w) for 3 h followed by washing (2) Finally extracted in water bath at 60 °C for 3 h followed by filtration and freeze drying	16–20 (W_w)	308–331	[56]

Table 2 (continued)

Source of gelatin	Extraction conditions	Yield (%)	Gel strength (g)	References
Clown feather back (*Chitala ornata*) skin	(1) Skin samples were pretreated with 0.1 mol L^{-1} NaOH at sample/solution ratio of (1/10, w/v) for 2 h at 15–20 °C followed by washing (2) Skin samples were then swollen by mixing with 0.05 mol L^{-1} of acetic acid with similar sample/solution ratio as above for 30 min at room temperature and 250 rpm followed by washing (3) Swollen skin was mixed with distilled water at different temperatures (45, 65 and 85 °C) at a speed of 150 rpm followed by filtration and freeze drying	–	225–284	[53]
Seabass (*Lates calcarifer*) swim bladder	(1) Alkali pretreatment with 0.1 M NaOH for 3 h at 28–30 °C with sample/solution ratio of 1/10 (w/v) (2) The deproteinized matters were then pretreatment with 0.05 M acetic acid with sample/solution ratio of 1/10 (w/v) for 2 h at room temperature until final extraction with distilled water for 6 h at 45,55, 65 and 75 °C (3) After filtration the filtrate was mixed with 1% (w/v) activated carbon for 1 h followed by centrifugation at 17,500*g* for 15 min at 25 °C (4) The supernatant was finally filtered and freeze-dried	44.83–71.95 (D_w)	188.3–280.9	[55]
Whiptail stingray (*Dasyatis brevis*) skin	(1) Alkaline pretreatment with 0.1 M NaOH for 1 h at 150 rpm (2) Acidic pretreatment with HCl or acetic acid (0.01–0.2 M) for 1 h at 125 rpm before final extraction with deionized water	5.5–7 (W_w)	380–653	[70]
Asian Swamp Eel (*Monopterus albus*) skin	(1) Skin samples were pretreated with 0.15% (w/v) NaOH and centrifuged at 4000*g* for 5 min followed by washing of resulting pellets (2) The pellets were mixed with 0.15% (v/v) H_2SO_4 then by 0.7% (w/v) citric acid followed by washing with distilled water (3) Centrifugation at 4000*g* for 10 min, final extraction in distilled water overnight at 45 °C, filtered and freeze-dried	12.75 (D_w)	215.96	[65]

Table 2 (continued)

Source of gelatin	Extraction conditions	Yield (%)	Gel strength (g)	References
2-Bovine: Bovine hide	(1) Hide was treated with 0.1 M NaOH for 6 h at room temperature to remove non-collagenous protein (2) Hide was then treated with 1% HCl for 20 h (3) Hides were then swollen with actindin enzyme for 20 h and finally sonicated for gelatin extraction.	8.64–19.65 (W_w)	451.5–627.5	[44, 59, 71]
3-Caprine: Goatskin	(1) Goatskin soaked in 3% (w/v) of sodium sulfide and 2% (w/v) calcium hydroxide to remove hair (2) Skin samples were soaked in 2% (v/v) of HCl for 48 h at 5 °C followed by washing (3) The final extraction was carried out in distilled water at 60 °C for 9 h followed by filtration and drying	10.26 (W_w)	301	[72]
4-Porcine: Pig skin	(1) After soaking the skin in isopropanol overnight alkaline pretreatment with 0.1 M NaOH at 15–20 °C for 2 h with sample/solution ratio of 1/10 (w/v) (2) Acidic pretreatment with 0.2 M hydrochloric acid for 4 h at 15–20 °C with sample/solution ratio of 1/10 (w/v) before final extraction with deionized water with 1/3 (w/v) ratio at different temperatures (55, 65 and 75 °C) followed by filtration and concentration	–	183.55–717.36	[44, 73–76]
5-Poultry: Chicken skin	(1) Defatted chicken skin was treated with 0.15% (w/v) NaOH at room temperature for 40 min followed by centrifugation at 3500–6500*g* for 10 min (2) The obtained pellets after centrifugation were treated with 0.15% (v/v) sulfuric acid after that with 0.7% (w/v) citric acid solution followed by final wash and centrifugation at 4 °C for 15 min (3) Final extraction was carried out by using distilled water overnight at 45 °C followed by filtration, concentration and freeze drying	16 (D_w)	355	[5, 46]

Table 2 (continued)

Source of gelatin	Extraction conditions	Yield (%)	Gel strength (g)	References
Broiler (*Gallus gallus domesticus*) skin	(1) NaOH (0.15%) was added into freeze-dried skin samples and blended for 30 min at 25 °C then centrifuged at 4500*g* for 10 min, resulting supernatant containing non-collagen protein was removed followed by washing of remaining pellets (2) Pellets were blended with H_2SO_4 (0.15%) at similar conditions stated above and rinsed with distilled water (3) Obtained pellets were then blended with citric acid (0.7%) at similar conditions (4) After final centrifugation at 4500*g* for 10 min, the extraction was carried out at 50 °C overnight followed by filtration, ion exchange and freeze drying	6.5 (W_w)	166.65	[35]
Duck feet	(1) The duck feet stored at −20 °C were soaked in soaking solution (0.1 N HCl, 40% NaOH and distilled water) for 24 h at 18 °C with varying pH values, followed by neutralization by using tap water at 18 °C for 48 h (2) The fully soaked duck feet were drained and stored in polyethylene bags and vacuum packed (3) Followed by extraction by either heating the sample in water bath at 95 °C for 2 h or heating in pressure cooker for 70 min at 88.2 kPa pressure or heating in a microwave oven at 350 W power for 5 min, finally filtered and freeze-dried	17.58–51.83 (D_w)	190–700	[77]

– not calculated. Whereas D_w indicates percentage yield on dry weight basis, while W_w represents percentage yield on wet weight basis

gelatin is extracted [58, 85], fish gelatin when compared to mammalian gelatin melts at a lower temperature due to low content of imino acids (Pro plus Hyp) [86], which could be due to the fact that imino acids stabilize the ordered structure of gelatin in the gel form, thus ultimately reducing the gel modulus [85].

Avena-Bustillos et al. [86] reported that WVP of cold water fish gelatin films was found to be significantly lower (0.932 g mm m^{-2} h kPa) as compared to hot water fish or mammalian gelatin films (1.309 and 1.884 g mm m^{-2} h kPa, respectively) due to higher amount of hydrophobic amino acids and lower amounts of Hyp. Sobral et al. [44] reported that mammalian skin gelatin films were mechanically stronger and more permeable to water vapors as compared to fish skin gelatin films, whereas fish skin gelatin films showed better elasticity. Furthermore, imino acid content of bovine skin gelatin (10.67%) was reported to be lower than chicken skin gelatin (12.66%) leading to lower melting and gelling temperatures (31.55 and 24.43 °C, respectively) of bovine gelatin as compared to chicken gelatin (33.57 and 24.28 °C, respectively). Thus, chicken gelatin exhibited higher viscous and elastic modulus values as compared to bovine gelatin for a wide range of frequencies and concentrations [46].

Influence of NPs on gelatin composite films

Films formulated by using gelatin as primary biopolymer are more advantageous to produce due to gelatin availability at low cost and better functional characteristics [87]. Gelatin has been widely used as an alternative to plastic-based packaging because of its unique characteristics as compared to other proteins and polysaccharides, i.e., carrier of bioactive compounds, film-forming ability and barrier properties [12]. The barrier and mechanical properties of gelatin-based films mainly depend on the chemical and physical characteristics of gelatin which are determined by amino acid composition, molecular weight distribution and extraction conditions of gelatin [12]. On the other hand, because of hygroscopic nature of gelatin the gelatin-based film tends to swell by absorbing moisture when comes in contact with the food products with high moisture content [1, 12]. Gelatin has been reported to improve mechanical and physicochemical properties of composite films with other biopolymers, i.e., chitosan, as compared to plain gelatin or chitosan films [88]. The incorporation of NPs in gelatin film matrix improves the barrier, optical and mechanical properties of gelatin films (Fig. 1). The presence of NPs in gelatin film matrix provides a physical barrier that restricts the penetration of light, oxygen and water vapors across the polymeric matrix. Neat gelatin films provide barrier and mechanic protection to the packaged materials; however, neat gelatin films lack in antimicrobial and antioxidant potential. The addition of NPs to neat gelatin films resulted in the entrapment of NPs in film matrix which improved the film barrier and mechanical properties. Moreover, the addition of NPs (i.e., ZnO and silver) to gelatin film matrix imparts antimicrobial potential and results in the development of active packaging [1, 15]. The effects of NPs on the mechanical and optical properties of gelatin films are summarized in Table 3.

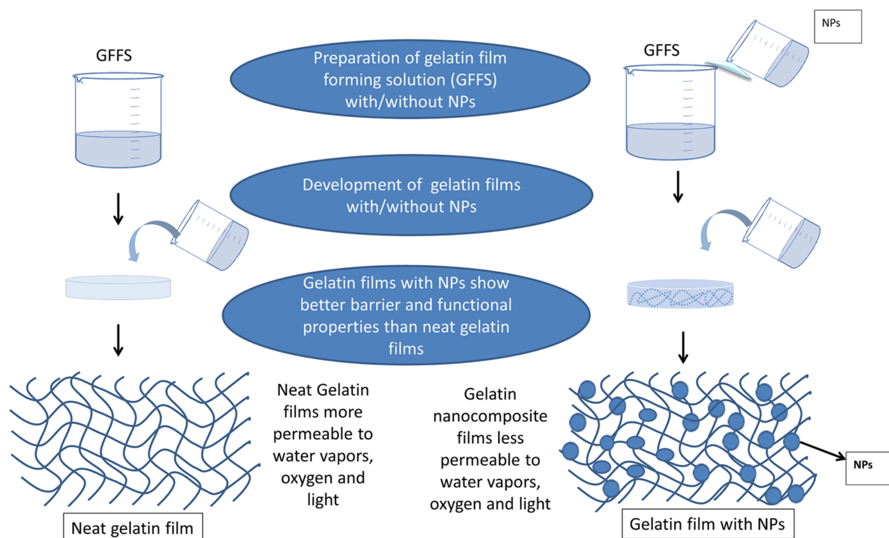

Fig. 1 Influence of nanoparticles (NPs) on gelatin composite films

Water vapor permeability and surface hydrophobicity

It is important to have an understanding of barrier characteristics and moisture content of food packaging. One of the permeability characteristics of food packaging materials is WVP. The water vapor barrier properties of packaged products have great importance in maintaining and extending the product's shelf life as the WVP and moisture content are directly related to physical and chemical deterioration of the products [11]. Generally, hydrophobic NPs and their interaction with biopolymer, i.e., gelatin, can create zig-zag pathways across water vapors leading to improved barrier properties, while hydrophilic organic NPs can create water clusters when employed into a polymer (i.e., gelatin) to form film, thus helping water diffusion through gelatin film matrix [18]. When hydrophilic NPs are used in high concentration, they tend to aggregate and make the gelatin films more permeable to water vapor [18]. The increased hydrophobicity of gelatin nanocomposites was mainly due to hydrophobic character of NPs [22]. The water contact angle (WCA) is dependent on the adhesive and cohesive molecular forces within the water and between film surface and water, respectively [16]. WCA above 65° is a characteristic of surfaces that are hydrophobic, while WCA below 65° is typical for hydrophilic surfaces [97]. It has been reported that roughness of the material surface is responsible for hydrophilic behavior of the material [16]. However, surface hydrophobicity of gelatin nanocomposite is not only influenced by the interaction between biopolymer chain and NPs, but also individual hydrophobicity of NPs and biopolymers [18].

Gelatin films containing ZnO NPs showed increase in WVP from 6.637 to 8.157×10^{-10} g^{-1} s^{-1} Pa^{-1} with increase in ZnO NPs concentration from 1 to 5%, respectively, but still remained lower than the control gelatin films ($8.478 \pm 0.448 \times 1$

Table 3 Characteristics of gelatin films incorporated with nanoparticles (NPs)

Gelatin nanocomposite composition	Thickness (mm)	TS (MPa)	EAB %	Transparency	ΔE	WCA°	References
Gelatin (4%) + ZnO NPs (1–5% based on total gelatin content) and nanochitin (3–10% based on total gelatin)	0.100–0.160	28.47–119.1	4.63–13.63	1.42–4.15	–	–	[18]
Gelatin type A (3.33%) + melanin NPs (0–1wt % of gelatin)	0.068–0.077	33.6–46.5	9.7–15	–	0.4–65.1	56.8–66.8	[22]
Gelatin (pig skin) type A (5%) + MMT NPs (0–10% of gelatin)	0.071–0.080	23.5–31.7	38–48.7	–	–	82.9–94.6	[16]
Gelatin (cold fish skin) (3%) + silver–copper NPs (0–4% of gelatin w/w)	0.061–0.098	19.2–28.6	32.9–54.2	1.45–8.22	3.17–63.85	–	[13]
Gelatin (bovine hide) type B (10%) + Na^+, H^+ and Cu(II) exchanged MMT NPs (5% w/w of gelatin)	0.16–0.21	3.9–10.9	48.4–96.9	–	–	–	[89]
Gelatin (pig skin) type A (90–100% of total solid content) + nanocellulose (0–10 wt% of total solid) and MMT NPs (0–10 wt% of total solid)	0.049–0.082	32–75	9–20	–	–	–	[90]
Gelatin (cold fish skin) (4%) + chitosan NPs (0–8% w/w of dry gelatin) Without essential oil	0.050–0.064	7.44–11.28	32.73–102.04	0.97–3.65	–	45.57–97.12	[91]
Gelatin (cold fish skin) (4%) + chitosan NPs (6% w/w of dry gelatin) With oregano essential oil (*Origanum vulgare* L.) (0.4–1.2% w/v)	0.062–0.089	3.28–10.57	44.71–151.82	–	–	58.15–80.22	[24, 92]
Gelatin (cold fish skin) (4% w/v) + agar (1.5% w/v) + TiO_2 NPs (0–2% w/w)	–	–	–	7.15–17.89	5.31–16.1	–	[21]
Gelatin (5%) + silver nanocolloid (1–4 mM)	0.0682–0.073	46.12–56.55	37.99–47.23	–	4.5–68.7	–	[17, 93, 94]
Gelatin (2%) + chitosan (2%) + silver NPs (0.05–0.1%)	0.083–0.092	21.19–28.87	17.99–27.23	0.81–3.87	–	–	
Gelatin (3.33%) + silver NPs (0–40 mg)	–	26.3–35	45.6–60.5	–	2.1–61.4	43.6–48.5	
Gelatin (Reagent grade) (5%) + guar gum benzoate NPs (5–20% w/w of gelatin)	0.345–0.392	2.25–3.75	1.0–4.0	2.08–4.69	–	–	[95]
Gelatin (2.66%) + NPs (1 mM) of gold/silver/copper/bimetallic	0.0531–0.0568	87.3–98.1	8.3–9.0	–	2.19–72.49	48.1–56.9	[20]
Gelatin type A (1% w/v) + chitosan (1.5% w/v) + ZnO NPs (0–10 mg)	–	–	–	1.65–2.21	–	–	[96]

– not reported, *NPs* nanoparticles

0^{-10} g^{-1} s^{-1} Pa^{-1}) without NPs. This can be explained due to the hydrophobic nature of ZnO NPs which provides a tortuous pass-way for water vapor through film matrix because of increased crystallinity of biopolymer or by reducing free hydrophilic groups (OH, NH), thus improving barrier properties [18]. The combined influence of ZnO NPs (i.e., from 1 to 5%) was observed with the addition of 3% chitin NPs in gelatin FFS on WVP, which was increased from 5.943 to 7.195×10^{-10} g^{-1} s^{-1} Pa^{-1}. However, chitin NPs did not present any significant influence on WVP when employed alone due to its hydrophilic nature [18]. The application of hydrophilic chitin NPs in gelatin films can create water clusters across the structure of film, helping water diffusion through the matrix of film [18, 98]. It has been previously reported that high concentration of chitin NPs increased the hydrophilic character of films, thus leading to more diffusion of water [99].

Shankar et al. [22] reported that the WVP of gelatin films incorporated with melanin NPs decreased significantly (from 1.04 to 0.78×10^{-9}g m^{-1} m^2 Pa s) with increasing concentrations of NPs from 0 to 1% in FFS. The compact structure was formed due to increased hydrogen bonds in biopolymeric chains, thus causing reduction in WVP of gelatin films with the addition of melanin NPs. Martucci and Ruseckaite [89] prepared copper (II)-exchanged MMT/gelatin nanocomposites with reduced WVP as compared to control gelatin film. The water uptake by capillarity at interface was reduced, due to consumption of hydrophilic groups by strong interactions between gelatin and MMT nanoclay. The delay in the transmission of water vapor across the polymer matrix might be due to presence of water vapor impermeable silicate platelets of nanoclay which are dispersed in the polymer matrix, thus obstructing the pathway of water vapor [17]. Similarly, TiO_2 NPs (0–5%) reduced the WVP of the bovine gelatin nanocomposites significantly from 8.90 to 1.61×10^{-11} g m^{-1} s^{-1} Pa^{-1} due to hindrance in the gelatin matrix by greater water resistance of TiO_2 NPs because of their hydrophobicity [100].

WCA is one of the basic wetting properties of nanocomposite films, which is used as an indicator for hydrophobic/hydrophilic properties of film surface. The WCA for gelatin composite films incorporated with melanin NPs increased (from 55.8° to 66.8°) with increase in the concentration of NPs up to 0.5%, followed by a decrease (62.2°) with increase in NPs from 0.5 to 1% [22]. Jorge et al. [16] reported an increase in WCA (from 90.3° to 94.6°) with increase in MMT NPs concentration up to 5 g/100 g gelatin due to increased hydrophobicity of the gelatin nanocomposites by the incorporation of hydrophobic NPs.

Thickness and mechanical properties

The biopolymeric film thickness is mainly influenced by solid content of FFS [101]. Addition of NPs into the gelatin FFS can increase the thickness of resulting gelatin nanocomposites as compared to control gelatin films [95]. The increase in concentration of NPs linearly increases the TS of gelatin composite films up to a certain critical concentration of NPs and decreases thereafter. The addition of NPs develops a strong network with gelatin matrix up to a critical concentration, and further increase in concentration of NPs results in overloading and jamming into the gelatin

matrix. Thus, the distribution of additional NPs lacks uniformity and decreases the mechanical properties [13, 18], while EAB decreased linearly with increasing concentration of NP due to improvement in rigidity of films. Furthermore, certain NPs due to their hydrophobic nature tend to aggregate and could impede the protein domains of gelatin films leading to reduced mechanical properties [13]. The thickness of gelatin nanocomposite films incorporated with silver nanocolloid and silver NP varied between 0.0682–0.0732 mm and 0.0834–0.0922 mm, respectively [17, 93]. The thickness of gelatin films incorporated with melanin NPs increased (from 0.0682 to 0.0776 mm) because of solid content by means of NPs incorporation [22].

Sufficient flexibility and mechanical strength are necessary for packaging film to endure external pressure as well as environmental stress and to maintain its integrity and barrier properties during packaging [11]. The TS of control gelatin films was decreased significantly from 65.199 ± 5.190 MPa to 29.32 ± 1.24 MPa when the concentration of incorporated ZnO NPs was increased up to 5%, while the EAB of control gelatin films was increased from 4.636 ± 0.371 to 12.963 ± 1.570% after incorporation of ZnO NPs [18]. It has been reported that mechanical properties of the films are influenced by both intermolecular and intramolecular interactions of polymeric chains. Shankar et al. [22] reported that TS of gelatin films reinforced with melanin NPs increased with increase in NPs concentration up to 0.5% (from 33.6 to 46.5 MPa) and decreased (from 46.5 to 41.8 MPa) when concentration of NPs was increased from 0.5% to 1%. EAB was decreased (from 15 to 9.7%) with increasing concentration of NPs from 0 to 1%. The increased strength and decreased flexibility of gelatin nanocomposite can be attributed to H-bonds between the melanin and quinone-amino group of gelatin [22]. For MMT/gelatin nanocomposites, TS was increased from 23.5 to 31.7 MPa with increase in MMT NPs concentration (0–5%). While EAB was decreased from 48.7 to 38.0% with increasing content of MMT from 0 to 10% due to reinforcement of polymeric matrix by the addition of NPs [16]. It has been reported that addition of chitosan NPs increased the TS of gelatin films and made them stiffer confirming the reinforcing effect of NPs on the film matrix [24], while a decrease in EAB was found with increasing content of chitosan NPs which indicates brittleness of gelatin films due to decrease in free volume by strong intermolecular attractive forces [24].

Optical properties

Color of films is an important factor which influences the demand of consumers. The addition of NPs into gelatin FFS mostly decreased the lightness (L value) and increased the yellowness (b value), redness (a value) and ΔE (total color difference) as compared to control gelatin films, while the L value decreased linearly with increasing concentration of NPs [22]. The change in color of gelatin films from colorless to yellowish-brown color is an indication of successful production and incorporation of NPs into gelatin film matrix [17]. It has been reported that protein-based biopolymers, i.e., gelatin, exhibit excellent barrier properties against UV light because of the presence of aromatic amino acids (mainly Tyr and tryptophan, while phenylalanine and disulfide bonds to a lesser extent) in its structure [24].

Furthermore, addition of NPs into gelatin films reduced light transmission through the film matrix, especially when NPs are well dispersed throughout the biopolymer; ultimately gelatin films having enhanced light barrier properties with ability to block and scatter light are obtained [18]. Generally, gelatin nanocomposites are opaque, while neat gelatin films are colorless; thus, opaque films can limit oxidation and discoloration induced by light by reducing visible and UV transmission [102]. It has been reported that transparency of gelatin films decreased significantly with increasing concentration of NPs due to resistance shown by NPs impregnated within film matrix to passage of light [93].

Kanmani and Rhim [17] have reported an increase in ΔE values (from 2.1 to 61.4) of gelatin films with increasing concentration of silver NPs from 10 to 40 mg into FFS. The films containing only gelatin usually exhibit very low values (0.4–5.35) for ΔE and are almost transparent, which is a characteristic of neat gelatin films [17, 22, 93]. It has been reported that gelatin/silver nanocomposite films revealed pale yellow to brownish yellow color depending on the concentration of NPs in FFS [17]. This might help in preventing the penetration of UV and visible light into the food packaging film and thus help in the retention of nutrients, flavor and color [17, 89].

One of the functions of food packaging films is to protect the food from effects of light, especially UV rays [11]. Transparency of gelatin films was reported to be greatly influenced by the addition of NPs as gelatin nanocomposites were opaque as compared to neat gelatin films (i.e., which are almost transparent) after incorporation of NPs [17]. Arfat et al. [13] reported that transparency values increased (from 1.45 to 8.22) with the increasing concentration of silver–copper NPs from 0.5 to 4% in gelatin nanocomposites. Thus, addition of NPs in composite films can play an important role by blocking UV light through the film [103]. Various researchers reported elevated composite film transparency values (i.e., higher transparency values lead to more opaque films) with increasing concentration of chitosan, silver, guar gum benzoate and ZnO NPs [24, 86, 89, 91]. So, it can be concluded that gelatin nanocomposite films with strong light and UV barrier properties can be used as a packaging material for oxidation-sensitive foods.

Thermal properties

The state of the polymer (i.e., glassy or rubbery) influences the thermal properties of the film [11], and it is therefore important to evaluate the glass transition temperature (T_g) of the packaging films. T_g is a value that corresponds with system mobility, and it is acquired as a central point of endothermic transition and can be defined as a temperature at which an amorphous material morphs into rubbery state from glassy state. T_g is an essential parameter for the selection of processing and storage conditions of the film along with the application of films [11]. Differential scanning calorimetry (DSC) is one of the methods that had been applied to study semi-crystalline materials. DSC is widely used to verify the T_g of the materials [11].

Shankar et al. [104] reported that ZnO NPs can act as a heat insulator and thus can increase heat stability of gelatin films. Generally, addition of NPs into polymer matrix can enhance the thermal properties of gelatin nanocomposites, because they

can increase the crystallinity of the biopolymer and can instigate ordered and compressive crystals (larger crystals have higher thermodynamic stability) [18]. Previous studies reported improved thermal stability of gelatin films after incorporation of NPs/nanoclays leading to higher heat resistance and higher onset temperatures required for thermal degradation as compared to neat/single gelatin films [24, 93, 94].

Arfat et al. [13] reported increased T_g during second heating scan for gelatin nanocomposite (i.e., 2% silver–copper NPs) from 50.32 to 89 °C that could be attributed to removal of absorbed water during first heating scan from gelatin films, thus leading to enhanced interactions between polymer chains and NPs; consequently, films with rigid network were formed. During the first scan gelatin nanocomposite incorporated with MMT showed increase in T_g with increasing content of MMT in FFS due to crystalline nature of the material, while no significant difference was observed during second heating scan due to amorphous nature of material [16]. Hosseini et al. [92] on the other hand reported increase in T_g of gelatin films incorporated with chitosan NPs from 2 to 6% concentration followed by complete disappearance of T_g above 6% concentration and was not observed on the film thermograms as it depends upon a number of factors such as the intermolecular interactions, molecular weight, chain flexibility, steric effects, cross-linking density and branching.

Chemical and crystalline structure

Film structure is an important parameter for film production because of its potential industrial usage and its contribution to the physicochemical properties of the film. X-ray diffraction (XRD) analysis is carried out to study the amorphous and crystalline structure of the biopolymers used as film materials. Protein-based biopolymers, i.e., gelatin, usually presents a predominant peak at $2\theta = 20°$ which is characteristic of an amorphous phase [13]. On the other hand, gelatin nanocomposites present a diffraction peak at $2\theta = 7°$ which is a characteristic of a crystalline triple-helix structure of gelatin. The overall crystallinity of gelatin nanocomposite is increased after incorporation of NPs, thus increasing major peak intensity [18, 83].

Two characteristic peaks were observed in neat gelatin film diffractogram at 2θ of 7° and 20° [16, 83, 105]. The first peak at 7° indicates crystalline structure of gelatin, and the second peak at 20° indicates amorphous phase of the composite films [106]. Extra peaks have appeared in diffractogram of the biopolymers by the incorporation of NPs into neat gelatin films between 31° and 79° of 2θ corresponding to the plane of different NPs [13, 18, 92]. The intensity of characteristic peak of gelatin (2θ at 7°) was reduced by the addition of chitin and ZnO NPs into gelatin films leading to reduced crystallinity [18]. This might be due to decrease of α-helix structure of gelatin chains by the interaction of NPs with gelatin [107].

Evaluation of film microstructure is of great value as it was used to determine barrier, physicochemical and mechanical properties of the film [108]. Scanning electron microscopy (SEM) was used to study the microstructural changes in the gelatin films and to evaluate cross-sectional and surface topography of the film samples.

Neat gelatin films were found to be smooth, without pores with excellent structural integrity when observed through SEM [13, 18, 24]. On the other hand, gelatin/nanocomposite films containing NPs were reported to have rough surfaces even though NPs were evenly distributed [13, 15, 18] leading to enhanced thermal stability and mechanical strength [18, 86]. Arfat et al. [13] reported that film surface became less homogenous when silver–copper NP concentration was increased above 2% leading to appearance of micro-cracks. This was mainly due to high concentration of NP/nanoclay leading to agglomeration or aggregation [16, 24, 109]. Such inhomogeneous structures can lead to poorer film properties, i.e., TS. On the other hand, well-dispersed NPs in the film matrix can reduce WVP by creating convoluted pathway across the film [18].

The analysis of chemical structure through Fourier transform infrared spectroscopy (FTIR) is crucial in film production as it is used to determine specific functional groups that impart specific functions to the gelatin films [11]. Generally, specific amide bands are associated with triple-helix structure in gelatin spectra [18]. Therefore peaks ranging between (3000 and 3500 cm^{-1}) are related to amide-A and amide-B which correspond to OH and NH stretching vibrations followed by amide-I (1600–1690 cm^{-1}) C=O stretching vibration, amide-II (1480–1575 cm^{-1}) NH stretching and amide-III (1229–1301 cm^{-1}) CN stretching and NH bending frequency [16, 18, 24, 83]. Additionally, amide-III peak can also be associated with Pro and Gly backbone of the chain [110].

It has been reported that there was a shift of NH stretching of amide-A band from 3500 cm^{-1} to higher or lower wave number due to incorporation of NPs, indicating the formation of possible hydrogen bonding between polymer matrix and NPs [13, 18, 24]. A higher wave number of amide-A band was reported with increasing content of NPs for gelatin nanocomposites incorporated with chitosan and ZnO NPs between 3295 and 3646 cm^{-1} [18, 24]. Similarly, amide-I, amide-II and amide-III have also been reported to be influenced by NPs concentration in composite films [13, 17, 18]. Addition of MMT clay shifted amide-I, amide-II and amide-III to higher frequencies confirming the hydrogen bonding between acceptor atoms such as oxygen from free hydroxyl group and Si–O–Si groups in MMT [89].

Antibacterial properties

Microbial contamination can decrease the shelf life of food/feed products and increase the risks of various food-borne infections. The antimicrobial active packaging is one of the promising technologies which is achieved by incorporation of potent antimicrobial agents (i.e., organic and inorganic NPs) into biopolymer matrix [17, 24]. Gelatin itself does not have any antimicrobial potential and in order to develop an active antimicrobial packaging, antimicrobial agents, i.e., NPs and natural preservatives are added into gelatin matrix. Generally, different inorganic NPs impart their antibacterial activity by targeting different cellular organelles of food-borne bacteria. However, NPs mainly accumulate in bacterial cytoplasmic membranes and cause a significant increase in membrane permeability, leading to cell death [20]. Among inorganic NPs (i.e., gold, silver, copper and zinc), reinforcement

of silver NPs in gelatin matrix has been reported to exhibit broad spectrum of antimicrobial effect against food-borne pathogens (i.e., *Escherichia coli*, *Salmonella typhimurium*, *Staphylococcus aureus* and *Bacillus cereus*) [17, 20]. Silver NPs in nanocomposites are believed to bind with negatively charge cell membrane of bacteria, which disrupts surface proteins and cell walls, inactivates enzymes and produce H_2O_2, leading to cell death. Gelatin-based silver nanocomposites are reported to be more effective against gram-negative bacteria because of their surface charge and thin cell wall [17, 20]. Copper NPs in gelatin nanocomposites inhibit the bacterial growth due to inhibition of DNA replication and destruction of bacterial cell wall [20]. TiO_2 NPs in nanocomposites can generate oxygen reactive species after entering the bacterial cell membrane and oxidize polyunsaturated phospholipid components of cell membrane [21, 111]. Alternatively, organic NPs are found to be less effective against food-borne pathogens as compared to inorganic counterparts because of their low stability [17, 91]. Gelatin nanocomposites can increase the shelf life of food products due to their antibacterial potential imparted by the reinforcement of NPs as compared to neat gelatin films.

Biodegradation of gelatin composite films

The neat gelatin films were reported to degrade 60% after 30 days of incubation under simulating soil burial conditions [112]. The biodegradation of polymeric matrix depends on the length of polymer chain, complexity of the matrix and degree of crystallinity. The biodegradation of a polymer increases with decrease in length of polymer chain, lack of complexity and crystallinity. Gelatin film stability in simulated body fluids was found to increase with the addition of NPs. After incorporation of NPs into gelatin matrix a decrease in the rate of biodegradation of gelatin nanocomposites was reported as compared to neat gelatin films due to the fact that NPs reinforced the gelatin structure in a way that amount of non-reinforced gelatin was decreased (since gelatin became more crystalline, it was difficult to degrade crystalline phase as compared to amorphous phase), furthermore, it might also be associated with the surface reaction of gelatin nanocomposites with the medium in which degradation was observed [113, 114].

Safety concerns and limitation of NPs in packaging

Safety of NPs has always been a concern in packaging, particularly in food packaging due to direct contact of NPs with food materials [17]. Despite from the beneficial effects of various NPs, the safety level of NPs should be evaluated by clinical studies before commercial application. The potential health risks associated with the consumption of food products containing nanofillers transferred from the packaging are not yet fully understood and mainly depend on the particle size, toxicity, morphology, rates of migration and ingestion of NPs [114]. Several metal oxides (i.e., TiO_2 and ZnO) used in gelatin-based food packaging were reported to be safe and approved by Food and Drug Administration (FDA) because of their broad range

of biological activities [111, 115]. On the other hand, ZnO NPs have been extensively used and reported to be safe for human consumption, but ZnO NPs were also reported to be highly toxic at a concentration of ≥ 15 ppm [116, 117]. For CuO NPs, European Food Safety Authority (ESFA) described the maximum level of release in food materials, which should not be more than 10 mg kg^{-1} [14]. The lack of information on risk assessment of NPs used in packaging is mainly due to the absence of studies on migration assays and in vivo experimentations, which needs to be explored for better understanding.

Conclusion

Gelatin-based nanocomposite films are gaining interest over the other biopolymeric films due to better barrier and functional properties. The incorporation of NPs in the gelatin films leads to modification in physical, mechanical and barrier properties. Therefore, a better understanding of the interactions between biopolymer nature and nanoparticles can lead to the formulation of biodegradable packaging films with better preservation potential. The gelatin as a biopolymer exhibits advantages due to its cheap cost, wide variety of sources and improved functional characteristics after incorporation of wide variety of NPs.

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Compliance with ethical standards

Conflict of interest The authors declare no conflict of interest.

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