Review



Nanoedible films for food packaging: a review

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ABSTRACT

Edible packaging is a thin layer formed on food surface, which can be eaten as an integral part of the food product. While an edible coating is formed as thin layer directly on the food surface for improving shelf life of fruits and vegetables, the edible film is formed as thin layer separately and wrapped on food surface later. The edible films have attracted much interest as it has potential to overcome the problems associated with plastic packaging. However, their film properties are not as good as the conventional packaging materials, such as plastics. The food and beverage industry is showing much interest to incorporate the benefits of nanotechnology. The nanomaterials have unique characteristics (such as, large surface area-to-volume ratio, distinct optical behaviour and high mechanical strength), which, when incorporated with the edible films, could improve the film properties of the edible films. Therefore, the right selection and incorporation of nanomaterials can improve the film properties. Most of the previous review articles on food packaging summarized the research findings of synthetic and/or biodegradable films and coatings. Only few review articles were devoted for edible films and coatings. Among them, very few review articles had discussion about the use of nanotechnology for all kinds of food packaging applications. However, there is no comprehensive review on nanoedible films. The objective of this review article is to cover the recent works on nanoedible films prepared incorporating the nanofillers (such as, nanostarch, nanocellulose, nanochitosan/nanochitin, nanoproteins and nanolipids), the film properties (such as, the mechanical properties, WVP and film colour of some of the recent nanoedible films), and the challenges and opportunities for future research.

Introduction

Food packaging is primarily used to store the food items in a cost-effective way, protect them from environmental influences or damage during transportation and maintain food quality from the time of packaging to the time of consumption [1, 2]. The most used packaging materials, according to FICCI, are plastics (42%), paper board (31%), metals (15%), glass (7%) and other materials (5%). Among

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these, plastics packaging has become the dominant packaging material, due to their high cost-to-performance ratio, light weight and convenience [3]. Food packaging is the largest and fast growing sector within the plastic packaging. It was forecasted for the period of 2003-2009 that 54% of plastic packaging would be consumed by food and beverages sectors, 40% and 14%, respectively [4]. Plastics used for food packaging are usually discarded after consumption of food, with a very limited quantity being recycled. The plastics waste, if recycled, might reduce the landfills. However, a little amount of plastics are only recovered through recycling. If not recycled, plastics are either dumped in landfills where they last forever or burnt for energy recovery (incineration) producing harmful gases causing air pollution. The space availability for landfills is a major issue in many countries, especially in highly populous cities. Another problem with plastics is its dependency on petroleum reserves [5]. Biodegradable plastics, on the other hand, decompose plastic molecules into carbon dioxide, water and other by-products like methane. Since landfills reduce the presence of oxygen and moisture which are essential for biodegradation, biodegradability is affected. Moreover, land space is still required for storing wastes until they are fully decomposed. Therefore, biodegradable materials show little impact on reducing landfills [1, 6].

Edible packaging is a thin layer (film or coating), formed on food surface, which can be eaten as an integral part of the food product. If the thin layer is formed directly on the food surface, it is termed as coating. If the layer is formed separately and

wrapped on food surface later, it is termed as film. Edible films and coatings are formed only when the packaging material can form a continuous and cohesive structure. Polymers and composites, that are edible and capable of forming such continuous cohesive structures, qualify as edible packaging materials [7]. As these edible packages can be consumed along with food or beverages, nothing will be left out for disposal. They do not harm the environment even if not consumed. As they are produced from food ingredients, the edible packaging degrades more readily than synthetic and biodegradable packaging materials. As a result, edible packaging materials have attracted researchers' attention as the promising alternative that has bright potential to replace synthetic as well as biodegradables plastics [8].

Edible films are produced from edible polymers which are categorized into polysaccharides, proteins and lipids. A list of the widely used edible polymers for making edible films is shown in Fig. 1. There is a basic difference between polysaccharides and proteins. Polysaccharides are homopolymers consisting of a monomer (glucose, for example) of repeating units, while proteins are heteropolymers consisting of more than at least 20 amino acids. Polysaccharides are better gas barriers but poor water vapour barriers. Proteins are also poor water vapour barriers, but show better mechanical strength. Unlike polysaccharides and proteins, lipids do not form self-supporting film structures, but are good water vapour barriers. So, lipids can be used either used as coating material that is directly applied onto the food surface,



or as an additive added to a self-supporting emulsion films containing polysaccharides or proteins for improving the hydrophobic nature and gloss of the resulting composite films. The edible composite film, where the type and amount of polymers (polysaccharides, proteins and lipids) are appropriately selected, could be made in order to obtain the improved film properties [9, 10].

The food and beverage industry is showing much interest to add the benefits of nanotechnology. With nanotechnology, one can achieve lightweight packaging material with stronger barrier properties, which can protect the food quality during packaging, transportation and consumption and can preserve meat or poultry products from the spoiling pathogens. The nanomaterials of size ranging from 10-100 nm are added to several kinds of packaging materials, and alternative packaging materials with improved film properties (for instance, barrier properties and mechanical strength) can be produced [11–13]. The unique characteristics of nanomaterials, which are possible only at nanosize, make them so interesting. The materials at nanoscale offer entirely different physical and chemical properties as compared to their micro- and macroscopic counterparts. Some of the unique properties of nanomaterials, such as, large surface area-to-volume ratio, distinct optical behaviour and high mechanical strength make them suitable for the packaging industry. The incorporation of the right nanomaterials in compatible polymers can offer superior mechanical and barrier properties, thermal stability and better optical properties of the packaging materials as compared to those of the conventional packaging materials [14–17]. The use of nanotechnology for food packaging has been reviewed by some of the researchers [18-28]. Almost all these articles focused on the synthetic as well as biodegradable films and coatings. None of these articles gave a comprehensive review on nanoedible films. Even though few of them included the details of the nanoedible films, they could only provide the brief note. Because of the scarcity of information about the use of nanotechnology for food packaging, it would be useful for the research community if the recent research works on nanoedible films are summarized. This article is therefore aimed to summarize the research findings of nanoedible films in recent times, problems against their progression and opportunities for future research.

Nanostarch reinforced edible films

Starch structure generally consists of starch granules (2-100 µm) as white powder, which is made of growth rings (120-500 nm). These growth rings are composed of blocklets (20-50 nm), which consisted of amorphous and crystalline lamellae (9 nm) into which we find amylopectin and amylose chains (0.1–1 nm) [29]. The first report on isolation of starch nanocrystals was found in 1996 by Dufresne et al. [30], in which acid hydrolysis of potato starch produced the so called then microcrystals of diameter a few tens of nanometres. In another work by Putaux et al. [31], starch nanocrystals were isolated from the native maize granules using acid hydrolysis. After 6 weeks of acid hydrolysis, the starch nanocrystals were produced with width of 5-7 nm and 30-45%crystallinity. In the work of Kim et al. [32], starch nanocrystals were produced through acid hydrolysis followed by centrifugation from various starch sources, such as waxy maize, normal maize, high amylose maize, potato and mung bean. The size of the nanocrystals varied from about 40-70 nm diameters with round or oval shapes. Starch nanocrystals typically exhibit the crystallinity of about 45%, as opposed to cellulose nanocrystals that exhibit the crystallinity close to 100%. Isolation of starch nanocrystals has been reported with various isolation methods, such as, acid hydrolysis [33-37], enzymatic hydrolysis [38], high-pressure homogenization [39, 40], ultrasonication [41] and nanoprecipitation [42].

Angellier et al. [43] studied the effects of the ageing of the waxy maize starch edible films reinforced with the waxy maize starch nanocrystals. During the 2 weeks of aging, the mechanical properties of the waxy maize starch nanocrystals reinforced edible films changed significantly. Ageing increased TS and YM while decreasing %E. The mechanical properties of non-aged starch films of 10 wt% starch nanocrystals are close to those of the aged starch films. The nanocrystals above 10 wt% resulted in the high YM and TS (after aging), with %E remaining almost a constant about 20%. Shi et al. [44] produced starch nanoparticles loaded corn starch-based edible films and investigated the effects of drying methods on physical and mechanical properties of the prepared films. Emulsion cross-linking method with the help of a high-pressure homogenizer, followed by two drying techniques, namely spray drying and vacuum freeze drying, was used for producing the nanoparticles. The results showed that the addition of starch nanoparticles, prepared by both drying techniques, in the corn starch films increased the surface roughness, reduced the degree of crystallinity by 23.5%, reduced WVP by 44% and reduced the glass transition temperature by 4.3 C, respectively, as compared to those of control films. The addition of corn starch nanoparticles slightly increased the creep strain, creep compliance and creep rate, but did not significantly different statistically. Drying technique did not affect these properties significantly. The drying method affected only the opacity of the starch films. The opacity of starch films containing the vacuum freeze-dried nanoparticles was found to be higher (30.499 AU nm), as compared to that of the starch films containing the spray-dried starch nanoparticles (23.965 AU nm).

Piyada et al. [45] prepared solution-cast rice starchbased film dispersed with rice starch nanocrystals (5 to 30%) and found that the addition of rice starch nanocrystals improved the mechanical properties, WVP and thermal stability of the rice starch edible films. The edible films containing up to 20% of rice starch nanoparticles exhibited the best mechanical properties, such as TS (16.43 MPa) and % E (5.76%), respectively. Li et al. [46] produced the pea starch edible composite films loaded with starch (waxy maize) nanocrystals and investigated the effects of the concentrations (1-9%) of starch nanocrystals on WVP and mechanical properties. The results showed that the moisture content and WVP of the composite films decreased significantly, while TS and YM increased with the increase of the concentration of starch nanocrystals up to 5%. The films with up to 5%starch nanocrystals produced smooth and dense surface. However, for the composite films produced with the concentrations of the starch nanocrystals above 7%, the longitudinal fibrous structure was developed on the film surface as the starch nanocrystals started to aggregate. Dai et al. [47] produced the nanoedible films from corn starch reinforced with varying proportions of taro starch nanoparticles (0.5–15%). The results showed that the addition of taro starch improved WVP, TS (from 1.11 MPa to 2.87 MPa) and thermal stabilities of the films. Liu et al. [48] prepared waxy corn starch-based nanocomposite films reinforced with corn starch nanoparticles (0-25%). The addition of corn starch nanoparticles, particularly below 15%, greatly improved the mechanical strength (TS from 1.40 to 2.35 MPa), water vapour barrier (from $5.89 \times 10-12$ to 3.08x10-12 g/m s Pa) and thermal stability as compared to the plain corn starch-based films. González and Igarzabal [49] prepared soy protein isolate edible films dispersed with native corn starch nanocrystals (5–40%) through casting method and found that the nanostarch addition improved the physical and mechanical properties, such as, water solubility, swelling, WVP and TS. The nanostarch incorporated films were found to be more transparent and homogeneous as the amount of nanostarch particles increased. However, the opacity and degree of crystallinity had slightly increased with the higher amounts of starch nanocrystals.

Condés et al. [50] investigated the effects of the origin of maize starch nanocrystals, waxy or normal, on the amarnath protein-based edible films prepared through film casting. The nanoedible films showed better physical properties (lower WVP, lower water uptake and higher surface hydrophobicity) and the mechanical properties as compared to those of the protein control films. The starch nanocrystals incorporated films were homogeneous and translucent than the neat protein film, without affecting the thickness and the optical properties. Jiang et al. [51] produced pea starch-based nanoedible films incorporated with the potato starch nanoparticles. The potato nanoparticles exhibited the spherical-shaped nanoparticles in the SEM characterization with diameter of about 15-30 nm. The relative crystallinity of potato starch nanoparticles reinforced nanoedible films was found to be higher than that of the native starch films. The addition of nanoparticles improved the mechanical properties (increased from 8.8 MPa to 15.0 MPa at 6% concentrations of nanoparticles). Moreover, the nanocomposite films also exhibited good water resistance and thermal stability as compared with native pea starch-based film. Oliveira et al. [52] used mango kernels for preparing both mango starch and starch nanocrystals and produced the starch-based nanoedible films with 0-10 wt% concentrations of starch nanocrystals. The corn starch nanoedible films were also prepared for the comparison purpose by adding 0–10 wt% of nanocrystals isolated from corn starch. The mechanical properties of both the films were compared. The incorporation of up to 7.5% starch nanoparticles to mango starchbased nanoedible films showed higher TS (90%) and YM (120%), while reducing the WVP (15%), as



compared to that of plain films. It was also noted that the mechanical properties were superior for mango starch-based nanoedible films as compared to those of corn starch-based nanoedible films.

Nanocellulose reinforced edible films

Nanocellulose is one of the most discussed emerging biopolymers in the past few decades with exceptional physicochemical properties, such as transparency, light weight, non-toxic, low density (around 1.6 g/ cm³) and high strength to weight ratio (8 times higher than that of stainless steel). It has high stiffness (YM of 220 GPa) which is higher than that of Kevlar fibre and has high tensile strength (TS of 10 GPa), which is higher than that of cast iron [53–57]. Nanocellulose can be extracted from the cellulosic sources, such as agricultural plants and residues, lignocellulosic biomass and wastes, microcrystalline cellulose, animals, bacteria and algae [55, 58, 59]. Isolation of nanocellulose has been reported in the literatures from different cellulose sources, such as, rice husk [60], rice straw [61, 62], banana pseudostem [63, 64], jute stem [65], pineapple leaf [63, 65], wheat straw [66], kenaf bast [67], potato pulp [68], corncob [69], mulberry [70], wood [71], cotton [72, 73], ramie [74], sisal [75, 76], soybean straw [77], sugarcane bagasse [78, 79], tunicates [80], bacteria [81, 82] and fruit & vegetable wastes [83-85]. The common methods reported for isolating such nanocellulose include acid hydrolysis [78, 79, 86], steam explosion [63, 65], ultrasonication [87-89] and high-pressure homogenization [90–92] and enzymatic hydrolysis [93].

Three kinds of nanocellulose structures are possible: cellulose nanocrystals, nanofibrillated cellulose and bacterial cellulose. Cellulose nanocrystals represent the smallest fundamental unit of about 5 nm width. The cellulose nanocrystals are generally rigid and reasonably straight with aspect ratio ranging from 11 to 67, making them to be suitable for thin film applications. While the crytallinity of the cellulose ranges from 25 to 75%, the crystallinity of the cellulose nanocrystals is about 85%. Nanofibrillated cellulose has relatively larger width and aspect ratio as compared to those of the cellulose nanocrystals. The individual fibrils of nanofibrillated cellulose typically range from 20 to30 nm in width. The crystallinity varies from 60 to70% representing the presence of higher amounts of non-crystalline matter. Hence, nanofibrillated cellulose is considered to be more flexible in wet conditions as compared to that of cellulose nanocrystals. Bacterial nanocellulose is synthesized primarily with the help of the bacterium 'Gluconacetobacter xylinus'. Although initial production of bacterial nanocellulose was studied in Gluconacetobacter xylinus, other microorganisms, such as Gluconacetobacter, Agrobacterium tumefaciens, Rhizobium spp. and Gram-positive Sarcina ventriculi, are also capable of synthesizing this nanocellulose. Unlike plant-derived nanocellulose, bacterial nanocellulose is devoid of lignin, hemicellulose, pectin, or any other compound present in the plant-derived nanocellulose. Therefore, bacterial cellulose is considered as the pure form of the nanocellulose without any further treatment and could be used in direct food applications. The width of individual fibrils of bacterial nanocellulose ranges from 20-100 nm with superior mechanical properties as compared to plant-derived nanocellulose [94–96].

The research works specific to nanocellulose reinforced nanoedible films are summarized as follows. In the work of Azeredo et al. [97], nanocomposite films were prepared from chitason reinforced with nanocellulose and the resulting films containing 15% nanocellulose and 18% glycerol had exhibited higher TS and YM as comparable to those of the synthetic polymers. However, their %*E* and WVP were poorer, making it suitable only for the applications that do not require WVP and flexibility. Bilbao-Sainz et al. [98] incorporated three types of nanocellulose particles, namely (a) nanofibrils cellulose, (b) oxidized nanofibrils cellulose using tempo reaction and (c) nanocellulose whiskers, into HPMC edible films. The results showed that incorporation of nanofibrils and oxidized nanofibrils exhibited negative effects on mechanical and water barrier properties. However, nanocellulose whiskers improved the mechanical properties (by 22% higher in TS and 55% higher in YM) and water barrier properties (by 14% lesser in WVP). Furthermore, the film transparency of nanocellulose whiskers was decreased only by 3-6% of the HPMC films, while nanofibrils and oxidized nanofibrils decreased the film transparency by 42% and 35%, respectively. George and Siddaramaiah [82] prepared bacterial cellulose nanocrystals (20 \pm 5 nm and 290 \pm 130 nm) from Gluconacetobacter xylinus using acid hydrolysis, and produced bionanocomposite with gelatin. The produced edible films showed improved mechanical properties, moisture sorption and water vapour permeability.

Shankar and Rhim [99] produced the nanocellulose from microcrystalline cellulose using NaOH/urea dissolution method, followed by regeneration, neutralization and ultrasonication and prepared edible films by incorporating the nanocellulose or microcellulose in agar matrix, and the film properties were evaluated. The results showed that the mechanical and water vapour barrier properties were significantly higher at lower amounts of nanocellulose as compared to those of microcrystalline cellulose addition. Chaichi et al. [100] produced pectin-based edible films reinforced with crystalline nanocelullose (2, 5 and 7 wt%). The results showed that the film having 5% nanocellulose produced the optimum mechanical and water vapour properties with up to 84% increase in TS and 40% decrease in WVP. Chaichi et al. [100] prepared the edible pectin-based films reinforced with celullose nanocrystals (2, 5 and 7% w/w) using solution casting method. The optimum film performance was achieved at 5% incorporation of the cellulose nanocrystals in which about 84% increase in TS and 40% decrease in WVP were observed. It was also observed that the addition of cellulose nanocrystals did not significantly affect the thermal properties, such as glass transition temperature. Viana et al. [101] produced nanoedible films from pectin and fruit puree matrix reinforced with nanofibrillated bacterial cellulose. The bacterial cellulose was produced from cashew apple juice by Komagataeibacter xylinus. Two different fruit purees, namely guava puree and mango puree, were used as the plasticizers. The edible films were prepared by replacing pectin with different amounts of bacterial nanocellulose (0, 25, 50, 75, and 100%). The addition of fruit purees improved the plasticizing effects due to the presence of fruit sugars, and the increase in WVP (13-18 times), decrease in TS (90%), decrease in YM (99%) and increase in % E (13 times) were noticed. The partial or complete replacement of pectin with bacterial nanocellulose improved film properties, making them stronger, stiffer, more resistant to water. It was suggested from the results that the fruit containing edible films based on pectin were suitable for sachets, while the same on bacterial nanocellulose was suitable for food wrapping or coating.

Wang et al. [102] produced the agar-based edible films reinforced with nanobacterial cellulose (0, 3, 5, 8

and 10%), and film properties were investigated. The addition of nanobacterial cellulose improved the crystallinity and the thermal stability of films. The dispersion of nanobacterial cellulose was good at low concentrations (3-5%) as compared to high concentrations (8–10%). The addition of high concentrations of nanobacterial cellulose (10%) significantly reduced the moisture content, water solubility and WVP by about 60.4%, 13.3% and 25.7%, respectively. With respect to mechanical properties, the addition of nanobacterial cellulose increased TS from 22.1 MPa to 44.51 MPa, while %E was initially increased up to 0-5% and then decreased for 8-10% concentrations. Shabanpour et al. [103] produced the fish myofibrillar protein-based edible films reinforced with bacterial nanocellulose of 2, 4 and 6%, w/w concentrations. The results showed that 6% addition of nanocellulose improved TS by 49%, while also improving WVP and solubility index. Ilyas et al. [104] prepared the nanoedible films from sugar palm starch and sugar palm cellulose nanocrystals (0-1.0 wt%) with sorbitol/glycerol as plasticizer using solution casting method. The length, diameter and L/D ratio of the cellulose nanocrystals are 130 ± 30.23 nm, 8.5 ± 1.82 nm and 15.3, respectively. Due to chemical similarities and compatibility of both sugar palm starch and sugar palm cellulose nanocrystals, good dispersion and adhesion were observed in the prepared edible films. The results showed that the addition of nanocellulose improved the crystallinity, the mechanical strength (TS from 4.80 to 11.47 MPa and YM from 54 to 178.83 MPa), thermal stability and water barrier properties compared to the plain sugar palm starch film.

Nanochitosan/nanochitin reinforced edible films

Chitin (and its derivative chitosan) is a naturally occurring polysaccharide, which is the second most abundant semi-crystalline biopolymer next to cellulose. It is mainly found in the exoskeletons of crabs, shellfish, shrimp, tortoise, insects and cell walls of yeast, fungus and mushrooms [105, 106]. The chitin fibres are made up of thin filaments, called microfibrils embedded in protein matrix. The diameters of the microfibrils range from 2.5 to 2.8 nm, which are tightly bonded to each other with strong hydrogen bonds and are arranged in the order of alternating crystalline and amorphous domains. The isolation treatments cut these microfrbrils in a longitudinal direction breaking the amorphous domains. This dissolution of amorphous domains can therefore separate the chitin crystallites, also called chitin nanowhiskers) [107, 108]. Chitin nanowhiskers can be extracted through a series of extraction methods and chemical treatments, such as acid hydrolysis [109–111], partial deacetylation [112, 113], ultrasonication [114, 115], mechanical treatment [116] and gelation treatment [117].

Fan et al. [118] prepared and compared the properties of the cast films produced from four types of chitins produced through different preparation methods (HCl-hydrolyzed chitin nanowhiskers, TEMPO-oxidized chitin nanowhiskers, partially deacetylated chitin nanowhiskers and squid-pen chitin nanofibre). TEMPO-oxidized and HCl-hydrolyzed chitin nanowhiskers were rod-like structures with few aggregates, while the partially deacetylated and squid-pen (mechanical treatment) chitin nanowhiskers were dispersions of individual nano-elements of approximately 4 nm width. All chitin nanowhisker had shown almost the similar thermal degradation point and O2 permeability, which were approximately 200 $\,$ C and 1 mL mm⁻² day^{-1} kPa⁻¹, respectively. While the partially deacetylated chitin nanowhiskers films exhibited the highest TS of 140 MPa, %E of 10% and light transmittance of about 87% at 400 nm, the squid-pen chitin nanofibre films had exhibited the lowest TS of 35 MPa, %*E* of 3.2% and light transmittance of about 75% at 400 nm. De Moura et al. [119] prepared the chitosan-tripolyphosphate nanoparticles (of different particle sizes of 21 nm, 110 nm and 221 nm, respectively) using ionic gelation of chitosan with tripolyphosphate anions and produced four hydroxypropyl methylcellulose (HPMC) edible films by incorporating with or without nanoparticles. The effect of sizes of nanoparticles were studied, and the results showed improved mechanical properties, WVP and thermal degradation in the order (best to worst) of 21 nm sized nanoparticles followed by 110 nm and 221 nm and HPMC film. Ifuku et al. [119–121] produced the transparent nanoedible films from chitosan reinforced with surface-deacetylated chitin nanofibres. With 10% addition of chitin nanofibres, the mechanical strength of the nanoedible films increased such that TS and YM by 65% and 94%, respectively, while reducing the thermal expansion coefficient from 35.3 ppm/K to 26.1 ppm/ K. These films also showed good antifungal activity against Alternaria alternata.

Antoniou et al. [122] produced tara gum films by incorporating the bulk chitosan or chitosan nanoparticles at varying concentrations (0, 5, 10, 15% w/w). With the addition of chitosan nanoparticles, mechanical and barrier properties were improved, i.e. TS increased by 35.73 MPa, elongation decreased by 7.21%, film solubility decreased by 74.3% and WVP decreased by 22.7%). The properties of tara gum films with chitosan nanoparticles were superior than that of with bulk chitosan. Hosseini et al. [123] prepared fish gelatin-based nanoedible films reinforced with chitosan nanoparticles (size ranging between 40 and 80 nm). The incorporation of chitosan nanoparticles (0%, 2%, 4%, 6% and 8%) had resulted in the improvement on the gelatin edible films. Mechanical strength (TS and YM) had increased and WVP decreased due to the addition of nanoparticles. Qin et al. [124]. prepared maize starch-based edible films dispersed with chitin nanowhiskers at different concentrations (0%, 0.5%, 1%, 2% and 5%). The addition of nanowhiskers had improved the mechanical and barrier properties of maize starch films with significant increase in film opacity. Vahedikia et al. [125] produced zein-based nanofilms loaded with chitosan nanoparticles (4%) and/or cinnamon essential oil (2%). The produced films, particularly, the one having both chitosan nanoparticles and cinnaon essential oil, decreased the WVP by 41% and increased TS by 112%, while decreasing % E by about 45%. It was also observed that the composite nanofilms could be used as an antibacterial agent against Gram-positive (Staphylococcus aureus) and Gram-negative bacteria (Escherichia coli).

Nanoproteins reinforced edible films

Protein is a linear heterogeneous biopolymer made up of different kinds of amino acids to form the unique three-dimensional network structures. The variations in the amino acid sequences determine the interactions of the functional groups leading to the possibility of more than thousand different protein structures [126]. Protein nanoparticles are typically biodegradable, non-antigenic, metabolizable and easily amenable for surface modification. The processing and control of desired size, morphology and weight of the protein nanoparticles are easy [127, 128]. The protein nanoparticles are generally obtained through (a) desolvation, (b) emulsification, (c) thermal gelation and (d) nanospray drying [129]. The isolation of protein nanoparticles were reported in the literature from the protein sources, such as, corn zein [130–132], gelatin [133, 134], whey protein [135, 136], egg albumin [137, 138], soy protein [139] and casein [140, 141].

Li et al. [142] produced the peanut protein nanoparticles through the anti-solvent method and produced the nanoparticles incorporated (0-4%) sov protein isolate and corn starch edible films. The experimental results showed that addition of protein nanoparticles improved the TS, WVP and thermal stability of both soy protein and corn starch-based edible films. While the corn starch films performed the best with 4% nanoparticles, the soy protein films performed the best with 2% nanoparticles, due to high compatibility and cohesion between protein matrix and protein nanoparticles reinforcement. Li et al. [143] prepared the antimicrobial sodium caseinate-based edible films with zein-sodium caseinate nanoparticles and thymol oil (thymol ro zein ration from 0 to 40%). The thymol oil loaded zein-sodium caseinate nanoparticles were first produced by antisolvent method, and the solution casting method was used to produce the edible films. Thymol oil improved the DPPH radical scavenging activity and the antimicrobial activity against Escherichia coli and Salmonella. The mechanical strength was improved for the small addition of nanoparticles up to 10%, and the films became weaker above 10% addition of nanoparticles. Moreover, there was no significant difference observed on WVP among the films. Oymaci & Altinkaya [144] produced whey protein isolate-based edible self standing films with corn zein nanoparticles through solution casting. Zein is a safe material for consumption with high hydrophobicity, which makes it a good choice for improving WVP. However, to achieve homogeneous distribution, sodium caseinate was coated over the films. The results showed that the incorporation of zein nanoparticles improved the mechanical (303% higher TS) and barrier properties (84% lower).

Zhang and Zhao [145] prepared the edible active films corn starch with zein–rutin composite nanoparticles incorporated at different concentrations (0, 0.5, 1, 2, 4 and 10%, w/w). The incorporation of nanoparticles improved the antioxidant property

of the corn starch films and the mechanical and physical properties. TS of the prepared edible films was increased from 1.19 MPa to 2.42 MPa, and %E was increased from 42.1 to 78.84% with the increase in the concentrations of nanoparticles, while decreasing the WVP. Gul et al. [146] investigated the effects of ultrasonic treatment on the film properties of the hazelnut meal protein nanoemulsion-based films enriched with clove essential oil. An ultrasound homogenizer was used in for the ultrasonic treatment in which hazelnut meal protein (4% (w/v)) and clove essential oil (CEO) (3% (v/v)) were homogenized at different time durations (2, 4 and 6 min) and different amplitudes (50, 75 and 100%) to produce the nanoemulsions. The nanoemulsion was then poured into an acrylic plate and dried to produce the nanoedible films. The average particle size was decreased with increasing amplitudes. The prepared films became more transparent and decreased the WVP. The clove oil improved the antimicrobial activity of the edible films against L. monocytogenes, B. subtilis, S. aureus, P. aeruginosa and E. coli. Aboul-Anean [147] isolated the nanoparticles separately from both quinoa protein and quinoa starch, and also the phenolic compounds extract from luria leaves and pomegranate peels. The quinoa protein nanoedible films as well as the quinoa starch nanoedible films were prepared using solution casting. The results showed that the addition of protein and starch nanoparticles on the edible films increased mechanical properties and improved the barrier properties. The films were also able to inhibit the microbial growth with the addition of phenolic compounds extract.

Nanolipids reinforced edible films

Lipids derived from animal and vegetable fats, such as waxes, acylglycerols and fatty acids, have been used for making the edible films and coatings. Lipid films exhibit excellent water barrier properties [148]. Nanolipids are very interesting for self-assembled nanofilms and other nano-structures. Lipid nanoparticles consist of a lipid core surrounded by one or more surfactant materials. The earlier nanolipids are nanoemulsions, liposomes, polymeric nanoparticles. Solid lipid nanoparticles, followed by nanostructured lipid carriers, were later developed in order to overcome the problems associated with these nanolipid carriers, such as emulsions, polymeric nanoparticles and liposomes. Solid lipid nanoparticles consist of 0.1–30% solid fat dispersed in an aqueous medium. Nanostructured lipid carriers are modified solid lipid nanoparticles where a solid fat and liquid lipid are in liquid phase at ambient temperature [149–152]. Some of the commonly used techniques used for the preparation of nanoemulsions include high-pressure valve homogenization, ultrasonic homogenization, high-pressure microfluidic homogenization, colloid mills, spontaneous emulsification, phase inversion temperature and phase inversion composition [153, 154].

Acevedo-Fani et al. [155] produced sodium alginate edible films with nanoemulsions made from thyme oil (Thymus vulgaris), lemongrass oil (Cymbopogon citratus) or sage oil (Salvia officinalis). The results indicated that the incorporation of nanoemulsions (1% v/v) improved the colour difference, mechanical and barrier properties of the edible films. Among three essential oils, sage oil produced the significant improvement in terms of mechanical and barrier properties without much penalty in colour difference as compared to other two oils. Wu et al. [156] produced fish gelatin-based nanoedible films incorporated with cinnamon essential oil nanoliposomes (5% v/v), and the film properties were compared with the plain gelatin films. The nanoliposomes-reinforced edible films showed TS of 6.5 MPa and %*E* of 85.71%, while plain gelatin films showed TS of 8.97 MPa and %E of 65.41%. Thus, the addition of nanoliposomes decreased the mechanical strength, but improved the elongation of the films. WVP of nanoliposomes-reinforced edible films is found to be lower than that of the plain films. The opacity of nanoliposomes-reinforced edible films was increased from 2.19 to 2.34, making the film more opaque. Cui et al. [157] produced agar-based edible film incorporated with Artemisia annua oil nanoliposomes and chitosan for improving the antibacterial effect on Escherichia coli. The mean size of liposomes was around 191.8 nm. The antibacterial effect was evaluated on cherry tomatoes, and the results showed that the addition of chitason and nanoliposomes brought the antibacterial effect on the agarbased edible films.

Hashemi-Gahruie et al. [158] produced the basil seed gum-based edible films reinforced with Zataria multiflora essential oil nanoemulsions and investigated the effect of the nanoemulsion droplet size on

the antibacterial activity. It was found that the decrease in the nanoemulsion particle size increased the antibacterial activity against Gram-positive and Gram-negative bacteria. In addition, the mechanical strength of the films was improved due to the addition of nanoemulsion. Robledo et al. [159] produced thymol nanoemulsions through spontaneous emulsification, ultrasound, and a combination of both these methods, and found that the best result was found in spontaneous emulsification method in terms of size and dispersion. The nanoedible films were then produced by incorporating thymol nanoemulsion in water in quinoa-chitosan films. The resulting edible film exhibited porous microstructure. There was no significant variation in WVP. The mechanical strength differed from the control film, where TS reduced and %E increased, with the addition of nanoemulsions. The nanoemulsion incorporated films significantly decreased the mould growth of Botrytis cinerea on cherry tomatoes. Frank et al. [160] produced alginate-based nanoedible films by incorporating cinnamon essential oil nanoemulsions, and the film properties were evaluated. While YM and YS increased with an increasing amount of nanoemulsions, %E decreased. The maximum TS and %E of the films were found to be 15.63 MPa and 23.67%, respectively, for the film with 20% nanoemulsion. The film containing 20% nanoemulsion also increased the antibacterial effects against Escherichia coli, Bacillus cereus, Salmonella typhimurium and Staphylococcus aureus. Restrepo et al. [161] produced green banana (Musa paradisiaca L.) starch-based nanoedible films reinforced with nanoemulsions of two essential oils. The nanoemulsions were prepared separately from lemongrass (Cymbopogon citratus) oil and rosemary (Rosmarinus officinalis) oil using the emulsion-phase inversion method. The nanoediprepared ble films were by incorporating nanoemulsions of three different proportions. The results showed that incorporation of the lemongrass nanoemulsion or rosemary nanoemulsion had increased the plasticizing effect of the banana starch edible films. The plasticizing effect had resulted in higher transparency and increased %E, TS and YM, but this plasticizing effect led to higher WVP. However, the increase in WVP was not significant as the hydrophobic nature of the lemongrass nanoemulsion or rosemary nanoemulsion counteracted this plasticizing effect.

Film properties of some nanoedible films

Mechanical properties

The mechanical strength of a material at the nanoscale is usually different from that of at its macroscopic scale. Nanofiller chemistry, filler size and the shape are the major contributors of the overall mechanical properties of the nanocomposites. The nanofiller chemistry affects the polymer-nanofiller interactions and inter-filler interactions. The polymer-nanofiller interactions contribute more for the stress transfer at the nanofiller-polymer interfaces, while the inter-filler interactions contribute to agglomerates at high volume fractions of the filler. The nanofiller chemistry thus greatly affects the mechanical properties, and it is essential to have the homogeneous dispersion of the fillers in the matrix, breaking of the agglomerates and good wettability of the fillers with the polymer for improving the stress transfer of the composite. The filler size is dictated by surface-to-volume ratio and excluded volume interactions. Surface-to-volume ratio is an indicator of the amount of interfacial region compared to the bulk material. For instance, the surface-to-volume ratio of the spherical filler material particles of radius, r, would be $\left(\frac{3}{r}\right)$. From this relationship, one can say that the available surface area per volume of the filler material increases with the decrease in radius. The total surface area or interfacial area within a composite is given by $\left(\frac{3\emptyset}{r}\right)$, where φ is the volume fraction of the filler material in the composite. From this relationship, one can say that, for the constant volume fraction of filler material, if the filler radius is reduced, the surface area available for interfacial interactions increases. With higher surface-to-volume ratio of the nanomaterials as filler material, quantity of chain-filler interactions is more facilitating the increased stress transfer between the filler material and polymer. As a result, the mechanical strength is increased. Filler shape also contributes a significant role to surface-to-volume ratios. For instance, the surface-to-volume ratio of the a cylindrical filler particle of radius (r) and length (L) is given by $\left(\frac{2}{r}+\frac{2}{T}\right)$. Now, if we determine the surface-to-volume ratio of sphere relative to the cylinder, the value would be $\left(\frac{3}{2\left(1+\frac{t}{L}\right)}\right)$. From this relationship, it is understood that if (r > L) and (L < 2r), the surface-to-volume ratio of

the cylindrical particles is higher than that of the spherical particles. When L > 2r, the spherical particles have greater surface-to-volume ratios as compared to the cylindrical particles. However, the maximum increase of the surface-to-volume ratios of spherical particles is only 50%. If surface-to-volume ratio is considered as the primary design factor, the plate-like structures have significant effect on improving the stress transfer and mechanical strength. Unfortunately, shape factor should also act as a critical design factor for mechanical strength. For instance, dispersion of cylindrical fillers isotropically is difficult as the aspect ratio increases. In such cases, the non-isotropically dispersed filler particles produce anisotropic nature in the mechanical strength of the resulting composite [162-165]. For microscale structures, the elastic strain energy controls the mechanical properties. But, for nanoscale structures, because of higher surface-to-volume ratios, the surface effects are predominant and therefore the surface effects control the mechanical properties. The elastic modulus of the nanomaterials was found to be significantly higher than that of the bulk material [166].

ASTM Standard D882 (Standard Test Method for Tensile Properties of Thin Plastic Sheeting) is used to determine the tensile properties of the edible films. The edible films are cut and placed in the metal grips of a universal testing machine. Ultimate tensile strength, expressed in force per unit area (MPa), is calculated by dividing the maximum load by the original cross-sectional area of test films. Per cent elongation at break, expressed in percentage (%), is calculated by dividing the elongation of the film by the initial gage length and multiplying by 100. Modulus of elasticity, expressed in force per unit area (MPa), is a measure of stiffness and is calculated by dividing applied stress by strain corresponding to the applied stress (in stress-strain curve) [167-169]. Table 1 shows the mechanical properties of the nanoedible films produced from different sources of matrix and reinforcement. The mechanical strength of the edible films was improved with the addition of nanomaterials. While TS and YM of nanoedible films increased, %E decreased with the addition of nanomaterials. Therefore, the addition of nanomaterials during the preparation of edible films has the potential to improve the mechanical properties with the penalty of reduced %E. In some cases, the mechanical strength was comparable with the synthetic plastics. However, %E of nanoedible films was



Table 1 Mechanical properties

S. no.	Matrix material	Fillers/additives	Film thickness μm	Tensile properties			Refs.
				TS MPa	YM MPa	%E &	
1	Gelatin	Bacterial cellulose (0 wt%)	na	83.7	2189.5	33.7	[82]
		Bacterial cellulose (1 wt%)		88.7	2225.3	33.1	
		Bacterial cellulose (2 wt%)		95.1	2272.8	29.8	
		Bacterial cellulose (3 wt%)		103.1	2335.1	27.5	
		Bacterial cellulose (4 wt%)		108.6	2350.4	23.4	
		Bacterial cellulose (5 wt%)		89.8	2321.9	20.8	
2	Hydroxypropyl	Chitosan-tripolyphosphate (0%)	na	na	900	8.1	[121]
	methylcellulose (3 g/	Chitosan-tripolyphosphate (21 nm)			1264	11.1	
	100 ml nanoparticle	Chitosan-tripolyphosphate (110 nm)			1190	5.2	
	solution)	Chitosan-tripolyphosphate (221 nm)			1204	5.7	
3	Maize starch	Chitin nanowhiskers (0%)	148	1.64	na	175	[126]
		Chitin nanowhiskers (0.5%)	144	2.79		176	L .]
		Chitin nanowhiskers (1%)	147	3.69		179	
		Chitin nanowhiskers (2%)	146	3.17		160	
		Chitin nanowhiskers (5%)	145	2.37		111	
4	Pea starch (5 g/100 ml	Waxy maize starch nanocrystals (0%)	104	5.76	21.15	29.23	[46]
	H_2O) + glycerol (1.5 g/	Waxy maize starch nanocrystals (1%)	109.4	6.56	27.95	26.18	
	100 ml H ₂ O)	Waxy maize starch nanocrystals (3%)	106.8	6.95	37.89	20.46	
		Waxy maize starch nanocrystals (5%)	117.8	9.96	85.72	12.58	
		Waxy maize starch nanocrystals (7%)	121	7.12	36.59	21.6	
		Waxy maize starch nanocrystals (9%)	131.2	6.68	27.56	26.7	
5	Waxy maize starch (5.5 g/ 35 g H ₂ O) + glycerol (20% of wt% starch)	Waxy maize starch nanocrystals (0%)	na	2.4	49	182	[43]
-		Waxy maize starch nanocrystals (5%)		13.2	298	8.2	[]
		Waxy maize starch nanocrystals (10%)		13.6	333	8.6	
		Waxy maize starch nanocrystals (15%)		7.6	_	4.5	
	Waxy maize starch (5.5 g)	Waxy maize starch nanocrystals (0%)		1.02	11	297	
	$35 \text{ g H}_2(0) + \text{glycerol}$	Waxy maize starch nanocrystals (5%)		3.6	80	97	
	(25% of wt% starch)	Waxy maize starch nanocrystals (10%)		4 2	82	57	
		Waxy maize starch nanocrystals (15%)		9.8	241	20	
	Waxy maize starch (5.5 g)	Waxy maize starch nanocrystals (0%)		0.26	0.46	551	
	$35 \text{ g H}_2\text{O}$ + glycerol (30% of wt% starch)	Waxy maize starch nanocrystals (5%)		1.3	3.4	236	
		Waxy maize starch nanocrystals (10%)		2.7	25	89	
		Waxy maize starch nanocrystals (15%)		3.6	44	82	
6	Soy protein isolate (0.25	Corn starch nanocrystals (0%)	na	1.1	26.89	65.95	[49]
-	g/30 ml	Corn starch nanocrystals (2%)		1.42	55.31	53.79	[]
	H_2O + glycerol (50	Corn starch nanocrystals (5%)		1.34	39.42	58.67	
	wt% protein)	Corn starch nanocrystals (10%)		1.79	71.05	32.17	
	(it) (protoni)	Corn starch nanocrystals (20%)		2.61	102.23	41.89	
		Corn starch nanocrystals (40%)		5.08	310.34	21.35	
7	Agar (3 g/150 ml	Nanocellulose (0%)	42.3	46.7	1340	15.7	[101]
	$H_2O) + glycerrol$	Nanocellulose (1%)	44.3	48.7	1370	15.4	[]
	$(0.9 \text{ g}/150 \text{ m} \text{H}_2\text{O})$	Nanocellulose (3%)	44.4	52.8	1390	15.8	
	(Nanocellulose (5%)	46.3	45.3	1230	17.6	
		Nanocellulose (10%)	50.7	38.4	1150	17.8	
8	Gelatin (4 g/100 ml	Nanochitin (1%)	0.1	65.19	1691.81	4.63	[170]
-	$H_2O) + glycerol (30)$	Nanochitin (3%)	0.1	75.02	1992.18	5.58	[1/V]
	wt% of gelatin)	Nanochitin (5%)	0.11	119.08	3247.61	5.82	
	U ,	Nanochitin (10%)	0.14	31.59	902.18	6.54	

S. no.	Matrix material	Fillers/additives	Film thickness	Tensile properties			Refs.
			μm	TS MPa	YM MPa	%E &	
9	Carrageenan (3 g/100 ml	Chitin nanofibrils (0%)	0.535	30.2	1070	21.8	[171]
	H_2O) + glycerol (1.2 g/	Chitin nanofibrils (3%)	0.592	40.7	1430	14.4	
	100 ml soln)	Chitin nanofibrils (5%)	0.622	44.7	1560	11.2	
		Chitin nanofibrils (10%)	0.668	29.8	1770	3.9	
10	Fish gelatin (4 g/100 ml	Chitason nanoparticle (0%)	50.58	7.44	287.03	102.04	[125]
	H_2O) + glycerol (0.3 g/	Chitason nanoparticle (2%)	54.52	7.99	371.93	70.09	
	1 g gelatin)	Chitason nanoparticle (4%)	61.16	8.77	392.25	64.72	
		Chitason nanoparticle (6%)	62.14	10.57	453.46	44.71	
		Chitason nanoparticle (8%)	64.87	11.28	467.2	32.73	

Table 1 continued

na Data not available

found to be much lower than that of synthetic plastic films. Further investigation is required in the improvement of mechanical strength, while not compromising on %E.

Water vapour permeability

The effect of nanofillers on barrier properties can be explained with a well-known theory developed by Nielsen [172]. According to him, the permeability of a polymer is decreased with the addition of a filler material. If the filler material is impenetrable to a gas or liquid that is diffusing into the polymer, then the diffusing gas or liquid must go around these filler material particles. This phenomenon is explained in Fig. 2, in which the diffusing gas or liquid is forced to take a tortuous path leading to an increase in path length and the permeability is decreased drastically. According to the theory, the addition of fillers should substantially increase the travel path of the diffusing gas and vapours so as to improve the barrier properties of the composite, if the filler particles are thin plates having a large length to width aspect ratio, homogeneously dispersed and oriented so that the filler surfaces are parallel to the polymer surface [172, 173]. Variations of Neilsen model were proposed by different researchers like Nape et al. [174], Aris [175], Cussler et al. [176], Fredrickson and Bicerano [177], Gusev and Lusti [178], Bharadwaj [179]. Modified-Neilsen mechanism is often used to explain the barrier properties in nanocomposites. However, from the data collected from more than 1000 articles by Wolf et al. [180], it was observed, however, that the improvement of barrier properties in the nanocomposites was not as high as what predicted from the theory. According to them, various interfering mechanisms, which influence tortuosity, sorption, polymer matrix crystallinity, polymer matrix free volume, molar mass of polymer matrix, interphase, voids and cracks, etc. could cause unexpected changes in the barrier properties of the nanocomposites.

The permeability is explained with three parameters: diffusion coefficient, solubility coefficient and permeability coefficient. Diffusion coefficient represents the movement of permeant molecules through a polymer film. Solubility coefficient represents dissolution of a permeant in a polymer film. Permeability coefficient combines both diffusion coefficient and solubility coefficient to provide a gross mass transport property. Diffusion coefficient can be described with the help of Fick's first law. It states that the mass flow rate of a permeant, *J*, is directly proportional to the concentration gradient $(\partial C/\partial X)$ and is expressed as I = -D. ($\partial C/\partial X$), where, C is the concentration of permeant, X is the thickness of the film, and D is the diffusion coefficient. Solubility coefficient is described by Nernst distribution function and is expressed as $C = S \cdot p$, where, p is the vapour pressure of the permeant, and S is the solubility coefficient. The permeability coefficient is related diffusion coefficient and solubility coefficient by the expression, $P = D \cdot S$. With two assumptions: (a) steady state diffusion and (b) constant diffusivity, mass flow rate is given by,

 $J = \frac{D(C_2 - C_1)}{X} = \frac{Q}{(A * t)}$, where, Q is the amount of







Pathway through Larger Filler Particles

Pathway through Smaller Filler Particles

permeant atoms diffused through the film (g), A is the area of the film (mm^2) and t is the diffusion time (s). Hentry's law allows to consider partial pressure differential of the permeant, and the expression is rearranged as, $\frac{Q}{(A*t)} = \frac{D*S*(p_2-p_1)}{X} = \frac{P*\Delta p}{X}$, where, S is Hentry's law solubility coefficient (mole/atom), Δp is the partial pressure difference across the film (Pa), and P is the permeability $(g mm/(m^2 s Pa))$. The permeability is thus expressed as the following expression, $P = \frac{Q*X}{(A*t*\Delta p)}$ [168, 181, 182]. ASTM E96 is generally used as the standard test procedure, also known as gravimetric or cup method, for determining water vapour transmission rate. This method uses a temperature and humidity controlled chamber that contains a cup or a dish filled with desiccant/water. The prepared films are sealed on cups, and weight changes are periodically monitored [168, 183]. Table 2 shows the WVTR of the nanoedible films produced from different sources of matrix and reinforcement. The WVTR of the edible films was increased when the nanoparticles were added to the nanoedible films. Due to the nanoscale dimensions of the nanoparticles, they could cover up the matrix material that prevents the diffusion of water molecules. This, in turn, decreased the WVP of the edible films making the nanoedible films the potential candidates with improved WVTR. However, WVTR values of the nanoedible films were much lower than those of the synthetic plastic films. Further investigation is required to explore the techniques to improve the WVTR to compete with the synthetic plastics.

Film colour

Another interesting property of nanoparticles is their optical properties. The optical properties of the nanoparticles strongly depend upon the particle size and shape. For example, gold in microscale appears

as yellow colour in reflected light, while the same gold in nanoscale appears blue in colour. When the particle size of gold is further reduced to about 3 nm, the blue colour is changed to orange. Hence, understanding the optical behaviour of nanoparticles is essential before their use in packaging applications [184–186]. Microscale particles used in a polymer matrix scatter light, which in turn reduces the light transmittance and optical clarity. But, when efficient nanoscale particles are used with good polymernanofiller interfacial adhesion, the composite eliminates the scattering effect which in turn develops transparent films and coatings [187-190]. The composites generally scatter more light due to differences in refractive indices resulting in a loss of transparency. However, in the work of Yano et al. [191], a high degree of transparent sheets was produced with transparent thermoset polymers (epoxy, acrylic and phenol formaldehyde) and 60-70 wt% cellulose nanofibres. Despite having high fibre content and variations in refractive indices, the composite sheets could maintain a high degree of transparency. The results indicate that the nanosize allowed combining the optically functional materials of different refractive indices without affecting the transparency. The optical properties depend on the size, shape and scattering behaviour of the nanofillers and the scattering behaviour of the polymer. The particle size of below 40 nm is essential to produce the transparent nanocomposites. This is due to Rayleighs law in which the intensity of scattered light increases with increasing particle size. The intensity ratio is given by: $\left(\frac{I}{I_0}\right) = e^{-\left[\frac{3\varphi_p xr^3}{4\delta^4}\left(\frac{n_p}{n_m}-1\right)\right]}$, where *I* is the intensity the transmitted light and I_0 is the intensity of the incident light, *r* is the radius of the spherical particles, n_p is the refractive index of the nanoparticles, $n_{\rm m}$ is the

refractive index nm of the matrix, δ is the wavelength

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Table 2 Water vapour permeability

No.	Matrix material	Fillers/additives	Film thickness	WVTR	Refs.	
			μm	g-mm/m ² day		
1	Undrowneronul	Chitagan tripolyphognhata (0%)	20	10.06	121	
1	methylcellulose (3 g/	Chitosan-tripolyphosphate (076)	IIa	7 95	121	
	100 ml nanoparticle	Chitosan-tripolyphosphate (21 mil)		10.83		
	solution)	Chitosan tripolyphosphate (221 nm)		14.00		
r	Solution)	Nanoomulaion (0%)	50	0.43	157	
2	Socium arginate	Thuma ail nancomulsion	11(070) 50		157	
		Lamongross oil noncomulsion	40	0.37		
		Sage ail non-complaint	42	0.33		
2	Maiza starsh	Chitin nonexpiritors (0%)	30 149	0.20	126	
3	Marze starch	Chitin nanowhiskers (0.59/)	146	2.64	120	
		Chitin nanowniskers (0.5%)	144	2.5		
		Chitin nanowniskers (1%)	14/	1.89		
		Chitic history (2%)	140	1.17		
	T	Chitin nanowniskers (5%)	145	1.33	104	
4	lara gum	Nanochitin (0%)	/3	11.09	124	
		Nanochitin (5%)	/9	9.84		
		Nanochitin (10%)	88	8.57		
_		Nanochitin (15%)	94	8.76		
5	Whey protein isolate	Zein nanoparticles (0%)	71	7.73	146	
		Zein nanoparticles (0.2%)	98	4.23		
		Zein nanoparticles (0.4%)	107	2.96		
		Zein nanoparticles (0.8%)	131	2.07		
		Zein Nanoparticles (1.2%)	151	1.25		
6	Pea starch (5 g/100 ml H ₂ O) + Glycerol (1.5 g/ 100 ml H ₂ O)	Waxy maize starch nanocrystals (0%)	104	268.32	46	
		Waxy maize starch nanocrystals (1%)	109.4	181.68		
		Waxy maize starch nanocrystals (3%)	106.8	146.16		
		Waxy maize starch nanocrystals (5%)	117.8	102.24		
		Waxy maize starch nanocrystals (7%)	121	129.84		
		Waxy maize starch nanocrystals (9%)	131.2	132		
7	Corn starch (7.5 g/100 ml H ₂ O) + glycerol (3 g/ 100 ml H ₂ O)	Taro starch nanoparticles (0%)	148	65.76	47	
		Taro starch nanoparticles (0.5%)	153.5	49.2		
		Taro starch nanoparticles (2%)	161	43.92		
		Taro starch nanoparticles (5%)	148	35.76		
		Taro starch nanoparticles (10%)	157.5	28.8		
		Taro starch nanoparticles (15%)	162.5	32.88		
8	Soy protein isolate (5 g/	Soy protein isolate nanoparticles (0%)	na	104.88	144	
	80 ml H ₂ O) + glycerol (1.25 g/80 ml H ₂ O)	Soy protein isolate nanoparticles (0.5%)		91.92		
		Soy protein isolate nanoparticles (1%)		86.4		
		Soy protein isolate nanoparticles (2%)		79.2		
		Soy protein isolate nanoparticles (4%)		71.52		
	Corn starch (5 g/80 ml	Soy protein isolate nanoparticles (0%)		100.56		
	H_2O) + glycerol (1.2	Soy protein isolate nanoparticles (0.5%)		80.16		
	5 g/80 ml H ₂ O)	Soy protein isolate nanoparticles (1%)		72.72		
		Soy protein isolate nanoparticles (2%)		75.36		
		Soy protein isolate nanoparticles (4%)		79.92		
9	Agar (3 g/150 ml	Nanocellulose (0%)	42.3	31.68	101	
	$H_2O)$ + glycerol (0.9 g/	Nanocellulose (1%)	44.3	28.32		
	150 ml H ₂ O)	Nanocellulose (3%)	44.4	23.28		
		Nanocellulose (5%)	46.3	29.28		
		Nanocellulose (10%)	50.7	31.68		



Table 2	continued
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No.	Matrix material	Fillers/additives	Film thickness μm	WVTR g-mm/m ² day	Refs.
10	Fish gelatin (4 g/100 ml	Chitason nanoparticle (0%)	50.58	34.104	125
	H_2O) + glycerol (0.3 g/	Chitason nanoparticle (2%)	54.52	24.144	
	1 g gelatin)	Chitason nanoparticle (4%)	61.16	19.968	
		Chitason nanoparticle (6%)	62.14	17.208	
		Chitason nanoparticle (8%)	64.87	21.216	

na Data not available

of the light, φ_p the volume fraction of the particles and *x* the optical path length [192].

ASTM D2244 is generally used for determining the colour of the films. Hunter lab scale and CIE lab scale are the widely used colour measurement methods for determining film colour. In hunter lab scale, the colorimeter uses a standard white plate with standard values, L^* , a^* and b^* as 93.49, -0.25 and -0.09, respectively. The colour parameters range represents the following: L = 0 (black) to L = 100 (white), a (greenness) to +a (redness) and b (blueness) to +b (yellowness). D65 illuminant and 2° observer angle are considered for making measurements. The film specimens are placed on the colorimeter to measure three colour parameters of the films, L, a and b [193–195]. In CIE Lab scale (recommended by The International Commission on Illumination), D65 illuminant and 10° observer angle are considered with a white reference plate of *L*, *a* and *b* as 93.52, - 0.81 and 1.58, respectively [196, 197]. Digital colorimetry is another method used for evaluating the film colour. Though this method is not as accurate as the above-mentioned methods, it is considered to be a low-cost method for evaluating the film colour. In this method, high-resolution picture of a film sample is captured using a digital camera. The picture is then imported in an image software (Adobe Photoshop), and the *L*, *a* and *b* colour parameters are evaluated. L values range from 0 (black) to 100 (white); a values range from - 80 (greenness) to 100 (redness); b values range from -80 (blueness) to 70 (yellowness) [198–200]. Table 3 shows the film colour parameters of the nanoedible films produced from different sources of matrix and reinforcement. It was found that the addition of nanomaterials improved the film colour. In most cases, parameter 'L' was increased to '>90', indicating that the whiteness of the edible films was increased with the addition of the nanomaterials. Parameter 'a' was slightly increased in negative direction, but the change of colour parameter was not significant. The values of 'a' ranged between -2.98and 1.13, which represents the neutrality in terms of greenness or redness. Parameter 'b' also showed the increasing trend in most cases indicating an increase in yellowness, but there were also ups and downs in some cases. On the whole, the whiteness of the nanoedible films was increased with slight increase in yellowness. However, due to limited research, further research is required to investigate more on improving film colour or effects of colour additives, printing inks, etc.

Challenges and opportunities

Inadequate film properties

The addition of nanomaterials has improved the overall performance of edible films as compared to the plain edible films (without nanomaterials) by enhancing their mechanical, thermal and barrier properties. Therefore, the use of nanomaterials is a promising way of improving the performance of food packaging films. Several nanoedible films can not only merely carry the food items passively, but also can impart active and/or smart properties to the packaging films, such as antimicrobial properties, multifunctionality, oxygen scavenging ability, enzyme immobilization and biosensing to indicate some level of food degradation [18]. Some of the most important factors deciding the choice of edible films for packaging applications include film flexibility, transparency and superior barrier properties. Even though exhibiting sufficient barrier properties to oxygen and light, most of the edible films are poor water barriers. The edible films also lack of other Table 3 Film colour

S. no.	Matrix material	Fillers/additives	Film colour				Refs.
			L	A _	b 	ΔE	
1	Sadium alginata	Noncomputation $(00/)$	90.61	0.47	4.4	24	157
1	Sodium alginate	Thuma ail nancomulsion	89.01 89.54	- 0.47	4.4	24 59	137
			02.10	- 0.85	0.9	12	
		Sage ail nanoemulsion	95.19	- 0.47	2.5	15	
2	Maiza starsh	Sage oil nanoemulsion	92.72	- 0.42	2.0	11.9	126
2	Maize starch	Chitin nanowhiskers (0.76)	00.0 96.02	- 1.39	1.09	lla	120
		Chitin nanowinskers (0.5%)	80.92	- 1.40	1.08		
		Chitian hanowniskers (1%)	80.02	- 1.55	1.14		
		Chitin nanowniskers (2%)	86.//	- 1.53	1.1/		
2	T	Chitin nanowhiskers (5%)	86.2	- 1.6	1.2	7.05	10.4
3	Tara gum	Nanochitin (0%)	92.58	- 1.29	2.61	7.25	124
		Nanochitin (5%)	91.45	- 1.95	6.8	10.44	
		Nanochitin (10%)	90.65	- 2.28	9.96	13.19	
	D	Nanochitin (15%)	89.8	- 2.48	12.75	16.79	
4	Pea starch (5 g/100 ml H ₂ O) + glycerol (1.5 g/	Waxy maize starch nanocrystals (0%)	86.93	1.15	2.1	na	46
		Waxy maize starch nanocrystals (1%)	86.71	1.11	2.29		
	100 ml H ₂ O)	Waxy maize starch nanocrystals (3%)	87.39	1.13	2.46		
		Waxy maize starch nanocrystals (5%)	86.81	1.08	2.55		
		Waxy maize starch nanocrystals (7%)	87.33	1.05	3.22		
		Waxy maize starch nanocrystals (9%)	86.79	1.07	4.01		
5	Amarnath protein isolate	Control (0%)	83.5	- 0.1	10.2	16.2	203
	(5% w/v	Normal maize starch nanocrystals (3%)	84.5	0	10	15.4	
	H_2O) + glycerol (1.25%)	Normal maize starch nanocrystals (6%)	83.3	- 0.2	12	17.4	
	w/v H ₂ O)	Normal maize starch nanocrystals (9%)	82.6	- 0.2	11.9	17.9	
		Normal maize starch nanocrystals (12%)	82.5	- 0.1	13.8	20.2	
		Waxy maize starch nanocrystals (3%)	81.5	- 0.1	13.2	19.6	
		Waxy maize starch nanocrystals (6%)	80.7	- 0.2	14.5	21	
		Waxy maize starch nanocrystals (9%)	79.7	- 0.1	16.1	22.7	
		Waxy maize starch nanocrystals (12%)	81.1	- 0.2	14.1	20.5	
6	Agar (3 g/150 ml	Nanocellulose (0%)	92.7	-0.58	4.9	2.74	101
	$H_2O) + glycerol (0.9 g/$	Nanocellulose (1%)	92.93	-0.62	4.93	2.64	
	150 ml H ₂ O)	Nanocellulose (3%)	92.71	-0.67	5.18	2.95	
		Nanocellulose (5%)	92.71	-0.70	5.31	3.05	
		Nanocellulose (10%)	92.63	-0.74	5.53	3.28	
7	Carrageenan (3 g/100 ml	Chitin nanofibrils (0%)	93.4	-0.34	4.1	3.52	173
	$H_2O) + glycerol (1.2 g/$	Chitin nanofibrils (3%)	93.2	-0.34	4.21	3.95	
	100 ml soln)	Chitin nanofibrils (5%)	92.5	-0.50	4.5	4.63	
	*	Chitin nanofibrils (10%)	91.8	-0.58	4.7	5.09	

na Data not available

issues, such as poor mechanical strength and poor flexibility as compared to the synthetic plastics, which prevents their commercialization [201].

Barrier and mechanical properties play a vital role for food packaging materials for protecting and carrying the packaged food products from external factors. There is no specific legislation available on the material properties of such food packaging materials. However, a conventional requirement for a food packaging material is that the tensile strength should be more than 3.5 MPa. In order to evaluate whether a new or modified films meet the requirements of other physical properties, those properties are usually compared with the physical properties of the commonly used synthetic packaging materials, such as such as LDPE (low density polyethylene), HDPE (high density polyethylene), PP (polypropylene) and PET (polyethylene terephthalate) [202].

The mechanical properties of edible films are important for food packaging applications. As poor mechanical properties may lead to poor flexibility and premature failure during its production, handling, storage or use, higher mechanical properties are expected from edible films. For films, puncture strength (ASTM F1306), seal strength (ASTM F88) and tear strength (ASTM D1922) are considered as important mechanical properties. However, most investigations focused on tensile tests, where ultimate tensile strength, elastic modulus of elasticity and percentage of elongation at break of edible films were calculated [203].

Most research works had that the nanoscale fillers were reinforced onto macroscale edible materials to produce nanoedible films. Only limited research was carried out to find the effects of nanomaterials as both matrix and fillers to make the complete nanoedible films. In the work of Taniguchi and Okamura [204], they produced direct nanocellulose films (of about 3–100 microns thickness) from cellulose nanofibrils of about 20-90 nm. The cellulose nanofibrils nanofilms were prepared from wood pulp, tunicin cellulose and a mixture of wood pulp, chitosan and collagen, and the mechanical properties of these films were compared with those of commercial print grade paper and LDPE films. The mechanical propteries of nanofilms were found to be 2-5 times higher than those of the print grade paper and LDPE films. Among, wood pulp, tunicin cellulose and hybrid composite, tunicin showed the highest tensile strength. The results showed that tensile strengths of the films obtained from nanoparticles are higher than the regular films. Leitner et al. [205] produced nanocellulose films from sugar beet pulp using solution casting method, and the films exhibited TS of 104 MPa and YM of 9 GPa, respectively. In another work by Shao et al. [206], the composite solutions (of pullulan, carboxymethylcellulose sodium and tea polyphenols) with different concentrations (0.5%, 1%, 1%)1.5%, w/v) of tea polyphenols were electrospun onto aluminium foil in order to produce the nanocomposite nanofibre films. When the concentration of tea polyphenols is lower, nanofibres of smaller sizes were produced (for example, average diameter is 127 nm at 1% concentration of tea polyphenols). When coated over strawberries, these edible coatings could reduce the weight loss and improve the firmness, quality and shelf life of the strawberries during the fruit storage. However, these kinds of research are rarely found in the literatures. Future investigation is required to produce and investigates on complete nanomaterials-based edible films.

Lack of evidence on biodegradability and edibility

The possible health risks associated with the use of food contained in the nanofilms are not yet clearly understood [207]. The use of nanoedible films is the right candidate for packaging materials where the food has direct contact with the packaging materials. Most of the researchers have proposed various nanobased edible films. However, these research studies did not consider the edibility test for evaluating edibility of such films. Rather, the edibility of such films was assumed from the edible nature of the materials used for the film preparation. If all materials used for the preparation of the edible films were edible, then the produced films were considered as edible [94]. The biodegradability of edible packaging materials themselves is susceptible, and their barrier and mechanical properties are stable for only a short period of time than synthetic packaging materials. Therefore, further investigation is required in the aspects of safety and stability of the intended functions [208].

The antimicrobial additives, added to the edible films and coatings, are approved by FDA, but some are not approved. It is also unsure whether some bacteria could develop resistance to these antimicrobial additives, which may cause food-borne diseases. There is a need of attention for reducing food-borne diseases due to the presence of antimicrobial additives [209]. The use of essential oils, as natural antimicrobials, is limited due to their poor aroma properties and toxicity problems. The essential oils, usually regarded as GRAS (Generally Regarded as Safe), are considered beneficial only at very low concentrations. But, higher dosage of essential oils may cause serious toxicity problems, as well as allergic problems [210].

Although most of the research works suggested that edible films and coatings could be eaten, a very little investigation was carried out on their digestibility. Film formation, film forming materials and additives, in fact, could largely modify their sensitivity to enzyme hydrolysis and digestibility [211]. Plasticizers, often necessary to improve the mechanical properties, weaken the hydrogen bonding and decrease the intermolecular attractions. Consequently, film flexibility and tensile strength decrease. The use of plasticizers also affects the digestion rate of the edible films and coatings. Starch, once considered as a completely digestible by human body, is not anymore. According to recent studies, the inclusion of additives can significantly modify the digestibility. Structural properties, composition, food form, molecular arrangement, chain length and degree of crystallinity are some of the reasons that determine the extent of enzyme digestion. In the work of Hernandez et al. [212], native banana (44%), potato (46%) and sagu (34%) starches showed significant resistance to enzyme digestion, while native maize starch showed high digestibility. However, the edible films produced from these starches contained completely different degree of digestibility. Films produced from banana (9.4%), potato (7.4%) and sagu (9.9%) showed a decrease in resistance to digestibility, while maize (5.4%) increased resistance to digestibility. An extensive study may be required in digestibility properties of edible films and coatings their for extended use in food packaging applications.

Scarcity of research

Most of the research works in the past on nanoedible films were carried to determine the effects of the addition of a nanofiller, such as nanocellulose, nanostarch, nanochitin, nanoprotein or nanolipid, on film properties of polysaccharides, proteins and/or lipids as matrix materials. Though isolation of nanofillers was carried out for most of the sources, the nanoedible films were prepared with only a few nanofillers. The effects of the rest of the nanofillers should be explored. The use of nanomaterials for both reinforcements and matrix was also not evaluated yet. Therefore, future investigations are required to explore clear understanding of the effects of the addition of nanomaterials to make the nanoedible films.

The packaging materials are often produced under high temperatures, which, in turn, can cause thermal decomposition of polymers to form volatile compounds that can migrate into food. For example, food-on-plastic trays, heated in a microwave oven, produce an off-flavour or odour. The reason for the odour is the volatiles compounds produced by decomposition of the plastic material or residual solvents left in the plastic trays. The sources of such migrants can be adhesives, residual monomers, food additives, residual solvents, inks for printing, coatings, decomposition products of polymers, etc. [213].

Several reports have indicated a high level of uncertainty in the prediction of toxicity effects of nanomaterials. There is a lack of knowledge about the toxicity of nanomaterials and the processes. On the other hand, few food products with unlabelled invisible nano-additives are already in the market place. However, a good understanding on the effects of nanomaterials is necessary in the aspect of human health and environment, before they are used in food packaging [132, 148, 214]. Institute of Food Science and Technology (IFST) recommend that nanomaterials should be considered as potentially harmful without clear evidences on their safety [215]. Nanomaterials, used to prepare edible films and coatings, exhibit significant improvement in the functional properties. However, there are safety issues. While the bulk properties of nanomaterials are well known (macroscale), the material properties might be different at nanoscale. If nanomaterials exhibit any toxicological effects, the nanomaterials could penetrate into the human body on consumption and resides in the body. Therefore, it is necessary to understand and have accurate information about the behaviour of nanomaterials on human health before they are available for usage [216–218].

The effects of polymer structure, orientation, degree of crystallinity, degree of cohesion, degree of adhesion, solvent temperature, pressure, salt, acid and base concentration (pH), free volume, additives, thickness, etc. might influence the properties of edible films. Only few researchers have worked on the effects of degree of cohesion and adhesion. However, the effects of other factors have not yet been explored, and it is essential to understand the film properties completely for their successful applications [182].

The future edible films and coatings can be multifunctional. The films and coatings should incorporate one or more functions, such as heat-proof, air-proof, anti-corrosion, insect prevention, disease-resistant, high film properties, etc. Furthermore, the currently available edible packaging materials could not satisfy the market requirements. A variety of packaging materials should be developed to meet the tailormade demands of the market [219].

Scale-up difficulties

Though the research towards nanocomposite films is constantly growing, the transition from the laboratory scale production into industrial-scale production is not easy. Different film preparation methods are proposed by the research community leading to nanocomposite films with different film properties. The size, source and fabrication methods of the nanomaterials and the matrix materials also affected the film properties. There are many problems faced with the existing fabrication methods, and there is a need of developing new or improved technology to reduce the production cost as well as the preparation time of such nanocomposite films [220, 221]. The selection of efficient isolation method and homogenous dispersion of nanofibres is the major challenges in the use of nanofibres. Even though reinforcement, of nanofibres is replaced by nanofibres, has shown environmental benefits, production of nanofibres is not considered economical due to heavy energy consumption. The use of different pre-treatments could be beneficial in order to reduce the energy consumption. Another problem is to use the efficient drying process that can reduce the drying time, because size change after drying has impact on its properties. Surface modifications, which can be used for improving the surface properties of the films, are (a) producing hydrophobic surface and (b) including ionic groups on fibre surface. The edibility of the nanofibres after surface modifications should be evaluated in order to ensure the safety of the films. Therefore, further research should be carried out in order to find efficient production methods, pretreatments, post-treatments, surface modifications and drying process [222]. The film properties, such as cohesion, adhesion and durability, depend on the composition of film material, coating method and drying method. For industrial applications, the film or coating material must meet certain requirement. The film must be applied or spread in wet condition on the food surface and the required film or coating must be formed on drying, while the film or coating has adequate film properties. Lack of attention, in these points, has resulted in unsatisfactory results in many cases [223].

Most of the lab-scale edible films are prepared in two steps: gelatinization followed by air-drying. Airdrying of edible films takes place through natural evaporation and hence is a very slow process (2–3 days). But, fairly rapid drying process should be developed in order to scale-up to industrial-scale. In the work of Kaya and Kaya [224], whey protein isolate-based edible films were dried using microwave drying (5 min) method or air-drying (18 h) method. The results showed that there was no significant difference in WVP of both drying methods, but the mechanical properties of microwave drying were higher than that of air-drying. In the work of Mayachiew et al. [225] and Thakhiew et al. [226], the film properties of the edible chitosan films were compared for three drying methods: (a) hot air-drying (at 40 °C), (b) vacuum drying (at 90 °C, 10 kPa) and (c) low-pressure superheated steam drying (at 90 °C, 10 kPa). The air-dried (at 30 °C) films were considered as the control. The results showed that vacuum and low-pressure superheated steam drying methods dried the unplasticized films at about 45 min and 60 min, respectively, which were much shorter drying time as compared to hot air-drying method (10 h) and ambient air-drying method (54 h). For the same drying method, the drying time increased with the glycerol concentrations. It was observed that the colour and WVP did not significantly differ with the drying methods, while the mechanical properties varied significantly. In the work of De Moraes and Laurindo [227], cellulose fibre-reinforced starchbased films were dried using three drying methods, (a) conduction drying, (b) infrared drying (42.3 Wm^{-2}) and (c) conduction-infrared drying with drying times of about 1 h, 30 min and 80 min, respectively. In the work of Ortiz et al. [228], the soy protein isolate (10.5% w/v)-based edible film was prepared using tape casting and the effects of two drying methods, namely, (a) heat conduction drying, (b) IR (42.3 Wm^{-2}) drying, were investigated. At 60 °C, the drying time for both the drying methods was found to be about 120 min, which was much lower than that of solution-cast films.

Despite being some progress in the reduction in total drying time of the edible films, the current progress is not sufficient for the industrial-scale production of edible films. Well-established fabrication techniques should be established so as to automate or continuously produce the edible films. One such continuous film fabrication was explained by Munhoz et al. [229] in which the pectin- and rindbased edible films were produced with IR pre-dryer followed by hot drying. The wet layer of film forming solution was first dried in a conveyor (at a speed of 12 cm/min) using a IR pre-dryer and then dried in an air circulating oven (of 80 cm length) at 120 °C. The total drying time was 7 min, and productivity was 0.03 m² film per min. However, the productivity of the edible films was very low. Research attention is required to establish low-cost fabrication methods to continuously produce the edible films with increased productivity.

Obstacles for commercialization

There are many issues in the commercialization of nanoedible films. First, the nanoedible films show poor barrier and mechanical properties as compared to that of the synthetic plastics. Secondly, the production of edible films and coatings is still in the laboratory level and is not yet expanded to industrial level due to their high cost of production. Research work is required to find new or better production methods for large scale production. Lack of awareness about edible materials is another hurdle in its utilization. For example, some people fear of accidental consumption of foreign particles and ingestion by children. Some marketing strategies, such as price discounts, customer attention towards edible packaged foods, consumption of edible packaged food items, etc. might be used for commercialization [208, 216, 230, 231]. Consumers accept edible films and coatings as long as they feel as safe and friendly [214]. The cost of edible films and coatings is 10 to 50 times higher than that made from polyethylene or polypropylene. However, the cost of edible films and coatings cannot be considered now, since the quantities are less used in the development phase [232]. The implementation cost of nanotechnology (including cost of nanomaterials, active ingredients and intelligent factors) will increase the total food packaging cost per package, leading to a low profit margin of the product cost. The total cost of the packaging should be less than 10% of the product cost. Therefore, proper cost-benefit analyses should be performed to justify the adaptation of nanotechnology [233].

The film forming materials and other additives, used to prepare the edible films and coatings, must be food grade and non-toxic; the equipments and process resources should meet a high level of standards for maintaining hygiene. Each country follows certain regulations defining the approved list of food additives. Europe regulations incorporated the use of fatty acids, fatty acid salts, shellac resins, pectin, lecithin, gums, beeswax, candelilla wax and carnauba wax, polysorbates into the edible films and coatings. US FDA (Food and Drug Administration) mentioned morpholine, polydextrose, sorbitan monostearate, sucrose fatty acid esters, cocoa butter and castor oil as materials used for making protective coatings on fruits and vegetables. In India, beeswax or carnauba wax might be coated on fruits and vegetable with proper labelling. People, for example vegetarians and others who avoid food made of animal products, may have concerns to use edible packaging materials containing animal fats and acids. Some people believe that edible films and coatings might attract pesticides that make food unsafe. If edible coatings are prepared from allergens, they could also cause allergic reactions. Some of the well-known allergens, which are used in edible packaging, include wheat, milk, fish, nuts, soy beans and peanuts. Thus, it is important to label the presence of any known allergens in the edible coating. Thus, it becomes essential to emphasize producers and packers to label the necessary information about coating materials, so that the customers can avoid foods coated with edible materials [168, 234-236].

Summary

In this review article, recent research activities carried out in the fabrication of nanoedible films were discussed along with problems and opportunities for future research. Nanoedible films are so called when an edible nanofiller is reinforced with the edible matrix. From the discussion, it is clear that the addition of nanofillers improved the mechanical properties, film colour and WVP of the resulting edible films as compared to plain edible films. However, these properties are not adequate to compete with the properties of synthetic plastic films. For example, the nanoedible films showed very low %*E* making them highly fragile and show lower WVP at high RH conditions making them more hydrophilic. Hence, further investigation is required to find techniques to improve the film properties. The use of nanocellulose, starch nanocrystals,



nanochitin/nanochitason, soy/whey protein isolate nanocrystals and nanolipids has been reviewed in this review article. The nanoscale particles of other edible polymers, such as polysaccharides, proteins and lipids and their effects on edible films are not vet explored. The research carried out is also limited with the available nanofillers. Moreover, the effects of high proportions of nanofillers as well as completely nanofillers-based edible films were not much discussed in the literatures. There are also voids in the literatures related to degradability and edibility of films, lack of research, processing issues, scale-up issues and lack of awareness. Therefore, further research must be directed towards overcoming the above-said challenges and research deficiencies so that the nanoedible films will soon replace the synthetic plastics for food packaging.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

References

- Marsh K, Bugusu B (2007) Food packaging—roles, materials, and environmental issues. J Food Sci 72:R39– R55
- [2] Petersen K, Nielsen PV, Bertelsen G, Lawther M, Olsen MB, Nilsson NH, Mortensen G (1999) Potential of biobased materials for food packaging. Trends Food Sci Technol 10:52–68
- [3] Federation of Indian Chambers of Commerce and Industry (FICCI) (2016) A report on plastic industry. In: 2nd national conference on plastic packaging—the sustainable choice, pp 1–32
- [4] World Packaging Organization (2008) market statistics and future trends in global packaging. https://www.worldpack aging.org, pp 1–44. Accessed 7 Feb 2019
- [5] Hopewell J, Dvorak R, Kosior E (2009) Plastics recycling: challenges and opportunities. Philos Trans R Soc B 364:2115–2126
- [6] Kale G, Kijchavengkul T, Auras R, Rubino M, Selke SE, Singh SP (2007) Compostability of Bioplastic packaging materials: an overview. Macromol Biosci 7:255–277
- [7] Guilbert S, Gontard N, Cuq B (1995) Technology and applications of edible protective films. Packag Technol Sci 8:339–346
- [8] Shit SC, Shah PM (2014) Edible polymers: challenges and opportunities. J Polym Article ID 427259, 1–13

- [9] Jeevahan J, Chandrasekaran M, Durairaj RB, Mageshwaran G, Joseph GB (2017) A brief review on edible packing materials. J Glob Eng Probl Solut 1:9–19
- [10] Jeevahan J, Chandrasekaran M (2018) Effect of olive oil concentrations on film properties of edible composite films prepared from corn starch and olive oil. Res J Pharm Technol 11:4934–4938
- [11] Mellinas C, Valdés A, Ramos M, Burgos N, Garrigós MDC, Jiménez A (2016) Active edible films. Current state and future trends. J Appl Polym Sci 133:1–15
- [12] Bumbudsanpharoke N, Ko S (2015) Nano-food packaging: an overview of market, migration research, and safety regulations. J Food Sci 80:R910–R923
- Tsagkaris AS, Tzegkas SG, Danezis GP (2018) Nanomaterials in food packaging: state of the art and analysis. J Food Sci Technol 55:2862–2870
- [14] Cerqueira MA, Costa MJ, Fuciños C, Pastrana LM, Vicente AA (2013) Development of active and nanotechnologybased smart edible packaging systems: physical-chemical characterization. Food Bioprocess Technol 7:1472–1482
- [15] Pan K, Zhong Q (2016) Organic nanoparticles in foods: fabrication, characterization, and utilization. Annu Rev Food Sci Technol 7:245–266
- [16] Cerqueira MA, Vicente AA, Pastrana LM (2018) Nanotechnology in food packaging: opportunities and challenges. Nanomater Food Packag. https://doi.org/10.1016/b 978-0-323-51271-8.00001-2
- [17] Eleftheriadou M, Pyrgiotakis G, Demokritou P (2017) Nanotechnology to the rescue: using nano-enabled approaches in microbiological food safety and quality. Curr Opin Biotechnol 44:87–93
- [18] De Azeredo MCH (2009) Nanocomposites for food packaging applications. Food Res Int 42:1240–1253
- [19] Arora A, Padua GW (2010) Review: nanocomposites in food packaging. J Food Sci 75:R43–R49
- [20] Reddy MM, Vivekanandhan S, Misra M, Bhatia SK, Mohanty AK (2013) Biobased plastics and bionanocomposites: current status and future opportunities. Prog Polym Sci 38:1653–1689
- [21] Attaran SA, Hassan A, Wahit MU (2015) Materials for food packaging applications based on bio-based polymer nanocomposites. J Thermoplast Compos Mater 30:143–173
- [22] Sarkar P, Choudhary R, Panigrahi S, Syed I, Sivapratha S, Dhumal CV (2017) Nano-inspired systems in food technology and packaging. Environ Chem Lett 15:607–622
- [23] Kumar N, Kaur P, Bhatia S (2017) Advances in bionanocomposite materials for food packaging: a review. Nutr Food Sci 47:591–606. https://doi.org/10.1108/nfs-11-2016-0176

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- [24] Vasile C (2018) Polymeric nanocomposites and nanocoatings for food packaging: a review. Materials 11:1–49. h ttps://doi.org/10.3390/ma11101834
- [25] Zambrano-Zaragoza M, González-Reza R, Mendoza-Muñoz N, Miranda-Linares V, Bernal-Couoh T, Mendoza-Elvira S, Quintanar-Guerrero D (2018) Nanosystems in edible coatings: a novel strategy for food preservation. Int J Mol Sci 19:1–24. https://doi.org/10.3390/ijms19030705
- [26] Haniffa MM, Ching Y, Abdullah L, Poh S, Chuah C (2016) Review of bionanocomposite coating films and their applications. Polymers 8:1–33. https://doi.org/10.3390/ polym8070246
- [27] Sharma B, Malik P, Jain P (2018) Biopolymer reinforced nanocomposites: a comprehensive review. Mater Today Commun 16:353–363
- [28] Shapi'i RA, Othman SH (2015) edible nanofiller for development of edible bionanocomposite film: a review. J Polym Sci Technol 1:37–45
- [29] Le Corre D, Bras J, Dufresne A (2010) Starch nanoparticles: a review. Biomacromol 11:1139–1153
- [30] Dufresne A, Cavaillé JY, Helbert W (1996) New nanocomposite materials: microcrystalline starch reinforced thermoplastic. Macromolecules 29:7624–7626
- [31] Putaux JL, Molina-Boisseau S, Momaur T, Dufresne A (2003) Platelet nanocrystals resulting from the disruption of waxy maize starch granules by acid hydrolysis. Biomacromol 4:1198–1202
- [32] Kim HY, Lee JH, Kim JY, Lim WJ, Lim ST (2012) Characterization of nanoparticles prepared by acid hydrolysis of various starches. Starch-Stärke 64:367–373
- [33] Kim HY, Park SS, Lim ST (2015) Preparation, characterization and utilization of starch nanoparticles. Colloids Surf B Biointerfaces 126:607–620
- [34] Kim HY, Park DJ, Kim JY, Lim ST (2013) Preparation of crystalline starch nanoparticles using cold acid hydrolysis and ultrasonication. Carbohydr Polym 98:295–301
- [35] Dufresne A (2014) Crystalline starch based nanoparticles. Curr Opin Colloid Interface Sci 19:397–408
- [36] Herrera MP, Vasanthan T, Chen L (2017) Rheology of starch nanoparticles as influenced by particle size, concentration and temperature. Food Hydrocolloids 66:237–245
- [37] Gonçalves PM, Noreña CPZ, da Silveira NP, Brandelli A (2014) Characterization of starch nanoparticles obtained from Araucaria angustifolia seeds by acid hydrolysis and ultrasound. LWT Food Sci Technol 58:21–27
- [38] Sun Q, Li G, Dai L, Ji N, Xiong L (2014) Green preparation and characterisation of waxy maize starch nanoparticles through enzymolysis and recrystallisation. Food Chem 162:223–228

- [39] Shi A, Li D, Wang L, Li B, Adhikari B (2011) Preparation of starch-based nanoparticles through high-pressure homogenization and miniemulsion cross-linking: influence of various process parameters on particle size and stability. Carbohydr Polym 83:1604–1610
- [40] Qiu S, Li Y, Chen H, Liu Y, Yin L (2014) Effects of highpressure homogenization on thermal and electrical properties of wheat starch. J Food Eng 128:53–59
- [41] Haaj SB, Magnin A, Pétrier C, Boufi S (2013) Starch nanoparticles formation via high power ultrasonication. Carbohydr Polym 92:1625–1632
- [42] Juna S, Hayden S, Damm M, Kappe CO, Huber A (2013) Microwave mediated preparation of nanoparticles from wx corn starch employing nanoprecipitation. Starch-Stärke 66:316–325
- [43] Angellier H, Molina-Boisseau S, Dole P, Dufresne A (2006) Thermoplastic starch-waxy maize starch nanocrystals nanocomposites. Biomacromol 7:531–539
- [44] Shi A, Wang L, Li D, Adhikari B (2013) Characterization of starch films containing starch nanoparticles. Carbohydr Polym 96:593–601
- [45] Piyada K, Waranyou S, Thawien W (2013) Mechanical, thermal and structural properties of rice starch films reinforced with rice starch nanocrystals. Int Food Res J 20:439–449
- [46] Li X, Qiu C, Ji N, Sun C, Xiong L, Sun Q (2015) Mechanical, barrier and morphological properties of starch nanocrystals-reinforced pea starch films. Carbohydr Polym 121:155–162
- [47] Dai L, Qiu C, Xiong L, Sun Q (2015) Characterisation of corn starch-based films reinforced with taro starch nanoparticles. Food Chem 174:82–88
- [48] Liu C, Jiang S, Zhang S, Xi T, Sun Q, Xiong L (2015) Characterization of edible corn starch nanocomposite films: the effect of self-assembled starch nanoparticles. Starch-Stärke 68:239–248
- [49] González A, Igarzabal CIA (2015) Nanocrystal-reinforced soy protein films and their application as active packaging. Food Hydrocolloids 43:777–784
- [50] Condés MC, Añón MC, Mauri AN, Dufresne A (2015) Amaranth protein films reinforced with maize starch nanocrystals. Food Hydrocolloids 47:146–157
- [51] Jiang S, Liu C, Wang X, Xiong L, Sun Q (2016) Physicochemical properties of starch nanocomposite films enhanced by self-assembled potato starch nanoparticles. LWT Food Sci Technol 69:251–257
- [52] Oliveira AV, da Silva APM, Barros MO, Filho MDMS, Rosa MF, Azeredo HMC (2018) Nanocomposite films from mango kernel or corn starch with starch nanocrystals.

Starch-Stärke 70:1–7. https://doi.org/10.1002/star. 201800028

- [53] Klemm D, Kramer F, Moritz S, Lindström T, Ankerfors M, Gray D, Dorris A (2011) Nanocelluloses: a new family of nature-based materials. Angew Chem Intl Ed 50:5438–5466
- [54] Afrin S, Karim Z (2017) Isolation and surface modification of nanocellulose: necessity of enzymes over chemicals. ChemBioEng Rev 4:1–16
- [55] Mishra RK, Sabu A, Tiwari SK (2018) Materials chemistry and the futurist eco-friendly applications of nanocellulose: status and prospect. J Saudi Chem Soc 22:949–978. http s://doi.org/10.1016/j.jscs.2018.02.005
- [56] Gómez HC, Serpa A, Velásquez-Cock J, Gañán P, Castro C, Vélez L, Zuluaga R (2016) Vegetable nanocellulose in food science: a review. Food Hydrocolloids 57:178–186
- [57] Phanthong P, Reubroycharoen P, Hao X, Xu G, Abudula A, Guan G (2018) Nanocellulose: extraction and application. Carbon Resour Convers 1:32–43
- [58] Lee HV, Hamid SBA, Zain SK (2014) Conversion of lignocellulosic biomass to nanocellulose: structure and chemical process. Sci World J 2014:1–20
- [59] George J, Sabapathi SN (2015) Cellulose nanocrystals: synthesis, functional properties, and applications. Nanotechnol Sci Appl 2015(8):45–54
- [60] Ludueña L, Fasce D, Alvarez VA, Stefani PM (2011) Nanocellulose from rice husk following alkaline treatment to remove silica. BioResources 6:1440–1453
- [61] Rosa SML, Rehman N, Miranda MIGMd, Nachtigall SMB, Bica CID (2012) Chlorine-free extraction of cellulose from rice husk and whisker isolation. Carbohydr Polym 87:1131–1138
- [62] Jiang F, Hsieh YL (2013) Chemically and mechanically isolated nanocellulose and their self-assembled structures. Carbohydr Polym 95:32–40
- [63] Abraham E, Deepa B, Pothan LA, Jacob M, Thomas S, Cvelbar U, Anandjiwala R (2011) Extraction of nanocellulose fibrils from lignocellulosic fibres: A novel approach. Carbohydr Polym 86:1468–1475
- [64] Mueller S, Weder C, Foster EJ (2014) Isolation of cellulose nanocrystals from pseudostems of banana plants. RSC Adv 4:907–915
- [65] Cherian BM, Leão AL, Souza SFd, Thomas S, Pothan LA, Kottaisamy MK (2010) Isolation of nanocellulose from pineapple leaf fibres by steam explosion. Carbohydr Polym 81:720–725
- [66] Alemdar A, Sain M (2008) Biocomposites from wheat straw nanofibers: morphology, thermal and mechanical properties. Compos Sci Technol 68:557–565

- [67] Babaee M, Jonoobi M, Hamzeh Y, Ashori A (2015) Biodegradability and mechanical properties of reinforced starch nanocomposites using cellulose nanofibers. Carbohydr Polym 132:1–8. https://doi.org/10.1016/j.carbpol.201 5.06.043
- [68] Dufresne A, Vignon MR (1998) Improvement of starch film performances using cellulose microfibril. Macromolecules 31:2693–2696
- [69] Liu C, Li B, Du H, Dong L, Zhang Y, Yu G, Mu X, Peng H (2016) Properties of nanocellulose isolated from corncob residue using sulfuric acid, formic acid, oxidative and mechanical methods. Carbohydr Polym 151:716–724
- [70] Li R, Fei J, Cai Y, Li Y, Feng J, Yao J (2009) Cellulose whiskers extracted from mulberry: a novel biomass production. Carbohydr Polym 76:94–99
- [71] Beck-Candanedo S, Roman M, Gray DG (2005) Effect of reaction conditions on the properties and behavior of wood cellulose nanocrystal suspensions. Biomacromolecules 6:1048–1054
- [72] Araki J, Wada M, Kuga S (2001) Steric stabilization of a cellulose microcrystal suspension by poly (ethylene glycol) grafting. Langmuir 17:21–27
- [73] Morais JPS, Rosa MdF, Filho MMS, Nascimento LD, Nascimento DMd, Cassales AR (2013) Extraction and characterization of nanocellulose structures from raw cotton linter. Carbohydr Polym 91:229–235
- [74] Menezes AJd, Siqueira G, Curvelo AA, Dufresne A (2009) Extrusion and characterization of functionalized cellulose whiskers reinforced polyethylene nanocomposites. Polymer 50:4552–4563
- [75] Rodriguez NLGd, Thielemans W, Dufresne A (2006) Sisal cellulose whiskers reinforced polyvinyl acetate nanocomposites. Cellulose 13:261–270
- [76] Moran JI, Alvarez VA, Cyras VP, Vazquez A (2008) Extraction of cellulose and preparation of nanocellulose from sisal fibers. Cellulose 15:149–159
- [77] Martelli-Tosi M, Torricillas MdS, Martins MA, Odílio Assis OBGd, Tapia-Blácido RT (2016) Using commercial enzymes to produce cellulose nanofibers from soybean straw. J Nanomater 2016:678–687. https://doi.org/10.1155/ 2016/8106814
- [78] Mandal A, Chakrabarty D (2011) Isolation of nanocellulose from waste sugarcane bagasse (SCB) and its characterization. Carbohydr Polym 86:1291–1299
- [79] Wulandari WT, Rochliadi A, Arcana IM (2016) Nanocellulose prepared by acid hydrolysis of isolated cellulose from sugarcane bagasse. In: IOP conference series: materials science and engineering 107: 012045, 1–7

- [80] Kimura F, Kimura T, Tamura M, Hirai A, Ikuno M, Horii F (2005) Magnetic alignment of the chiral nematic phase of a cellulose microfbril suspension. Langmuir 21:2034–2037
- [81] George J, Bawa AS, Siddaramaiah (2010) Synthesis and characterization of bacterial cellulose nanocrystals and their PVA nanocomposites. Adv Mater Res 123:383–386
- [82] George J, Siddaramaiah (2012) High performance edible nanocomposite flms containing bacterial cellulose nanocrystals. Carbohydr Polym 87:2031–2037
- [83] Tsukamoto J, Durán N, Tasic L (2013) Nanocellulose and bioethanol production from orange waste using isolated microorganisms. J Braz Chem Soc 24:1537–1543
- [84] Mariño M, Silva Ld, Durán N, Tasic L (2015) Enhanced materials from nature: nanocellulose from citrus waste. Molecules 20:5908–5923
- [85] Xu C, Zhu S, Xing C, Li D, Zhu N, Zhou H (2015) Isolation and properties of cellulose nanofibrils from coconut palm petioles by different mechanical process. PLoS ONE 10:1–11. https://doi.org/10.1371/journal.pone.0122123
- [86] Yongvanich N (2015) Isolation of nanocellulose from pomelo fruit fibers by chemical treatments. J Nat Fibers 12:323–331
- [87] Zhao J, Zhang W, Zhang X, Zhang X, Lu C, Deng Y (2013) Extraction of cellulose nanofibrils from dry softwood pulp using high shear homogenization. Carbohydr Polym 97:695–702
- [88] Li W, Zhao X, Huang Z, Liu S (2013) Nanocellulose fibrils isolated from BHKP using ultrasonication and their reinforcing properties in transparent poly (vinyl alcohol) films. J Polym Res 20:1–7
- [89] Wang H, Zhang X, Jiang Z, Yu Z, Yu Y (2016) Isolating nanocellulose fibrills from bamboo parenchymal cells with high intensity ultrasonication. Holzforschung 70:401–409
- [90] Li J, Wei X, Wang Q, Chen J, Chang G, Kong L, Su J, Liu Y (2012) Homogeneous isolation of nanocellulose from sugarcane bagasse by high pressure homogenization. Carbohydr Polym 90:1609–1613
- [91] Wang Y, Wei X, Li J, Wang Q, Wang F, Kong L (2013) Homogeneous isolation of nanocellulose from cotton cellulose by high pressure homogenization. J Mater Sci Chem Eng 1:49–52
- [92] Wang Y, Wei X, Li J, Wang F, Wang Q, Zhang Y, Kong L (2017) Homogeneous isolation of nanocellulose from eucalyptus pulp by high pressure homogenization. Ind Crops Prod 104:237–241
- [93] Cui S, Zhang S, Ge S, Xiong L, Sun Q (2016) Green preparation and characterization of size-controlled nanocrystalline cellulose via ultrasonic-assisted enzymatic hydrolysis. Ind Crops Prod 83:346–352

- [94] Hubbe MA, Ferrer A, Tyagi P, Yin Y, Salas C, Pal L, Rojas OJ (2017) Nanocellulose in thin films, coatings, and plies for packaging applications: a review. BioResources 12:2413–2433. https://doi.org/10.15376/biores.12.1.2143-2233
- [95] Charreau HL, Foresti M, Vazquez A (2013) Nanocellulose patents trends: a comprehensive review on patents on cellulose nanocrystals, microfibrillated and bacterial cellulose. Recent Pat Nanotechnol 7:56–80
- [96] Jozala AF, de Lencastre-Novaes LC, Lopes AM, Santos-Ebinuma VdC, Mazzola PG, Pessoa-Jr A, Grotto D, Gerenutti M, Chaud MV (2016) Bacterial nanocellulose production and application: a 10-year overview. Appl Microbiol Biotechnol 100:2063–2072
- [97] Azeredo HMC, Mattoso LHC, Avena-Bustillos RJ, Filho GC, Munford ML, Wood D, McHugh TH (2010) Nanocellulose reinforced chitosan composite films as affected by nanofiller loading and plasticizer content. J Food Sci 75:N1–N7
- [98] Bilbao-Sainz C, Bras J, Williams T, Sénechal T, Orts W (2011) HPMC reinforced with different cellulose nanoparticles. Carbohydr Polym 86:1549–1557
- [99] Shankar S, Rhim JW (2016) Preparation of nanocellulose from micro-crystalline cellulose: the effect on the performance and properties of agar-based composite films. Carbohydr Polym 135:18–26
- [100] Chaichi M, Hashemi M, Badii F, Mohammadi A (2017) Preparation and characterization of a novel bionanocomposite edible film based on pectin and crystalline nanocellulose. Carbohydr Polym 157:167–175
- [101] Viana RM, Sá NMSM, Barros MO, Borges MdF, Azeredo HMC (2018) Nanofibrillated bacterial cellulose and pectin edible films added with fruit purees. Carbohydr Polym 196:27–32
- [102] Wang X, Guo C, Hao W, Ullah N, Chen L, Li Z, Feng X (2018) Development and characterization of agar-based edible films reinforced with nano-bacterial cellulose. Int J Biol Macromol 118:722–730
- [103] Shabanpour B, Kazemi M, Ojagh SM, Pourashouri P (2018) Bacterial cellulose nanofibers as reinforce in edible fish myofibrillar protein nanocomposite films. Int J Biol Macromol 117:742–751
- [104] Ilyas RA, Sapuan SM, Ishak MR, Zainudin ES (2018) Development and characterization of sugar palm nanocrystalline cellulose reinforced sugar palm starch bionanocomposites. Carbohydr Polym 202:186–202. https://d oi.org/10.1016/j.carbpol.2018.09.002
- [105] Zeng JB, He YS, Li SL, Wang YZ (2011) Chitin whiskers: an overview. Biomacromolecules 13:1–11

- [106] Ifuku S (2014) Chitin and chitosan nanofibers: preparation and chemical modifications. Molecules 19:18367–18380
- [107] Mincea M, Negrulescu A, Ostafe V (2012) Preparation, modification, and applications of chitin nanowhiskers: a review. Rev Adv Mater Sci 30:225–242
- [108] Salaberria AM, Labidi J, Fernandes SCM (2015) Different routes to turn chitin into stunning nano-objects. Eur Polym J 68:503–515
- [109] Pereira AGB, Muniz EC, Hsieh YL (2014) Chitosan-sheath and chitin-core nanowhiskers. Carbohydr Polym 107:158–166
- [110] Ifuku S, Saimoto H (2012) Chitin nanofibers: preparations, modifications, and applications. Nanoscale 4:3308
- [111] Ji Y, Wolfe PS, Rodriguez IA, Bowlin GL (2012) Preparation of chitin nanofibril/polycaprolactone nanocomposite from a nonaqueous medium suspension. Carbohydr Polym 87:2313–2319
- [112] Fan Y, Saito T, Isogai A (2010) Individual chitin nanowhiskers prepared from partially deacetylated α-chitin by fibril surface cationization. Carbohydr Polym 79:1046–1051
- [113] Lertwattanaseri T, Ichikawa N, Mizoguchi T, Tanaka Y, Chirachanchai S (2009) Microwave technique for efficient deacetylation of chitin nanowhiskers to a chitosan nanoscaffold. Carbohydr Res 344:331–335
- [114] Wijesena RN, Tissera N, Kannangara YY, Lin Y, Amaratunga GAJ, de Silva KMN (2015) A method for top down preparation of chitosan nanoparticles and nanofibers. Carbohydr Polym 117:731–738
- [115] Lu Y, Sun Q, She X, Xia Y, Liu Y, Li J, Yang D (2013) Fabrication and characterisation of α -chitin nanofibers and highly transparent chitin films by pulsed ultrasonication. Carbohydr Polym 98:1497–1504
- [116] Chen C, Li D, Hu Q, Wang R (2014) Properties of polymethyl methacrylate-based nanocomposites: reinforced with ultra-long chitin nanofiber extracted from crab shells. Mater Des 1980–2015(56):1049–1056
- [117] Garcia I, Azcune I, Casuso P, Carrasco PM, Grande HJ, Cabañero G, Katsigiannopoulos D, Grana E, Dimos K, Karakassides MA, Odriozola I (2015) Carbon nanotubes/chitin nanowhiskers aerogel achieved by quaternization-induced gelation. J Appl Polym Sci 132:42547–1– 9
- [118] Fan Y, Fukuzumi H, Saito T, Isogai A (2012) Comparative characterization of aqueous dispersions and cast films of different chitin nanowhiskers/nanofibers. Int J Biol Macromol 50:69–76
- [119] De Moura MR, Aouada FA, Avena-Bustillos RJ, McHugh TH, Krochta JM, Mattoso LHC (2009) Improved barrier and mechanical properties of novel hydroxypropyl

methylcellulose edible films with chitosan/tripolyphosphate nanoparticles. J Food Eng 92:448–453

- [120] Ifuku S, Ikuta A, Egusa M, Kaminaka H, Izawa H, Morimoto M, Saimoto H (2013) Preparation of high-strength transparent chitosan film reinforced with surface-deacetylated chitin nanofibers. Carbohydr Polym 98:1198–1202
- [121] Ifuku S, Ikuta A, Izawa H, Morimoto M, Saimoto H (2014) Control of mechanical properties of chitin nanofiber film using glycerol without losing its characteristics. Carbohydr Polym 101:714–717
- [122] Antoniou J, Liu F, Majeed H, Zhong F (2015) Characterization of tara gum edible films incorporated with bulk chitosan and chitosan nanoparticles: a comparative study. Food Hydrocolloids 44:309–319
- [123] Hosseini SF, Rezaei M, Zandi M, Farahmandghavi F (2015) Fabrication of bio-nanocomposite films based on fish gelatin reinforced with chitosan nanoparticles. Food Hydrocolloids 44:172–182
- [124] Qin Y, Zhang S, Yu J, Yang J, Xiong L, Sun Q (2016) Effects of chitin nano-whiskers on the antibacterial and physicochemical properties of maize starch films. Carbohydr Polym 147:372–378
- [125] Vahedikia N, Garavand F, Tajeddin B, Cacciotti I, Jafari SM, Omidi T, Zahedi Z (2019) Biodegradable zein film composites reinforced with chitosan nanoparticles and cinnamon essential oil: physical, mechanical, structural and antimicrobial attributes. Biointerfaces Colloids Surf B 177:25–32. https://doi.org/10.1016/j.colsurfb.2019.01.045
- [126] Zubair M, Ullah A (2019) Recent advances in protein derived bionanocomposites for food packaging applications. Crit Rev Food Sci Nutr. https://doi.org/10.1080/104 08398.2018.1534800
- [127] DeFrates K, Markiewicz T, Gallo P, Rack A, Weyhmiller A, Jarmusik B, Hu X (2018) Protein polymer-based nanoparticles: fabrication and medical applications. Int J Mol Sci 19:1–20
- [128] Jahanshahi M, Babaei Z (2008) Protein nanoparticle: a unique system as drug delivery vehicles. Afr J Biotechnol 7:4926–4934
- [129] Sripriyalakshmi S, Jose P, Ravindran A, Anjali CH (2014) Recent trends in drug delivery system using protein nanoparticles. Cell Biochem Biophys 70:17–26
- [130] Davidov-Pardo G, Joye IJ, Espinal-Ruiz M, McClements DJ (2015) Effect of maillard conjugates on the physical stability of zein nanoparticles prepared by liquid antisolvent coprecipitation. J Agric Food Chem 63:8510–8518
- [131] Kasaai MR (2017) Zein and zein-based nanoparticles for food packaging applications: a global view. Adv Sci Eng Med 9:439–444

- [132] Kasaai MR (2018) Zein and zein- based nano-materials for food and nutrition applications: a review. Trends Food Sci Technol 79:184–197
- [133] Zwiorek K, Kloeckner J, Wagner E, Coester C (2005) Gelatin nanoparticles as a new and simple gene delivery system. J Pharm Pharm Sci 7:22–28
- [134] Yasmina R, Shaha M, Khan SA, Ali R (2017) Gelatin nanoparticles: a potential candidate for medical applications. Nanotechnol Rev 6:191–207
- [135] Wu J, Shi M, Li W, Zhao L, Wang Z, Yan X, Norde W, Li Y (2015) Pickering emulsions stabilized by whey protein nanoparticles prepared by thermal cross-linking. Colloids Surf B 127:96–104
- [136] Gülseren İ, Fang Y, Corredig M (2012) Whey protein nanoparticles prepared with desolvation with ethanol: characterization, thermal stability and interfacial behaviour. Food Hydrocolloids 29:258–264
- [137] Arzeni C, Pérez OE, Pilosof AMR (2015) Power ultrasound assisted design of egg albumin nanoparticles. Food Biophys 10:439–446
- [138] Taheri ES, Jahanshahi M, Mosavian MTH (2012) Preparation, characterization and optimization of egg albumin nanoparticles as low molecular-weight drug delivery vehicle. Part Part Syst Charact 29:211–222
- [139] Zhang J, Liang L, Tian Z, Chen L, Subirade M (2012) Preparation and in vitro evaluation of calcium-induced soy protein isolate nanoparticles and their formation mechanism study. Food Chem 133:390–399
- [140] Elzoghby A, Helmy Samy, Elgindy NA (2013) Novel ionically crosslinked casein nanoparticles for flutamide delivery: formulation, characterization, and in vivo pharmacokinetics. Int J Nanomed 8:1721–1732. https://doi.org/ 10.2147/ijn.s40674
- [141] Nakagawa K, Kagemoto M (2013) Characterization of casein-based nanoparticles formed upon freezing by in situ SAXS measurement. Colloids Surf B 103:366–374
- [142] Li X, Ji N, Qiu C, Xia M, Xiong L, Sun Q (2015) The effect of peanut protein nanoparticles on characteristics of protein- and starch-based nanocomposite films: a comparative study. Ind Crops Prod 77:565–574
- [143] Li KK, Yin SW, Yang XQ, Tang CH, Wei ZH (2012) Fabrication and characterization of novel antimicrobial films derived from thymol-loaded zein–sodium caseinate (SC) nanoparticles. J Agric Food Chem 60:11592–11600
- [144] Oymaci P, Altinkaya SA (2016) Improvement of barrier and mechanical properties of whey protein isolate based food packaging films by incorporation of zein nanoparticles as a novel bionanocomposite. Food Hydrocolloids 54:1–9

- [145] Zhang S, Zhao H (2017) Preparation and properties of zein-rutin composite nanoparticle/corn starch films. Carbohydr Polym 169:385–392
- [146] Gul O, Saricaoglu FT, Besir A, Atalar I, Yazici F (2018) Effect of ultrasound treatment on the properties of nanoemulsion films obtained from hazelnut meal protein and clove essential oil. Ultrason Sonochem 41:466–474
- [147] Aboul-Anean HED (2018) Using quinoa protein and starch nano particles to produce edible films. J Nut Health Food Eng 8:297–308
- [148] Sorrentino A, Gorrasi G, Vittoria V (2007) Potential perspectives of bio-nanocomposites for food packaging applications. Trends Food Sci Technol 18:84–95
- [149] Augustin MA, Sanguansri P (2009) Chapter 5 nanostructured materials in the food industry. Adv Food Nutr Res 58:183–213
- [150] Zambrano-Zaragoza ML, Mercado-Silva E, Ramirez-Zamorano P, Cornejo-Villegas MA, Gutiérrez-Cortez E, Quintanar-Guerrero D (2013) Use of solid lipid nanoparticles (SLNs) in edible coatings to increase guava (Psidium guajava L.) shelf-life. Food Res Int 51:946–953
- [151] Mashaghi S, Jadidi T, Koenderink G, Mashaghi A (2013) Lipid nanotechnology. Int J Mol Sci 14:4242–4282
- [152] Naser N, Valizadeh H, Zakeri-Milani P (2015) Solid lipid nanoparticles and nanostructured lipid carrieres: structure, preparation and application. Adv Pharm Bull 5:305–313
- [153] Espitia PJP, Fuenmayor CA, Otoni CG (2018) Nanoemulsions: synthesis, characterization, and application in biobased active food packaging. Compr Rev Food Sci Food Saf 18:264–285. https://doi.org/10.1111/1541-4337.12405
- [154] Amoabediny G, Haghiralsadat F, Naderinezhad S, Helder MN, Kharanaghi EA, Arough JM, Zandieh-Doulabi B (2017) Overview of preparation methods of polymeric and lipid-based (niosome, solid lipid, liposome) nanoparticles: a comprehensive review. Int J Polym Mater Polym Biomater 67:383–400
- [155] Acevedo-Fani A, Salvia-Trujillo L, Rojas-Graü MA, Martín-Belloso O (2015) Edible films from essential-oil-loaded nanoemulsions: physicochemical characterization and antimicrobial properties. Food Hydrocolloids 47:168–177
- [156] Wu J, Liu H, Ge S, Wang S, Qin Z, Chen L, Zheng Q, Liu Q, Zhang Q (2015) The preparation, characterization, antimicrobial stability and in vitro release evaluation of fish gelatin films incorporated with cinnamon essential oil nanoliposomes. Food Hydrocolloids 43:427–435
- [157] Cui H, Yuan L, Li W, Lin L (2016) Edible film incorporated with chitosan andArtemisia annuaoil nanoliposomes for inactivation of Escherichia coli O157: H7 on cherry tomato. Int J Food Sci Technol 52:687–698

- [158] Hashemi-Gahruie H, Ziaee E, Eskandari MH, Hosseini SMH (2017) Characterization of basil seed gum-based edible films incorporated with Zataria multiflora essential oil nanoemulsion. Carbohydr Polym 166:93–103
- [159] Robledo N, Vera P, López L, Yazdani-Pedram M, Tapia C, Abugoch L (2018) Thymol nanoemulsions incorporated in quinoa protein/chitosan edible films; antifungal effect in cherry tomatoes. Food Chem 246:211–219
- [160] Frank K, Garcia CV, Shin GH, Kim JT (2018) Alginate biocomposite films incorporated with cinnamon essential oil nanoemulsions: physical, mechanical, and antibacterial properties. Int J Polym Sci 2018:1519407–1–8
- [161] Restrepo AE, Rojas JD, Garcia OR, Sanchez LT, Pinzon MI, Villa CC (2018) Mechanical, barrier, and color properties of banana starch edible films incorporated with nanoemulsions of lemongrass (Cymbopogon citratus) and rosemary (Rosmarinus officinalis) essential oils. Food Sci Technol Int 24:705–712. https://doi.org/10.1177/ 1082013218792133
- [162] Crosby AJ, Lee J (2007) Polymer nanocomposites: the "nano" effect on mechanical properties. Polym Rev 47:217–229
- [163] Winey KI, Vaia RA (2007) Polymer nanocomposites. MRS Bull 32:314–322
- [164] Fiedler B, Gojny FH, Wichmann MHG, Nolte MCM, Schulte K (2006) Fundamental aspects of nano-reinforced composites. Compos Sci Technol 66:3115–3125
- [165] Guo D, Xie G, Luo J (2013) Mechanical properties of nanoparticles: basics and applications. J Phys D Appl Phys 47:013001-1–013001-25
- [166] Cuenot S, Frétigny C, Demoustier-Champagne S, Nysten B (2004) Surface tension effect on the mechanical properties of nanomaterials measured by atomic force microscopy. Phys Rev B 69:165410-1-5. https://doi.org/10.1103/physre vb.69.165410
- [167] ASTM D882–91 (1996) Standard test methods for tensile properties of thin plastic sheeting, annual book of ASTM. American Society for Testing and Materials, Philadelphia
- [168] Bertuzzi MA, Vidaurre EFC, Armada M, Gottifredi JC (2007) Water vapor permeability of edible starch based films. J Food Eng 80:972–978
- [169] Bertuzzi MA, Gottifredi JC, Armada M (2012) Mechanical properties of a high amylose content corn starch based film, gelatinized at low temperature. Braz J Food Technology 15:219–227
- [170] Sahraee S, Milani JM, Ghanbarzadeh B, Hamishehkar H (2017) Physicochemical and antifungal properties of bionanocomposite film based on gelatin-chitin nanoparticles. Int J Biol Macromol 97:373–381

- [171] Shankar S, Reddy JP, Rhim JW, Kim HY (2015) Preparation, characterization, and antimicrobial activity of chitin nanofibrils reinforced carrageenan nanocomposite films. Carbohydr Polym 117:468–475
- [172] Nielsen LE (1967) Models for the permeability of filled polymer systems. J Macromol Sci Part A Chem 1:929–942
- [173] Silvestre C, Duraccio D, Cimmino S (2011) Food packaging based on polymer nanomaterials. Prog Polym Sci 36:1766–1782
- [174] Lape NK, Nuxoll EE, Cussler EL (2004) Polydisperse flakes in barrier films. J Membr Sci 236:29–37
- [175] Aris R (1986) On a problem in hindered diffusion. Arch Ration Mech Anal 95:83–91
- [176] Cussler EL, Hughes SE, Ward WJ, Aris R (1988) Barrier membranes. J Membr Sci 38:161–174
- [177] Fredrickson GH, Bicerano J (1999) Barrier properties of oriented disk composites. J Chem Phys 110:2181–2188
- [178] Gusev AA, Lusti HR (2001) Rational design of nanocomposites for barrier applications. Adv Mater 13:1641–1643
- [179] Bharadwaj RK (2001) Modeling the barrier properties of polymer-layered silicate nanocomposites. Macromolecules 34:9189–9192
- [180] Wolf C, Angellier-Coussy H, Gontard N, Doghieri F, Guillard V (2018) How the shape of fillers affects the barrier properties of polymer/non-porous particles nanocomposites: a review. J Membr Sci 556:393–418
- [181] Park HJ, Chinnan MS (1995) Gas and water vapor barrier properties of edible films from protein and cellulosic materials. J Food Eng 25:497–507
- [182] Miller KS, Krochta JM (1997) Oxygen and aroma barrier properties of edible films: a review. Trends Food Sci Technol 8:228–237
- [183] ASTM E96–95 (1995) Standard test methods for water vapor transmission of material, annual book of ASTM. American Society for Testing and Materials, Philadelphia
- [184] Liz-Marzán LM (2004) Nanometals. Mater Today 7:26-31
- [185] Kooij SE, Poelsema B (2006) Shape and size effects in the optical properties of metallic nanorods. Phys Chem Chem Phys 8:3349–3357. https://doi.org/10.1039/b518389h
- [186] Choudhury AKR (2014) Object appearance and colour. In: Principles of colour and appearance measurement, pp 53–102. https://doi.org/10.1533/9780857099242.53
- [187] Schmidt G, Malwitz MM (2003) Properties of polymernanoparticle composites. Curr Opin Colloid Interface Sci 8:103–108
- [188] Ray PC (2010) Size and Shape dependent second order nonlinear optical properties of nanomaterials and their application in biological and chemical sensing. Chem Rev 110:5332–5365

Deringer

- [189] Quinten M (2001) The color of finely dispersed nanoparticles. Appl Phys B Lasers Opt 73:317–326
- [190] Nobbmann U, Morfesis A (2009) Light scattering and nanoparticles. Mater Today 12:52–54
- [191] Yano H, Sugiyama J, Nakagaito AN, Nogi M, Matsuura T, Hikita M, Handa K (2005) Optically transparent composites reinforced with networks of bacterial nanofibers. Adv Mater 17:153–155
- [192] Althues H, Henle J, Kaskel S (2007) Functional inorganic nanofillers for transparent polymers. Chem Soc Rev 36:1454–1465. https://doi.org/10.1039/b608177k
- [193] Hunter RS (1942) Photoelectric tristimulus colorimetry with three filters. JOSA 32:509–538
- [194] Ghasemlou M, Aliheidari N, Fahmi R, Shojaee-Aliabadi S, Keshavarz B, Cran MJ, Khaksar R (2013) Physical, mechanical and barrier properties of corn starch films incorporated with plant essential oils. Carbohydr Polym 98:1117–1126
- [195] Flores S, Fama L, Rojas AM, Goyanes S, Gerschenson L (2007) Physical properties of tapioca-starch edible films: influence of filmmaking and potassium sorbate. Food Res Int 40:257–265
- [196] McLaren K (1976) XIII—the development of the CIE 1976 (L* a* b*) uniform colour space and colour-difference formula. J Soc Dyers Colour 92:338–341
- [197] Robertson AR (1977) The CIE 1976 color-difference formulae. Color Res Appl 2:7–11
- [198] Afshari-Jouybari H, Farahnaky A (2011) Evaluation of Photoshop software potential for food colorimetry. J Food Eng 106:170–175
- [199] Caglar A, Yamanel K, Gulsahi K, Bagis B, Ozcan M (2010) Could digital imaging be an alternative for digital colorimeters? Clin Oral Investig 14:713–718
- [200] Farahnaky A, Saberi B, Majzoobi M (2013) Effect of glycerol on physical and mechanical properties of wheat starch edible films. J Texture Stud 44:176–186
- [201] Hosseini SF, Gómez-Guillén MC (2018) A state-of-the-art review on the elaboration of fish gelatin as bioactive packaging: special emphasis on nanotechnology-based approaches. Trends Food Sci Technol 79:125–135
- [202] Azeredo HMC, Rosa MF, Mattoso LHC (2017) Nanocellulose in bio-based food packaging applications. Ind Crops Prod 97:664–671
- [203] Otoni CG, Avena-Bustillos RJ, Azeredo HMC, Lorevice MV, Moura MR, Mattoso LHC, McHugh TH (2017) Recent advances on edible films based on fruits and vegetables—a review. Compr Rev Food Sci Food Saf 16:1151–1169
- [204] Taniguchi T, Okamura K (1998) New films produced from microfibrillated natural fibres. Polym Int 47:291–294

- [205] Leitner J, Hinterstoisser B, Wastyn M, Keckes J, Gindl W (2007) Sugar beet cellulose nanofibril-reinforced composites. Cellulose 14:419–425
- [206] Shao P, Niu B, Chen H, Sun P (2018) Fabrication and characterization of tea polyphenols loaded pullulan-CMC electrospun nanofiber for fruit preservation. Int J Biol Macromol 107:1908–1914
- [207] Souza VGL, Fernando AL (2016) Nanoparticles in food packaging: biodegradability and potential migration to food—a review. Food Packag Shelf Life 8:63–70
- [208] Janjarasskul T, Krochta JM (2010) Edible packaging materials. Annu Rev Food Sci Technol 1:415–448
- [209] Cooksey K (2001) Antimicrobial food packaging materials. Addit Polym 2001:6–10
- [210] Sanchez-Gonzalez L, Vargas M, Gonzalez-Martinez C, Chiralt A, Chafer M (2011) Use of essential oils in bioactive edible coatings. Food Eng Rev 3:1–16
- [211] Cuq B, Gontard N, Guilbert S (1998) Proteins as agricultural polymers for packaging production. Cereal Chem 75:1–9
- [212] Hernandez O, Emaldi U, Tovar J (2008) In vitro digestibility of edible films from various starch sources. Carbohydr Polym 71:648–655
- [213] Kim-Kang H (1990) Volatiles in packaging materials. Crit Rev Food Sci Nutr 29:255–271
- [214] Vargas M, Pastor C, Chiralt A, McClements DJ, González-Martínez C (2008) Recent advances in edible coatings for fresh and minimally processed fruits. Crit Rev Food Sci Nutr 48:496–511
- [215] Weiss J, Takhistov P, McClements J (2006) Functional materials in food nanotechnology. J Food Sci 71:R107– R116
- [216] Falguera V, Quintero JP, Jimenez A, Munoz JA, Ibarz A (2011) Edible films and coatings: structures, active functions and trends in their use. Trends Food Sci Technol 22:292–303
- [217] Magnuson BA, Jonaitis TS, Card JW (2011) A brief review of the occurrence, use, and safety of food-related nanomaterials. J Food Sci 76:R126–R133
- [218] Sekhon SB (2010) Food nanotechnology—an overview. Nanotechnol Sci Appl 3:1–15
- [219] Zhang S, Gu W, Cheng Z, Li YY, Gu W (2014) Development of Edible Packaging Materials. Adv Mater Res 904:189–191
- [220] Siqueira G, Bras J, Dufresne A (2010) Cellulosic bionanocomposites: a review of preparation. Prop Appl Polym 2:728–765
- [221] Solans C, Solé I (2012) Nano-emulsions: formation by lowenergy methods. Curr Opin Colloid Interface Sci 17:246–254

- [222] Abdul-Khalil HPS, Davoudpour Y, Islam MN, Mustapha A, Sudesh K, Dungani R, Jawaid M (2014) Production and modification of nanofibrillated cellulose using various mechanical processes: a review. Carbohydr Polym 99:649–665
- [223] Krochta JM, Mulder-Johnston CD (1997) Edible and biodegradable polymer films: challenges and opportunities. Food Technol 51:61–74
- [224] Kaya S, Kaya A (2000) Microwave drying effects on properties of whey protein isolate edible films. J Food Eng 43:91–96
- [225] Mayachiew P, Devahastin S (2008) Comparative evaluation of physical properties of edible chitosan films prepared by different drying methods. Dry Technol 26:176–185
- [226] Thakhiew W, Devahastin S, Soponronnarit S (2010) Effects of drying methods and plasticizer concentration on some physical and mechanical properties of edible chitosan films. J Food Eng 99:216–224
- [227] De Moraes JO, Laurindo JB (2017) Properties of starchcellulose fiber films produced by tape casting coupled with infrared radiation. Dry Technol 36:830–840
- [228] Ortiz CM, de Moraes JO, Vicente AR, Laurindo JB, Mauri AN (2017) Scale-up of the production of soy (Glycine max L.) protein films using tape casting: formulation of filmforming suspension and drying conditions. Food Hydrocolloids 66:110–117
- [229] Munhoz DR, Moreira FKV, Bresolin JD, Bernardo MP, De Sousa CP, Mattoso LHC (2018) Sustainable production and in vitro biodegradability of edible films from yellow

passion fruit coproducts via continuous casting. ACS Sustain Chem Eng 6:9883–9892

- [230] Ozdemir M, Floros JD (2004) Active food packaging technologies. Crit Rev Food Sci Nutr 44:185–193
- [231] Abdul-Khalil HPS, Davoudpour Y, Saurabh CK, Hossain MS, Adnan AS, Dungani R, Haafiz MKM (2016) A review on nanocellulosic fibres as new material for sustainable packaging: process and applications. Renew Sustain Energy Rev 64:823–836
- [232] Debeaufort F, Quezada-Gallo JA, Voilley A (1998) Edible films and coatings: tomorrow's packagings: a review. Crit Rev Food Sci Nutr 38:299–313
- [233] Mihindukulasuriya SDF, Lim LT (2014) Nanotechnology development in food packaging: a review. Trends Food Sci Technol 40:149–167
- [234] Dhall RK (2013) Advances in edible coatings for fresh fruits and vegetables: a review. Crit Rev Food Sci Nutr 53:435–450
- [235] Hannon JC, Kerry J, Cruz-Romero M, Morris M, Cummins E (2015) Advances and challenges for the use of engineered nanoparticles in food contact materials. Trends Food Sci Technol 43:43–62
- [236] Han JW, Ruiz-Garcia L, Qian JP, Yang XT (2018) Food packaging: a comprehensive review and future trends. Compr Rev Food Sci Food Saf 17:860–877

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