

# Chapter 5

## Recent Studies on Soy Protein Based Blends, Composites and Nanocomposites

Lucia H. Innocentini-Mei and Farayde Matta Fakhouri

### 5.1 Introduction

With the environmental appeal around the planet for a sustainable development, there is the need to develop new materials from renewable resources, which can be degraded in a short time in the environment, thereby maintaining the proper balance of the carbon cycle. The utilization of hydrocolloids, such as soy protein, to prepare biodegradable materials with suitable properties, has been a great challenge for the scientific community, since these materials do not possess all the desirable characteristics of the synthetic polymers, being mostly often, highly hydrophilic and also presenting poor mechanical properties to be used as engineering's materials. In this context, the studies with application of nanotechnology to biodegradable polymers can open new possibilities to improve not only the properties of these materials, but also its efficiency.

Although the oil is important as energy source, its main utilization is directed toward the industrial production of raw material for packaging. For those industries that manufacture plastics packaging's, there is an enormous challenge to ensure the sustainability of the raw material since the conventional plastics, when discarded, will take decades or centuries to degrade. Among the most used plastic, in food packaging industries, are: (1) low density polyethylene (LDPE), which has low cost and is used in general to pack dehydrated products, pasteurized milk and in the manufacture of laminated packing; (2) high density polyethylene (HDPE), which is mainly used to produce plastic bags for supermarkets, (3) polystyrene (PS), used in the preparation of expanded trays (Styrofoam) for various food

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L. H. Innocentini-Mei (✉) · F. M. Fakhouri  
School of Chemical Engineering, State University of Campinas—UNICAMP, Av. Albert Einstein 500, CP 6066 13083-970 Campinas, São Paulo, Brazil  
e-mail: lumei@feq.unicamp.br

products; (4) poly vinyl chloride (PVC), widely used for packaging of vinegar, mineral water, edible oils, sauces and food wraps, among others; (5) oriented and bi-oriented polypropylene (PP), used for foods containing high fat content, possess excellent gloss and transparency; (6) poly ethylene terephthalate (PET), used mainly in carbonated beverages and (7) poly ethylene naphthalate (PEN), which has more specific applications than PET, such as fruit juices and preserves beer, wine and returnable packaging, due to its high price [1]. The four alternatives to treat plastic waste are incineration, recycling, landfills and biodegradation. The incineration of plastic waste will always produce a large amount of carbon dioxide causing global warming and sometimes producing toxic gases that contribute to environmental pollution [2]. Furthermore, recycling is a great option in this context, but requires education on the proper disposal of garbage and a subsequent processing to obtain the recycled plastic, which leads to higher cost packaging with inferior quality compared to those plastics produced with virgin resin. To attend the growing appeal for environmentally friendly materials to replace some traditional plastics packages, that take decades or centuries to decompose in landfills and dumps, many scientists around the world have dedicated themselves to transform the raw material from renewable sources in packages which have good properties and which have a very short life after its discharge [2].

The properties required for plastic packaging mainly depend on the alterations presented by the product being protected and the conditions where it is stored. When a flexible packaging is required for food packing, the bioplastic must: (1) present good sensory properties of gas barrier and mechanical properties, (2) sufficient biochemical, physical-chemical and microbiological stability, (3) be free of toxic and be safe for consumption, (4) has a simple manufacturing technology, (5) should not be polluting and, finally, (6) have good availability and low cost, both with respect to raw material and with the process of obtaining [3]. Among the main natural materials for the production of bioplastics are proteins, such as gelatin and soy protein, cellulose derivatives, alginate, pectin, starch and other polysaccharides. The water solubility of films based on polysaccharides is advantageous in situations where the film is consumed with the product, resulting in minor changes in the sensory properties of food [4]. Edible films based on proteins, polysaccharides or lipids, in addition to increasing food quality, minimize special care with the final package [5].

Bioplastics can be simple (made with a simple macromolecule), compounds (two or more macromolecules), and be compounded with two or more layers. The compounds have the advantage of bringing together the strengths of each component, since hydrophobic films have good barrier to water vapor and hydrophilic films have good barrier to gases, in addition to providing effective mechanical properties [6]. As commented by Sorrentino et al., the application of nanotechnology to biodegradable polymers can open new possibilities to improve not only the properties of these materials, but at the same time improve the cost and the efficiency [7]. The cultivation of raw materials and the industrial production of nanocomposites based on natural polymers may contribute for employment

generation, thereby promoting social inclusion, especially considering the vast potential of productive family farms.

## 5.2 Biodegradable Films and Coatings

As defined by the term “biodegradable material” [8] is used to describe those materials that can be degraded by the enzymatic action of living organisms such as bacteria, yeasts and fungi, presenting as end products of the degradation process: CO<sub>2</sub>, H<sub>2</sub>O and biomass under aerobic conditions, and hydrocarbons, methane and biomass under anaerobic conditions. Biopolymers have been proposed to formulate edible films. Polysaccharides, proteins and lipids has been used in several ways: simple or composite materials or films or multiple layers [9]. Bioplastics, or organic plastics, are derived from renewable biomass sources, such as vegetable oil, corn starch, pea starch, or microbiota, rather than fossil-fuel plastics which are derived from petroleum. Some, but not all, bioplastics are designed to be biodegradable. They can be molded by the action of heat and pressure, like the conventional plastics, and are potential alternatives to conventional thermoplastic of petrochemical origin, such as polyolefins and polyesters [10], in strategic applications such as packaging area that generates much waste. Industries that produce bioplastics generally use microbes, or their enzymes, to convert biomass to feedstocks, building blocks for biodegradable plastics. The water solubility of films from polysaccharides is advantageous in situations where the film is consumed with the product, resulting in minor changes in the sensory properties of food [4]. Edible films based on proteins, polysaccharides or lipids, in addition to increasing food quality, minimize special care with the final package [11].

Edible films differ from edible coatings since they are formed before their application to the product, while the cover is formed during the application on it. Edible coatings can be defined as a layer of edible material formed around the food or placed between the components thereof [12].

For the development of edible and biodegradable bioplastics, it is required solvents and a pH regulating agent, when necessary, in addition to the plasticizer and polymer. The pH adjustment in the case of proteins is necessary to control the solubility of the polymer. Some regulators of pH found in the literature [13]: acetic acid and sodium hydroxide. The solvents commonly used to prepare these bioplastics are: water, ethanol or a combination of both [14]. A crucial aspect in the preparation of films is the solubility of proteins and the ability to interact with the same solvent used, since the total solubility of the protein is required for films’ formation [15]. The dispersion of the protein molecule in water is possible due to the large number of amino acid residues that interact with the polar solvents. These interactions can be improved depending on the dielectric constant of the solvent, since this constant is inversely proportional to the strength of intermolecular attraction. Films can be simple, made with one type of macromolecule or composed by two or more types of macromolecules, and can be formed with two or

more layers. The composed films have the advantage of bringing together the strengths of each component, as in the case of hydrophobic films that have good water vapor barrier, and hydrophilic films which have good barrier to gases, propitiating effective mechanical properties [6].

## 5.3 Soy Protein Isolate

### 5.3.1 Structure of Soy Protein

The soybean (U.S.) or soya bean (UK) (*Glycine max*) is a species of legume native of East Asia [16]. Soybean consists of 36–42 % protein and is also rich in oil (18–22 %). Due to the high protein amount, soy has a high demand for nitrogen. It is an annual plant that has been used in China for 5,000 years to primarily add nitrogen into the soil as part of crop rotation. Similar to other plants, soy protein has the primary function of providing amino acids for germination and protein synthesis. It is estimated that the polypeptide chain of soybean has a molecular weight between  $300\text{--}600 \times 10^3$  Da and a complex structure with bonds of type SS and hydrogen [17].

Soybean production in the United States, Argentina and Brazil accounted for 82 % of world production. According to the report of February 2009 the Department of Agriculture (USDA) soybean production worldwide was 224.15 million tons, while in Brazil it was around 57 and in Argentina 43.8 [18]. For the 2010/2011 harvest, second the United States Department of Agriculture, the world production of soybeans was 2,637 million tons.

### 5.3.2 Amino Acids Composition, Structures and Thermal Behaviour of Soy Proteins

The largest portion of the soy proteins is present as globulins and its incorporation in amino acids can be seen in Table 5.1.

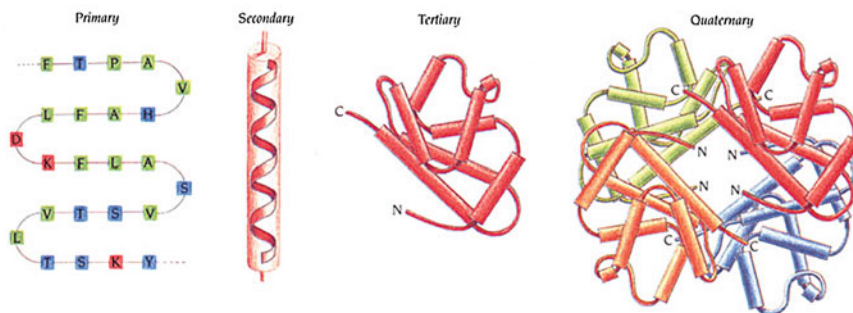
The soy protein isolate (SPI) comprises a set of macromolecules with different structures, formed by 18 different amino acids and with a molecular weight that may reach 600.000 gmol. When a solution containing SPI is subjected to ultra-filtration, it generally reveals about fifteen distinct fractions and four dominant fractions identified as 2S (20–22 %) 7S (37 %), 11S (31–40 %) and 15S (10–11 %), according to Schmidt et al. [19, 20]. To these four dominant fractions are attributed the characteristics of SPI. Like other proteins, can be organized into four different types of structures, i.e., the primary structure, secondary, tertiary and quaternary what will depend on their amino acids sequence.

The primary structure of proteins consists of a sequence of amino acids, linked together by peptide bonds (Fig. 5.1a). In the case of soy, the sequence consists of the amino acids of Table 5.1. All the protein structures, like other proteins, are

**Table 5.1** Amino acids composition of SPI

Amino acids	Composition (g, 16 g N)
Isoleucine	4,54
Leucine	7,78
Lysine	6,38
Methionine	1,26
Cysteine	1,33
Phenylalanine	4,94
Tyrosine	3,14
Threonine	3,86
Tryptophan	1,28
Valine	4,80
Arginine	7,23
Histidine	2,53
Alanine	4,26
Aspartic acid	11,70
Glutamic acid	18,70
Glycine	4,18
Proline	5,49
Serine	5,12

Source [19]



**Fig. 5.1** Amino acids forming peptide bonds (a) and protein (b, c, d) structures [22]

illustrated in Fig. 5.1b. The peptide bond is formed between the carbon atom (C) of the carboxylic group and nitrogen atom (N) of the amino group with elimination of water. The two ends of the chain thus formed are termed “amino terminal” and terminal carboxylic acid or N-terminus and C-terminus [21].

The secondary structure includes the spatial arrangement of peptides’ segments, stabilized by H bonding between carboxyl and amine groups of atoms. There are two types of arrangement, i.e.,  $\alpha$  helix (occurs between groups of the same chain segments, parallel to the chain) or  $\beta$  sheet (occurs between groups of different inter-chain segments, perpendicular to the chain). The tertiary structure describes the folding of the tertiary structural elements, including the lateral chains. The

quaternary structure appears only in oligomeric proteins, with a spatial distribution comprising more than one polypeptide chain in space, which are the subunits of the molecule mainly linked by covalent forces, like disulfide bonds, and noncovalent linkages, like hydrogen bonding and hydrophobic interactions [4]. The secondary, tertiary and quaternary structures of these macromolecules can be changed by the protein denaturation. Denaturing promotes increased interaction between chains of amino acids without the breaking of peptide bonds, thus improving the quality of films and gels formed.

Protein denaturation can be made through mechanical, physical (heat, radiation, ultra violet, ultrasonic, etc.) or chemical (guanidinium chloride, urea, lithium bromide, organic solvents, detergents, and extremes of pH) agents, affecting mainly the quaternary, tertiary and secondary structures. Some proteins are more stable at cooling temperature while some are more stable at ambient temperature. Usually between 40–50 °C the majority becomes unstable and there is the tendency of a change in the original conformation (denaturation) [23]. Denaturing presents several advantages such as: (1) Improves digestibility/bioavailability, i.e., improves enzymatic hydrolysis; (2) destruction of antinutritional factors; (3) Create desirable functional conditions for processing and (4) destroys toxins. Some of the disadvantages presented are: (a) destruction of amino acids, decreasing nutritional value and (b) complex formation (Maillard reaction) [24]. When the denaturation is done smoothly, it can be reversible. The studies linking soy protein denaturation, according to Kumar et al. [25], show that for this protein, structural modifications have been promoted by the use of bases, enzymes, surface-active agents (sodium dodecyl sulfate and sodium dodecylbenzenesulfonate). Such treatment promotes the exposure of polar functional groups, facilitating possible interactions with other molecules.

### ***5.3.3 Functional Uses of Soy Protein Isolate***

Soy protein is used for emulsification and texturizing. Specific applications include adhesives, asphalts, resins, cleaning materials, cosmetics, inks, pleather, paints, paper coatings, pesticides/fungicides, plastics, polyesters, and textile fibres [26]. Foods originating from plants have non-nutrient compounds (phytochemicals) with biological activities said health-promoting activities such as antioxidant, inflammatory and hypocholesterolemic like the soy isoflavones [27]. Soybean and its derivatives provide varying levels of isoflavones (daidzein, genistein and glycitein), bioactive compounds with diverse biological activities apparently associated with their forms [28].

The isoflavones exist in four chemical forms with a total of 12 isomers: the aglycones daidzein, genistein and glycitein; the  $\beta$ -glycosides daidzin, genistin and glycitin, and derivatives 6-O-acetylated such as 6-O-acetyldaidzin, 6-O-acetylgenistin, 6-O-acetylglycitin; and glucosyl-O-malonyl derivatives, as 6-O-malonyldaidzin, 6-O-malonylgenistin and 6-O-malonylglycitin [29]. Soy protein derivatives,

such as flours, isolates, concentrates and textured protein are widely used in food industry due to its functional properties. These products contain appreciable amounts of isoflavones, which during the processing stage of soybeans may be lost or modified. The main isoflavones present in soy non-processed, i.e., malonylgenistin, genistin, daidzein and malonyldaidzin are transformed into other forms during processing, such as acetylglicosides and aglycones [30]. Wang et al. [31] comment that the antioxidant activity of isoflavones has been linked to the number of hydroxyl groups present in their chemical structure. Pratt and Birac [32] reported that defatted flour, concentrate and isolated soy protein showed significant antioxidant activity by the method of co-oxidation of both  $\beta$ -caroteno and linoleic acid.

Besides the soybeans commercialization, the food industries also sell flour, concentrates, and soy isolates, and textured soy, fermented foods like miso, soy sauce and still the extract of soy or soy milk (soymilk) [29]. There are also other possibilities for the use of soy, like the elaboration of bioplastics, which have opened a new range of option for soy products, such as the use of Soy Protein Isolate (SPI) in the production of films and edible coatings. As published [25, 33], the separation of (SPI) from the other components of soybeans goes through a process of purification, based on chemical reactivity and solubility. A process for obtaining the SPI starts after the removal of bark and oil of soya beans, resulting in defatted flour. From this stage, and performed a typical process of aqueous extraction based in dissolution with pH lower than 5 or greater than 6.5, the SPI precipitation occurs. It is possible to study the SPI solubility as a function of the pH used in the protein extraction process [20]. The SPI has always been used as ingredient in food products due to their nutritional properties and physiological function, and have capacity for hydration, solubility, colloidal stability, gelation and also acts as emulsifier (technological functional properties) [34]. The IPS contains at least 90 % protein ( $N \times 6.25$ ) having as main components, the fractions  $\beta$ -conglycinin and glycinin, being free of fat and carbohydrate [35]. Moreover, lowering cholesterol in humans, due to consumption of soy protein isolates, is related to its isoflavone content [31].

### ***5.3.4 Mechanical Properties***

Films based on IPS, according to Gontard [9], the mechanical properties or barrier to water vapor unsatisfactory for practical applications, what became become worse under conditions of high humidity. Several studies have been made to improve the properties of the films based on IPS. It is reported that the addition of cross linking agents the film-forming solutions, or other physical methods, can improve the mechanical properties and/or the water vapor permeation (WVP) of films based on proteins [36]. Ou et al. [37] prepared films with 5.0 g of SPI, 3.0 g glycerol and different amounts (0, 50, 100, 150, 200 mg) of ferulic acid in two pH levels (8.0 and 9.0). At pH 9, the films showed better mechanical properties, as shown in Table 5.2.

**Table 5.2** Effect of ferulic acid on tensile strength (TS) and percentage of elongation at break (E) of SPI films plasticized with glycerol

Ferulic acid (mg/100 g)	Tensile strength (MPa)		Elongation (%)		Thickness ( $\mu\text{m}$ )	
	pH = 8,0		pH = 9,0		pH = 8,0	
	pH = 9,0		pH = 8,0		pH = 9,0	
0	1.472 $\pm$ 0.044 <sup>a</sup>	1.598 $\pm$ 0.056 <sup>a</sup>	61.7 $\pm$ 7.8 <sup>a</sup>	156.3 $\pm$ 7.1 <sup>a</sup>	70.3 $\pm$ 0.4 <sup>a</sup>	70.8 $\pm$ 1.6 <sup>a</sup>
50	1.688 $\pm$ 0.139 <sup>b</sup>	2.065 $\pm$ 0.138 <sup>b</sup>	94.7 $\pm$ 3.9 <sup>a</sup>	167.0 $\pm$ 3.5 <sup>b</sup>	72.4 $\pm$ 3.2 <sup>a</sup>	72.8 $\pm$ 1.5 <sup>b</sup>
100	1.622 $\pm$ 0.057 <sup>b</sup>	2.602 $\pm$ 0.073 <sup>d</sup>	85.1 $\pm$ 10.9 <sup>b</sup>	165.3 $\pm$ 7.2 <sup>b</sup>	71.0 $\pm$ 1.2 <sup>a</sup>	70.2 $\pm$ 1.0 <sup>a</sup>
150	1.638 $\pm$ 0.098 <sup>b</sup>	2.438 $\pm$ 0.059 <sup>c</sup>	93.0 $\pm$ 12.4 <sup>b</sup>	166.5 $\pm$ 5.3 <sup>b</sup>	71.4 $\pm$ 1.8 <sup>a</sup>	71.8 $\pm$ 1.3 <sup>a</sup>
200	1.476 $\pm$ 0.062 <sup>a</sup>	2.172 $\pm$ 0.175 <sup>b</sup>	86.4 $\pm$ 7.6 <sup>b</sup>	155.4 $\pm$ 8.5 <sup>a</sup>	71.4 $\pm$ 0.7 <sup>a</sup>	72.2 $\pm$ 1.3 <sup>a,b</sup>

<sup>a-h</sup> Values (mean  $\pm$  standard deviation,  $n = 5$ ), with different letters in the same column are significantly different at 5 % level

Source [37]



Physical properties of films consisting of isolated soy protein and gelatin has been studied [38]. Film forming solutions with 10 % protein and 0.1 % glycerol were prepared by mixing different proportions of IPS solutions and gelatin (10:0, 8:2, 6:4, 4:6, 2:8; 0:10). The results showed that the tensile strength of films prepared increased with the content of gelatin. The film made only with IPS was stiffer and less flexible. With the addition of gelatin, the film became more transparent, homogeneous and easier to handle. An important factor to be considered is related to the hydrophilicity/hydrophobicity of films prepared with proteins, what may interfere in the mechanical properties, beyond the barrier property. To control the balance desired on mechanical and barrier properties, one may control the hydrophilicity of the hydrocolloidal films and the formation of composite films, by preparing double-layer films or even a single homogeneous layer, formulated to combine the advantages of a hydrophobic lipid with the protein hydrocolloid, reducing the disadvantages that each one presented separately [39]. There are several publications reporting the development of composite films with the addition of beeswax and/or fatty acids such as palmitic, stearic and lauric acids in hydrocolloids. Rhim et al. [40] observed an increase in water solubility of films based on soy protein isolate, when lauric acid was added.

## 5.4 Protein Extrusion

It is well established that the extrusion of proteins is a potential for obtaining edible films on a large scale [41]. For these authors, the incorporation of polysaccharides and inclusion of nanoparticles in these films tend to improve their mechanical properties. Park et al. [42] studied bioplastics made from extruded gelatin plasticized with glycerol, sorbitol or a mixture of both. In this study, they discard sorbitol as a plasticizer because of low flow in the extrusion of the material.

Liu et al. [43] studied the incorporation of corn oil and olive oil in biofilms based on gelatin and sodium alginate, prepared by extrusion. The addition of oil caused an increase in the thickness of biofilms, also causing a decrease in tensile strain and an increase in the elongation. Both corn oil as olive oil reduced the water vapor permeability of biofilms, but no significant difference was observed regarding the use of different concentrations of the same. Extruded flexible films produced with thermoplastic potato starch and glycerol, were studied by Thunwall et al. [44]. The authors concluded that with appropriate control of process conditions, the amount of plasticizer and moisture of the mixture, it is possible to obtain extruded films of this material. The authors also reported that the preparation of biofilms from native starch, by this process, was significantly more difficult.

## 5.5 Nanocomposites

Nanocomposites are finding several applications as new researches prove their efficiency in the areas that they are used, mainly engineer. They are more popular among researchers from academia and industry, due to their excellent properties that are superior compared to the virgin polymers and conventional composites [45]. They can cover a wide area of applications such as automotive, medical, packaging and aerospace among others, since they have better thermal, mechanical and barrier properties that exceed those found in conventional composites [46]. Nanocomposites are a class of hybrid materials consisting of organic and inorganic components, where the inorganic phase in nanometer level is dispersed in a polymeric [47–49]. Montmorillonite (MMT), hectorite and saponite are the most used clays (phyllosilicates 2:1) in the preparation of polymer nanocomposites (PN). Details of the formula and chemical structure of these clays are presented in Table 5.3 and Fig. 5.2 [2].

### 5.5.1 Structure of Clays and Nanoclays

Clays have a layered structure consisting of 2 types of sheets, the silica tetrahedral and alumina octahedral sheets. The silica tetrahedral sheet consists of  $\text{SiO}_4$  groups linked together to form a hexagonal network of the repeating units of composition  $\text{Si}_4\text{O}_{10}$ . The alumina sheet consists of two planes of close packed oxygens or hydroxyls between which octahedrally coordinated aluminum atoms are imbedded in such a position that they are equidistant from six oxygens or hydroxyls. The two tetrahedral sheets sandwich the octahedral, sharing their apex oxygens with the latter. These 3 sheets form one clay layer. See Fig. 5.2.

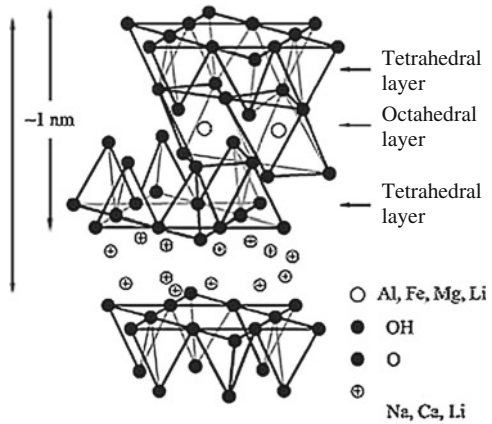
If the octahedral positions were occupied by alumina, we would not be looking at clay at all, but the inert mineral *pyrophyllite*. So, extremely important to the structure of clays is the phenomena of *isomorphous substitution*. Replacement of trivalent aluminum by divalent magnesium or iron II results in a negative crystal charge. The excess negative charge is compensated on the clays' surface by cations that are too large to be accommodated in the interior of the crystal. Further, in low pH environments, the edges of the clay crystal are positive, and compensated by anions. The result is a polyionic, supercharged nano-wafer that is unique in the world of minerals.

**Table 5.3** Chemical formula and the characteristic parameters of phyllosilicates 2:1

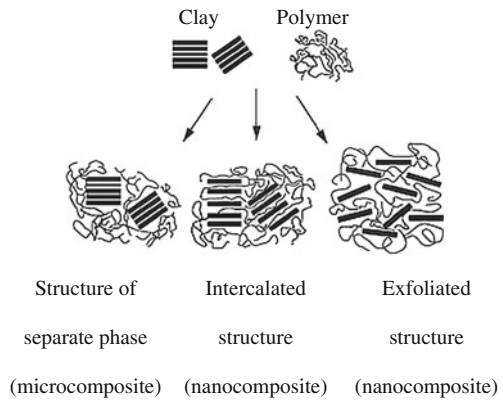
Phyllosilicates 2:1	Chemical formula <sup>a</sup>	CEC (meq/100 g)	Particle size (nm)
Montmorillonite	$\text{Mx}(\text{Al}_{4-x}\text{Mg}_x)\text{Si}_8\text{O}_{20}(\text{OH})_4$	110	100–150
Hectorite	$\text{Mx}(\text{Mg}_{6-x}\text{Li}_x)\text{Si}_8\text{O}_{20}(\text{OH})_4$	120	200–300
Saponite	$\text{MxMg}_6(\text{Si}_{8-x}\text{Al}_x)\text{Si}_8\text{O}_{20}(\text{OH})_4$	86,6	50–60

<sup>a</sup> M = monovalent cation; x = degree of isomorphous substitution (between 0,5 and 1,3)  
CEC = Cation exchange capacity

**Fig. 5.2** Illustration of clay structure [2, 45]



**Fig. 5.3** Illustrative structures of micro and nanocomposites [45]



Three main types of structures, which are shown in Fig. 5.3, can be obtained when a clay is dispersed in a polymer matrix: (1) phase-separated structure, where the polymer chains did not intercalate the clay layers, leading to a structure similar to those of a conventional composite, (2) intercalated structure, where the polymer chains are intercalated between clay layers, forming a well ordered multilayer structure, which has superior properties to those of a conventional composite, and (3) structure exfoliated, where the clay is completely and uniformly dispersed in a polymeric matrix, maximizing the interactions polymer–clay and leading to significant improvements in physical and mechanical properties [2, 50–52]. Production of nanocomposites based on polymer/clay can be done basically in three ways: (a) in situ polymerization, (b) prepared in solution and (c) preparation of the melt or melt blending [53].

The structures of nanocomposites have been characterized mainly by the techniques of X-Ray Diffraction (XRD) and transmission electron microscopy (TEM). The TEM analysis gives qualitative information of the sample as a whole, helping to understand the internal structure, partial distribution of various phases

and a vision of a structural defect by direct visualization, while the peaks in low angle XRD allow qualifying changes in the interlayer [45].

## 5.6 Conclusions

Based on text above and the references included in it, it is possible to note that there is great interest in other bioplastics based on natural sources of polysaccharides and proteins due to its low cost. However, the use of biofilms for food packaging has been greatly limited due to poor barrier properties and mechanical properties presented by natural polymers. Recent studies are exploring blends composed of soy protein isolate with other hydrocolloids and other biodegradable materials as an alternative to improve the properties of these materials. Compatibilizers have been incorporated into these blends to reach more uniformity in the material and influence directly its characteristics. Another possibility much studied is the preparation of nanocomposites from soy protein isolate. The fillers added to these materials have been clays, in general, which have shown good results. Finally, it is feasible to study biodegradable plastics based on soy protein isolate since it is possible to get this raw material in great amount and in a good price.

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