We are IntechOpen, the world's leading publisher of Open Access books Built by scientists, for scientists



185,000

200M



Our authors are among the

TOP 1% most cited scientists





WEB OF SCIENCE

Selection of our books indexed in the Book Citation Index in Web of Science™ Core Collection (BKCI)

# Interested in publishing with us? Contact book.department@intechopen.com

Numbers displayed above are based on latest data collected. For more information visit www.intechopen.com



## Chapter

# Development of Efficient Composites via Renewable, Recyclable, and Degradable Additives

Seyyed Alireza Hashemi, Seyyed Mojtaba Mousavi, Ali Mohammad Shakibafard, Ghasem Shouli, Yousef Amrollahi and Aziz Babapoor

## Abstract

Rapid development of composite industries raised the demand for tough, effective, practical, and sustainable composites with enhanced mechanical, electrical, thermal, and physical properties. However, several major problems such as high production cost and nonrecyclability and nondegradability of fillers and composites limited their selectivity and approaches toward their aims. Herein, our aim is to present and review the recent achievements in matter of reinforced composites with renewable, recyclable, or degradable additives toward development of composites through a mass-scale production strategy with least charges, high efficiency, and fine mechanical, thermal, and physical properties. Additionally, such aim can reduce the overall amount of nondegradable pollutions such as plastics in the nature and decline overall the charges of composite industries by reusing such materials and decreasing their demand for raw material. Last but not least, to clean up the nature from industrial wastes, green strategies should be developed to whether reuse such material or degrade them via practical strategies.

**Keywords:** renewable resources, degradable materials, recyclable fillers, composites, nanocomposites

## 1. Introduction

Polymeric-based composites have been frequently used in last decades due to their promising properties and selectable specification. It is widely known that the usage of a polymer and one (or more) type of solid fillers allows us to obtain several advantages. In this case, by using diverse kinds of fillers, we can develop composite structures with selectable mechanical, thermal, electrical, and chemical properties. Among the most common fillers in the composite industry, calcium carbonate, glass fibers, talc, kaolin, mica, wollastonite, silica, graphite, synthetic fillers (e.g., PETor PVA-based fibers), and high-performance fibers (carbon, aramid, etc.) can be mentioned [1]. However, the usage of such additives leads to one of the main limitations of polymer composites; the presence of two different components within the composite structure makes their recyclability and reuse a bit difficult to the level that it becomes much more economical to dispose them into the dump rather than reusing them [2–8]. Such method is often considered as an unappealing alternative due to its high cost, environmental impacts, and technical limitation/difficulties. Furthermore, it is worsened by the fact that plastic production requires a remarkable consumption of oil-based resources, which are notoriously nonrenewable [9]. Increased pressure from environmental activists, preservation of natural resources, and attended stringency of laws passed by developing countries lead to the invention and development of natural materials with a focus on renewable raw materials [10–12].

In case of chronological order, primary attempts were focused on the fabrication and characterization of polymeric composites based on recyclable polymers (e.g., polyolefin) filled with natural organic fillers, i.e., derived fibers and materials derived from natural plants. Several factors follow this choice. In this regard, the usage of natural organic fillers rather than traditional mineral inorganic additives leads to massive reduction in the usage of nonbiodegradable polymers and nonrenewable resources. Furthermore, these fillers are usually obtained from wastes or abundant plants; thus the final product becomes fairly economical and cheap. Moreover, these additives are less abrasive compared with inorganic mineral counterparts in matter of processing machinery. Likewise, these additives are also less dangerous for employees all of whom working in the production line due to their easy incineration and safe inhalation. The usage of such natural resources for reinforcement of composites can lead to the development of composite structures with lower density (or specific weight), ideal thermal properties, and fine acoustic insulation performance than with their mineral reinforced counterparts. Natural bio-fiber composites are emerging as a viable alternative to glass fiber reinforced plastics, especially in automotive applications. Thermoplastic biocomposites and biopolymer including PP, PE, PS, and PLA all of which were reinforced with natural fibers have attracted great attention toward themselves during the past few years. Such thermoplastic biocomposites, compared with those made using a thermoset matrix, can be processed into different shapes and have the potential to be mechanically recycled [13]. The synthesis of recyclable and degradable polymeric materials with a highly cross-linked three-dimensional network structure like PLA is very important for polymer science and technology for supporting a green sustainable society. PLA offers some advantages over the traditional nonbiodegradable polymers especially when their recycling is quite difficult or not economical. Likewise, there are also many restrictions due to the specification and properties of used materials, while a number of these challenges are expected to be resolved via blending PLA with other polymers, by making micro- and nanocomposites of PLA, coating with high barrier materials, and also through enhancement with practical polymeric materials [13].

The annual disposal of over 10 million tons of plastic wastes in both the EU countries and the USA has raised the demand for management and reuse of these nonbiodegradable waste streams. The synthetic polymeric materials containing displaced glasses, metals, wood, and ceramics have been widely used in many products, especially in the case of packaging. The commodity plastics, the so-called big four including PE, PP, PS, and PVC have been used in variety of forms such as films, rigid containers, and flexible bags which have revolutionized the packaging industry. However, disposal of these kinds of materials within the environment without the possibility of degradation led to rise of multitude environmental and ecological concerns. The important feature of composite materials is that they can be designed and tailored based on selectable properties to meet different requirements. Since biofibers are cheap and biodegradable, biocomposites from biofiber reinforced biodegradable polymers will render a contribution in the twenty-first century due to the serious environmental problem [14].

## 2. Green composites

When traditional nonbiodegradable polymeric matrices are reinforcing with natural fibers (e.g., green fiber and biofibers), the resulting composite structure is becoming partially biodegradable. If the polymeric matrix being selected forms biodegradable resins (i.e., green polymer or biopolymer) and reinforced with natural fibers, the final composite structure is considered as green or fully biodegradable composite. Two or more diverse natural fibers as additives in combination with polymeric matrices (biopolymer or petroleum-based polymer) lead to the development of hybrid green composites. The aim of hybrid composite development is to customize the properties and specifications of resulting green composites. Another subclassification of green composites can be based on the natural fillers and functional behavior of green composites. Depending upon the type of natural fillers, green composites can be classified as unidirectional and bi-directional continuous fiber green composites or discontinuous reinforcement composites (aligned or randomly oriented in the form of particulates, short fibers, and whiskers). According to the functional behavior, they can be classified as functionally graded and smart green composites. At the end of their life cycle, green composites can be easily disposed into the soil without deteriorating the environment by creating pollution sources. Many scientific evaluations have been developed to address the potential and specifications of green and partially biodegradable composites [15]. Table 1 reviews some of the reported works in case of these kinds of degradable composites.

Some processing techniques were widely used for fabrication of composites, while among them extrusion, subsequent injection, and compression molding can be mentioned. It was rarely observed that the overall amount of fillers exceed 50–60 wt%; however, in some cases higher percentages about 70–80 wt% were reported (especially in the USA). Besides, in more excessive situations, the compression molding method is preferred rather than injection molding method. Some common flaws related to the processing of such materials are due to the hygroscopic and hydrophilic nature of these additives along with their low thermal resistance which restrict the production procedure to temperatures lower than 200°C. In fact,

Fiber	Matrix	<b>Composition type</b>	Ref.
Rice husk, wheat, husk, and coconut coir	Epoxy resin (LY556) and hardener (HY951)	Partially biodegradable	[16]
Banana	Polylactic acid	Green	[17]
Banana	Epoxy resin (LY556) and hardener (HY951)	Partially biodegradable	[18, 19]
Jute	Polypropylene	Partially biodegradable	[20]
Jute	Polylactic acid	Green	[21, 22]
Sisal	Polylactic acid	Green	[23]
Sisal	Polypropylene	Partially biodegradable	[24]
Flax	Starch	Green	[25]
Flax	Polylactic acid	Green	[26, 27]
Kenaf	Polylactic acid	Green	[28, 29]

#### Table 1.

List of important partially biodegradable/green composites [15].

among the most highlighted problems of this field, we can refer to hygroscopic and hydrophilic nature of additives. Such features can significantly affect the distribution of fillers within the matrix and the interfacial adhesion between fillers and matrix. Additionally, in case of injection molding and during the processing of composites, the presence of humidity throughout materials leads to the formation of water vapor which can cause several systematic problems in the following molding steps where a venting or drying system is not present. In general, the formation of water vapor and also various kinds of gases during the processing leads to the formation of voids within the developed composite structure, thereby deteriorating their mechanical properties [9].

So far, the biodegradability of Mater-Bi-based green composites has not been perfectly investigated. However, available scientific research regarding the biodegradability of this type of composite revealed that they become biodegradable after their burial within the soil [30–32]. Rutkowska et al. [33] studied the biodegradation of bi-based green composites in different natural environments and observed a complete biodegradation after 4 weeks, and conducted these investigations only in case of neat materials. While these investigations were conducted only in case of neat materials. In another study, Scaffaro et al. [34] have studied Mater-Bi/wood flour composite biodegradation in active sewage sludge, while their obtained results showed that the developed composites undergo biodegradation with higher weight loss rates than the neat Mater-Bi. This effect was attributed primarily to the obtained morphology and capability of wood fibers to act as support for the bacterial growth [34]. Huda et al. [35] investigated the properties of PLA/recycled cellulose composites prepared by extrusion and injection molding. Their outcome showed that an increase in the filler loading up to the 30wt% significantly improved the rigidity without affecting the crystallinity degree or thermal stability of the final product. In another research, Plackett et al. [36] prepared PLA/jute composites through a film stacking technique. Their obtained data showed that the addition of jute can lead to considerable enhancement in the tensile properties, while it can also increase the brittle fracture and deteriorate the impact strength of the final product [9].

## 3. Bio-based and biodegradable polymeric composites

When a biodegradable material (neat polymer, blended product, or composite) is obtained completely from renewable resources, we usually call it a green polymeric material. Biopolymers from renewable resources have attracted much attention in recent years due to their advantages to nature [37]. Renewable resources of polymeric materials offer a solution to maintain sustainable development of economically and ecologically attractive technologies. The innovations in the development of materials from biopolymers, preservation of fossil-based raw materials, complete biological degradability of polymeric materials, reduction in the volume of garbage and composability in the natural cycle, protection of the climate through the reduction of carbon dioxide released, as well as the applications of agricultural resources for the production of bio/green materials are among the reasons that attracted the public attention toward such materials [38–40].

Bio-based and biodegradable polymeric materials have a wide range of applications in diverse fields such as packaging, biomedical, and agricultural fields. The most common application of biodegradable materials is in case of blends for TPS and aliphatic/aromatic polyesters, such as PLA or polylactides, PCL, PBAT, and PHB [41]. Recently, PLA has attracted considerable attention toward itself due to its biodegradability in the composting situation. PLA has turned into an economically plastic material for industries which is commonly used for production of films and

packaging materials, e.g., trays, bottles, and thin films for food packaging industry [42–44]. This cost affordable polymeric material can meet the requirement of industries for composite materials with ideal mechanical properties along with easy processability. However, PLA has some environmental impacts according to the life cycle assessment (LCA) [45]. PLA is a thermoplastic aliphatic polyester obtained from the ring-opening polymerization of lactide, which may be derived from the fermentation of sugar feedstock at competitive prices [14]. The same as most of thermoplastics, PLA can be molded into bottles, containers, and so on by injection molding, blow molding, etc. or be extruded into fibers, films, and sheets [13].

Polyester-derived by-products are playing an important role in case of biodegradable plastics due to their highly potential hydrolysable ester bonds. Polyesterderived materials are consisted from two main chemical groups, i.e., aliphatic (liner) and aromatic (aromatic rings) polyesters. Recently, various kinds of commercially biodegradable polyester materials were developed; among which we can refer to PHA, PHH, PHB, PHV, PLA, PCL, PBS, PBSA, AAC, PET, PBAT, and PTMAT [46].

The major constituents of biofibers (lignocelluloses) are cellulose, hemicellulose, and lignin. The overall amount of cellulose in lignocellulosic systems can vary based on the type and age of the plants or species that were used. Cellulose is a hydrophilic glucan polymer, while lignin is a phenolic compound, generally resistant to microbial degradation, but the pretreatment of fiber renders it susceptible to the cellulose enzyme [47, 48]. The exact chemical nature of the principal component of biofiber (the lignin) still remains obscure [14, 38]. The chemical compositions and structural parameters of some important biofibers are represented in **Table 2**. As shown in **Table 2**, specifications of natural fiber significantly vary from each other. Such variation may be due to the origin, age, retting (mode of extraction of fiber from the source) process adopted, etc. [14].

Type of fiber	Cellulose (wt%)	Lignin (wt%)	Hemicellulose (wt%)	Pectin (wt%)	Wax (wt%)	Microfibrillar/ spiral angle (deg.)	Moisture content (wt%)	Ref
Bast	61–71.5	12–13	13.6–20.4	0.2	0.5	8.0	12.6	[49, 50]
Jute	71	2.2	18.6–20.6	2.3	1.7	10.0	10.0	[51, 52]
Flax	70.2–74.4	3.7–5.7	17.9–22.4	1.9	0.8	6.2	10.8	[49]
Hemp	68.6–76.2	0.6–0.7	13.1–16.7	1.9	0.3	7.5	8.0	[53]
Ramie Kenaf	31–39	15–19	21.5				70	[54]
Leaf	67–78	8–11	10–14.2	10	2	20	11	[55]
Sisal	67–78	8–11	10–14.2	10	2	20	11	[55]
PALF	70–82	5–12	_	_	_	14	11	[56]
Henequen	77.6	13.1	4–8	_	_	_	8	[56]
Seed	82.7	—	5.7	_	6	—	_	[57, 58]
Cotton	82.7	_	5.7	_	6	_	_	[57, 58]
Fruit coir	36–43	41–45	0.15–0.25	3–4		41–45	8	[57, 58]

## Table 2. Chemical composition and structural parameters of some natural fibers [14].

## 4. PLA bioplastics

PLA is one of the most studied bioplastics due to its recyclability and attractive specifications. PLA can be biodegraded under certain conditions, such as the presence of oxygen and moisture [59]. Moreover, PLA is a biodegradable material, and its usage within the biodegradable polymers has a high level of importance, because it significantly reduces the negative environmental impacts of wastes due to its biodegradability, improves the material recyclability, and eases monitoring external effects on the properties of the final composition. In this matter, the first reason is that the generation of wastes from industrial processes where the grinding process of the offcuts and blending with the neat polymeric matrix is integrated is unavoidable [60]. The second reason is the possibility of PLA wastes to be reused and recycled, because it is necessary to extend the lifetime service of PLA prior to discarding them into the biodisposal sites (i.e., composting sites) [61, 62]. Finally, the negligible environmental impacts of PLA could be ignored if it is not being possible for industries to recycle the obtained wastes [60].

In a work by Ohya et al. [63], they reported a procedure to develop graft polymerization of PLA on polysaccharides via TMS-protected polysaccharides. Based on their study, introduction of TMS protecting groups to chlorotrimethylsilane/ pyridine can make the low molecular weight polysaccharides soluble in organic solvents and lead to an approach which can control the overall amount of initiating groups (i.e., the overall amount of graft chains). Moreover, via usage of trimethylsilyl protection method, PLA-grafted polysaccharides with variable lengths and number of graft chains were developed. The graft copolymer films present lower glass transition temperature ( $T_g$ ), crystallinity, melting temperature, and higher viscosity than pure PLA film. What is more, the usefulness of graft copolymer as a plasticizer was investigated with 1:4 blend films prepared from the graft copolymers and PLA. The blend films showed lower  $T_g$  and crystallinity and higher viscosity than pure PLA film [64].

In another research, multiple extrusions of PLA up to ten times were investigated by Enkiewicz et al. [61] using a double screw extruder for granulation of PLA followed by laboratory injection molding press, for preparation of specimens. In this case, their obtained results showed that the tensile strength values of PLA did not significantly depend on the number of extrusion cycles, and they showed only a little diversity from each other, showing a slight total reduction of about 5.2% after ten extrusion processes (**Figure 1(a)**). Besides, a negligible decrease in the tensile strain was observed (about 2.2–2.4%) which did not depend on the number of extrusion process. On the other hand, by an increase in the number of preformed

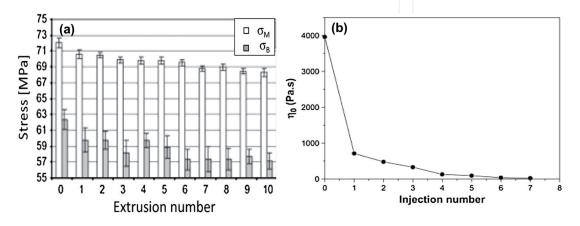


Figure 1.

(a) Tensile strength ( $\sigma_M$ ) and tensile stress at break ( $\sigma_B$ ) as functions of the extrusion number [61] and (b) zero viscosity of PLA as a function of injection number [60].

extrusions, the impact strength clearly declined (20.2% after ten extrusion processes), while the melt flow rate (MFR), water vapor, and oxygen transmission rates were significantly increased. The MFR value of the specimen subjected to ten extrusion processes was threefold higher than the original specimen. The thermal characteristics of specimens showed a slight decrease in the thermal stability of PLA, lower cold crystallization temperature, and a slight reduction in the melting point by increasing the number of cycles without any effect on the glass transition temperature ( $T_g$ ) [61, 64].

PLA degradation has been found to be dependent on several factors, including molecular weight, purity, crystallinity, temperature, pH, the presence of terminal carboxyl or hydroxyl groups, water permeability, and additives all of which acting catalytically that may include enzymes, bacteria, or inorganic fillers [65]. Pyrolysis is a method which can lead to the treatment of polylactide, in which applied heat to a condensed substrate can lead to its chemical decomposition. Co-pyrolytic methods attracted considerable attention toward themselves due to their presented alternatives for disposal and convert wastes original form into valuable fuel sources. Likewise, the specific benefit of this procedure could be attributed to its potential for reduction of wastes within the nature, recovery of used chemical compounds, and replacing the developed fuel with frequently used fossil fuels. Furthermore, co-pyrolysis treatment of PLA-derived materials and biomass offers a potential alternative way for removal of wastes from the nature and could act as an upgrading approach during the pyrolysis of biomass and wastes [66]. PLA can be recycled to its primary form (i.e., monomer) through hydrolyze by boiling water or steam of lactic acid. This approach could lead to molecular recycling and provide an alternative for recycling both fabricated plant materials and post-consumed packages. In this regard, PLA could be hydrolyzed to its primary form by applying heat at 180–350°C for 30min, which can lead to recycle of L-lactic acid as the final outcome [66].

In a research by De Jong et al. [67], they depolymerized PLA through the release of dimers in alkaline conditions, in which the end chain degradation could be explained via intramolecular transesterifications. In this matter, an electrophilic attack which is catalyzed via the base of the hydroxyl (OH<sup>-</sup>) end group on the second carbonyl group can lead to the formation of a ring and shorten the polymeric chain by hydrolysis of the final lactide. In the next step, free lactide hydrolysis into two lactic acid molecules. During this step, a random alkaline attacks to the carbon atoms of the ester groups, followed by the hydrolysis of the ester bond which leads to the intramolecular degradation of the primary chemical compound. Thereby, new low molecular weight molecules are produced; however, in the acidic conditions, the protonation process of the hydroxyl end group leads to an intramolecular hydrogen bond. The hydrolysis of the ester group can decrease the degree of polymerization of PLA due to the release of lactic acid molecules. Besides, random intramolecular protonation of ester groups' carbon atoms condenses the hydrolysis of ester bonds and produces diverse kinds of low molecular weight fragments [46]. Another study [60] on the reprocessing of PLA (containing 92% L-lactide and 8% D-lactide) showed that only the tensile modulus remains constant with thermomechanical cycles of up to seven injection moldings. Moreover, stress and strain at break, modulus, hardness, and rheological factors generally showed a small decline. Additionally, the viscosity of PLA decreased greatly (from 3960 to 713 Pa.s) after only one injection cycle. Zero viscosity  $(g_0)$  of PLA as a function of injection number is presented in **Figure 1(b)**. The mechanical properties of the recycled PLA had become too poor for an industrial application of the polymer [14]. In **Table 3**, a list of starchy and cellulosic materials that were used for the production of lactic acid can be seen.

Substrate	Microorganism	Lactic acid yield (g/	
Wheat and rice bran	Lactobacillus sp.	129	
Corn cob	Rhizopus sp. MK-96–1196	90	
Pretreated wood	Lactobacillus delbrueckii	48–62	
Cellulose	Lactobacillus coryniformis ssp. torquens	0.89	
Barley	Lactobacillus casei NRRLB-441	0.87–0.98	
Cassava bagasse	L. delbrueckii NCIM 2025 and L. casei	0.9–0.98	
Wheat starch	Lactococcus lactis ssp. lactis ATCC 19435	0.77–1	
Whole wheat	Lactococcus lactis and Lactobacillus delbrueckii	0.93–0.95	
Potato starch	Rhizopus oryzae and R. arrhizus	0.87–0.97	
Corn, rice, and wheat starches	Lactobacillus amylovorus ATCC 33620	<0.70	
Corn starch	L. amylovorus NRRL B-4542	0.935	

Table 3.

Starchy and cellulosic materials used for the production of lactic acid [46].

## 5. Nano-biocomposites

Nano-biocomposites are among the materials that contain bio-based polymers and low additions of nanoparticles of natural fibers such as cellulose and lignin for reinforcement purposes [68–70]. They are shaded orange in the bioplastics Spectrum because nanoparticles pose unknown hazards, and fibers are obtained via the craft method and can be treated with chemicals (isocyanates, alkalis) to enhance their properties as reinforcements [70]. Hazards of the craft method and these chemicals were described above. The health effects of nanoparticles are a major concern due to the lack of knowledge about their stability during processing, and there are potential toxicity concerns related to decomposition and/or migration during service [71]. Toxicologists hypothesize that nanoparticles may not be detected by the normal defense system of organisms; their small size can modify protein structures, and they can travel from respiratory system to the brain and other organs [72–74].

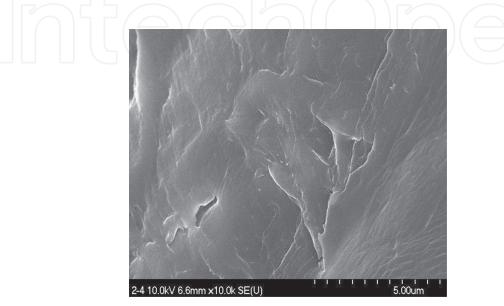
In a work by Hubbe et al. [75], they investigated about cellulose nanocrystals for their possible applications within the industry and stated that retention of developed nanocrystal properties should be controlled and guaranteed by using water miscible polymer matrices (e.g., starch products, latex, polyvinyl alcohol) to ease production procedure of cellulose nanocrystals and make them much more compatible with matrix. In another study, Eichhorn et al. [76] reported possible procedures of cellulose nanofiller recovery and then focused on the usage of cellulose nanowhiskers for the manufacturing of shape memory nanocomposites, as well as on the interfacial phenomena occurring in polymer/nanocellulose filler composites [9].

In another study by Mousavi et al. [77], NBR was composed with the natural polymers such as starch and glycerol. They also used silica nanoparticles to enhance the physical and mechanical properties of NBR. It was revealed that by the increase in the overall amount of starch, the mechanical properties of developed composite have considerably declined, while by an increase in the amount of starch, the module has increased, and impact resistance and also elongation at break have decreased. However, by the addition of silica nanoparticles, physical and mechanical properties of final composition were slightly improved, and the highest mechanical properties were achieved when silica nanoparticle filler

loading increased to 3 wt%. Besides, the low weight percentage of starch can convert polymers into the biodegradable polymer which has a variety of applications in the automotive industry. In this matter, SEM examinations of specimens justified the biodegradability of reinforced composites by adding starch and silica nanoparticles to their system. SEM image of developed specimen can be seen in **Figure 2**.

Also recently, Mousavi et al. [3] developed polypropylene-wood composite as a new source of raw material with a combination of maleic anhydride and eggshell nanoparticles in order to improve their overall performance. They indicated that by creating cross-link between these additives, the tensile properties of polypropylene improve at high temperatures. The obtained results also showed that the tensile strength and fracture strain of samples containing cross-linked fillers and matrix are higher than samples without it at high temperatures. They also revealed that an optimum amount of peroxide is needed to achieve the desired tensile properties. Furthermore, their results showed that an increase in the overall amount of natural polymers and additives such as starch, corn, and eggshell nanoparticles as fillers can significantly enhance the physical and mechanical properties of developed composites. Their obtained results can be seen in **Tables 4–6** and **Figure 3**.

In a study by Farazi et al. [78], they prepared an antimicrobial substrate using LDPE/EVA/PE-MA/clay nanoparticle blend along with PS and GO. In this regard, LDPE was selected due to its fine elasticity, transparency, low melting point, and simple reversible process. Besides, developed matrix was well-reinforced with various kinds of natural, biodegradable, and antimicrobial additives. In this case, a twin screw extruder was used to produce the related specimens. Furthermore, specifications of developed samples were examined using mechanical, SEM, TEM, XRD, water absorption, water vapor, permeability, oxygen permeability, and microbial permeability tests. Moreover, the outcome of their study showed that developed specimens have appropriate mechanical properties along with their antimicrobial performance. Additionally, the outcome of water absorption, water vapor, and oxygen permeability tests showed that the sample containing GO/clay nanoparticles is presenting the best results. In this matter, the outcome of mechanical tests can be seen in **Figure 4**(a-c), while the cultivating conditions of microorganisms and antimicrobial performance of developed samples can be seen to be negative on antimicrobial films (Table 7).



**Figure 2.** SEM image of a specimen containing 5 wt% starch and 18.7 wt% NBR [77].

#### Renewable and Sustainable Composites

Sample number	Propylene (wt%)	Corn (wt%)	Starch (wt%)	Eggshell nanoparticles (wt%)	Compatibilizer (wt%)	Foaming agent (wt%)
1	100	0	0	0	0	0
2	50	46	0	0	4	0
3	50	44	0	0	4	2
4	50	33	10	1	4	2
5	50	21	20	3	4	2
6	50	9	30	5	4	2

Sample number	Bending module (MPa)	Bending strength (MPa)	Tensile modulus (MPa)	Tensile strength (MPa)
1	522.57	7.249	6.08	17.42
2	1185.63	15.711	7.09	20.45
3	1224.27	15.908	6.68	19.8
4	1190.47	15.293	7.1	19.1
5	1025.55	13.713	1.66	18.49
6	925.1	12.478	12.41	19.59

#### Table 5.

Physical and mechanical properties of polypropylene-based composite [3].

Sample number	1	2	3	4	5	6
Impact strength (J/m)	17.55	30.86	14.88	51.43	222.9	55.28

The test conditions were as follow: Standard ISO 180, humidity about 45.1%, temperature about 22.2°C, touch back width of 8 mm, weights of 1, and touch radius of 0.25 mm [3].

#### Table 6.

Impact test of polypropylene composites based on ASTM 256 standard.

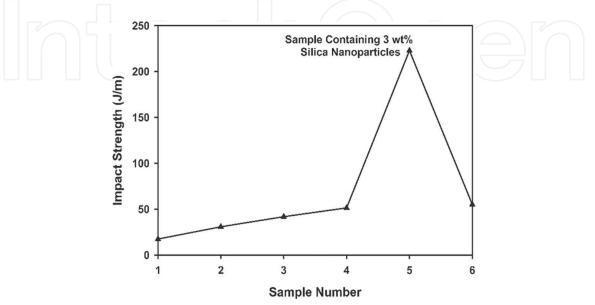
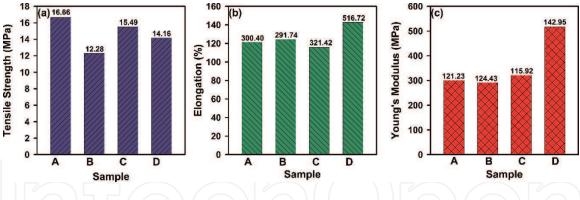


Figure 3.

Impact test results of polypropylene-based composites containing glycerol and starch. Sample 5 has the highest impact strength which is containing 3 wt% silica nanoparticles [3].



#### Figure 4.

(a) Tensile strength, (b) elongation at break, and (c) Young's modulus of developed samples by Farazi et al. Sample A (LDPE-EVA-PE-MA), sample B (LDPE-EVA-PE-MA-PS), sample C (LDPE-EVA-PE-MA-GO), and sample D (LDPE-EVA-PE-MA-PS-GO) [78].

Type of microorganism	ISIRI standard	Cultivating conditions 20–25°C/3–5 days	
Plural	10,899-1ISIRI		
Yeast	10,899-2ISIRI	20–25°C/3–5 day	
E. coli	2946ISIRI/gram negative	37°C/17 h	
Staphylococcus aureus	6806ISIR/gram negative	37°C/24 h	
Salmonella	1810ISIRI/gram negative	37°C/24 h	
Coliform	9263ISIRI/gram negative	37°C/24 h	
Enterococcus	2198ISIRI/gram positive	35°C/24 h	
Lactic acid bacteria (LAB)	13559ISIRI	30°C/72 h	
Esporas Clostridium Sulfito reductor	9432ISIRI	37°C/48 h	
Bacillus cereus	2324ISIRI/gram positive	30°C/18 h	

#### Table 7.

Microorganism, medium, and cultivating conditions [78].

### 6. Conclusions

Green composites have attracted great attention toward themselves due to the ecological issues and decline of petroleum-based resources because of their hazards. Different types of natural fibers and their properties have been studied as a potential replacement for synthetic fibers. In recent years, various kinds of polymer composites were reinforced with organic fillers (rather than mineral inorganic additives) to reduce the usage of whether petroleum-based or mineral products and replace them with renewable resources. These "green" composites have numerous industrial applications. One of the main advantages of these materials is their low cost and degradability which protect the environment against nondegradable products. On the other hand, these composites are suffering from some disadvantages including ductility, processability, and dimensional stability. To resolve the aforementioned problems, researcher from all around the globe conducted great efforts to provide practical and appropriate alternatives through chemical treatment of additives and the usage of adhesion promoters to improve the interaction between matrix and fillers. Additionally, to obtain a completely biodegradable composite structure, it is bare bone essential to reinforce polymeric matrices with biodegradable additives rather than obtained additives form mineral or nonrenewable

resources. In this regard, recent efforts were focused on the selection of biodegradable additives and matrices along with the optimization of their production procedures and processing parameters. Last but not least, the usage and development of sustainable and renewable resources not only can save our planet but also reduce production cost and pollutions within the environment.

## Abbreviations

PET	polyethylene terephthalate
PVA	polyvinyl alcohol
PP	polypropylene
PE	polyethylene
PS	polystyrene
PLA	polylactic acid
PVC	polyvinyl chloride
TPS	thermoplastic starch
PCL	polycaprolactone
PBAT	polybutylene adipate terephthalate
PHB	polyhydroxybutyrate
PHA	polyhydroxyalkanoates
РНН	polyhydroxyhexanoate
PHV	polyhydroxyvalerate
EVA	ethylene-vinyl acetate
PBS	polybutylene succinate
GO	garlic oil
PALF	pineapple leaf fiber
TEM	transmission electron microscopy
SEM	scanning electron microscopy
XRD	X-ray diffraction
PBSA	polybutylene succinate adipate
LCA	life cycle assessment
AAC	aliphatic-aromatic copolyesters
PBAT	polybutylene adipate terephthalate
PTMAT	polymethylene adipate/terephthalate
TMS	trimethylsilyl
MFR	melt flow rate
NBR	nitrile-butadiene rubber
LDPE	low-density polyethylene
MA	maleic anhydride
PS	potassium sorbate

# IntechOpen

## **Author details**

Seyyed Alireza Hashemi<sup>1,2\*</sup>, Seyyed Mojtaba Mousavi<sup>1,2</sup>, Ali Mohammad Shakibafard<sup>1,2,3</sup>, Ghasem Shouli<sup>3</sup>, Yousef Amrollahi<sup>4</sup> and Aziz Babapoor<sup>5</sup>

1 Department of Medical Nanotechnology, School of Advanced Medical Sciences and Technologies, Shiraz University of Medical Sciences, Shiraz, Iran

2 Pharmaceutical Sciences Research Center, Shiraz University of Medical Sciences, Shiraz, Iran

3 Zagros Petrochemical co. Asalouyeh, Buahehr, Iran

4 Borj va Baru Fars Research Center, Fars Science and Technology Park, Shiraz, Iran

5 Department of Chemical Engineering, University of Mohaghegh Ardabili, Ardabil, Iran

\*Address all correspondence to: s.a.hashemi0@gmail.com

## **IntechOpen**

© 2019 The Author(s). Licensee IntechOpen. This chapter is distributed under the terms of the Creative Commons Attribution License (http://creativecommons.org/licenses/by/3.0), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

## References

[1] Mousavi SM et al. Silica nanoparticles/phenol Novolac epoxy resin Nanocomposites via vacuum shock technique: Effect of silica nanoparticles on curing kinetic. Journal of Chemical, Biological and Physical Sciences. 2018;**8**:361-376

[2] Mousavi SM, et al. Investigation thermal stability and Activation energy composite of Epoxy phenol novelac Novalac/Unsaturated poly Ester/silica nano particle. 2015

[3] Mousavi SM et al. Modifying the properties of polypropylene-wood composite by natural polymers and eggshell Nano-particles. Polymers from Renewable Resources. 2015;**6**:157-174

[4] Hashemi SA et al. Electrified single-walled carbon nanotube/ epoxy nanocomposite via vacuum shock technique: Effect of alignment on electrical conductivity and electromagnetic interference shielding. Polymer Composites. 2018;**39**(S2):E1139-E1148

[5] Hashemi SA, Mousavi SM. Effect of bubble based degradation on the physical properties of single wall carbon nanotube/epoxy resin composite and new approach in bubbles reduction. Composites Part A: Applied Science and Manufacturing. 2016;**90**:457-469

[6] Mousavi S et al. Improved morphology and properties of Nanocomposites, linear low density polyethylene, ethylene-co-vinyl acetate and Nano clay particles by Electron beam. Polymers from Renewable Resources. 2016;7(4):135

[7] Hashemi SA et al. Lead oxidedecorated graphene oxide/epoxy composite towards X-ray radiation shielding. Radiation Physics and Chemistry. 2018;**146**:77-85 [8] Mousavi SM et al. Enhancement of rheological and mechanical properties of bitumen by Polythiophene doped with Nano Fe<sub>3</sub>O<sub>4</sub>. JOM. 2018;7(2):1-10

[9] La Mantia F, Morreale M. Green composites: A brief review. Composites Part A: Applied Science and Manufacturing. 2011;**42**(6):579-588

[10] Anandjiwala RD, Blouw S. Composites from bast fibres-prospects and potential in the changing market environment. Journal of Natural Fibers. 2007;4(2):91-109

[11] Wittig W. Einsatz von Naturfasern in Kfz-Bauteilen. Kunstoffe im Automobilbau. 1994:1-7

[12] Mousavi SM et al. Green synthesis of silver nanoparticles toward bio and medical applications: Review study. Artificial Cells, Nanomedicine, and Biotechnology. 2018;**46**:1-18

[13] Soroudi A, Jakubowicz I.
Recycling of bioplastics, their blends and biocomposites: A review. European Polymer Journal.
2013;49(10):2839-2858

[14] Mohanty AK, Misra Ma, Hinrichsen G. Biofibres, biodegradable polymers and biocomposites: An overview. Macromolecular Materials and Engineering. 2000;**276**(1):1-24

[15] Singh JIP, Dhawan V, Singh S. Development and characterization of green composites: A review. Asia Pacific Journals. 2015;**2**(1)

[16] Dhawan V, Singh S, SinghI. Effect of natural fillers on mechanical properties of GFRP composites. Journal of Composites.2013;2013:792620

[17] Sajna V, Mohanty S, Nayak SK. Hybrid green nanocomposites of

poly (lactic acid) reinforced with banana fibre and nanoclay. Journal of Reinforced Plastics and Composites. 2014;**33**(18):1717-1732

[18] Srinivasan V et al. Evaluation of mechanical and thermal properties of banana–flax based natural fibre composite. Materials & Design. 2014;**60**:620-627

[19] Uthayakumar M et al. Influence of red mud on the mechanical, damping and chemical resistance properties of banana/polyester hybrid composites. Materials & Design. 2014;**64**:270-279

[20] Haydaruzzaman et al. Fabrication and characterization of jute reinforced polypropylene composite: Effectiveness of coupling agents.
Journal of Composite Materials.
2010;44(16):1945-1963

[21] George G et al. Thermal, calorimetric and crystallisation behaviour of polypropylene/jute yarn bio-composites fabricated by commingling technique. Composites Part A: Applied Science and Manufacturing. 2013;**48**:110-120

[22] Arao Y et al. Strength improvement in injection-molded jute-fiber-reinforced polylactide green-composites. Composites Part B: Engineering. 2015;**68**:200-206

[23] Fu W, Xu X, Wu H. Mechanical and biodegradable properties of L-lactide-grafted sisal fiber reinforced polylactide composites. Journal of Reinforced Plastics and Composites. 2014;**33**(22):2034-2045

[24] Bajpai PK, Singh I. Drilling behavior of sisal fiber-reinforced polypropylene composite laminates. Journal of Reinforced Plastics and Composites. 2013;**32**(20):1569-1576

[25] Ibrahim H et al. Characteristics of starch-based biodegradable composites

reinforced with date palm and flax fibers. Carbohydrate Polymers. 2014;**101**:11-19

[26] Hallila T et al. Enzymatic pretreatment of seed flax-and polylactide-commingled nonwovens for composite processing. Journal of Thermoplastic Composite Materials. 2014;**27**(10):1387-1398

[27] Yuan Y, Guo M, Wang Y. Flax
fibers as reinforcement in poly (lactic acid) biodegradable composites. In:
Intelligent Computing and Information
Science. Berlin, Heidelberg: Springer;
2011. pp. 547-553

[28] Ochi S. Mechanical properties of kenaf fibers and kenaf/PLA composites. Mechanics of Materials. 2008;**40**(4-5):446-452

[29] Tawakkal IS, Cran MJ, Bigger SW. Effect of kenaf fibre loading and thymol concentration on the mechanical and thermal properties of PLA/kenaf/thymol composites. Industrial Crops and Products. 2014;**61**:74-83

[30] Di Franco C et al. Degradation of polycaprolactone/starch blends and composites with sisal fibre. Polymer Degradation and Stability. 2004;**86**(1):95-103

[31] Bastioli C. Properties and applications of mater-Bi starch-based materials. Polymer Degradation and Stability. 1998;**59**(1-3):263-272

[32] Alvarez V, Ruseckaite R, Vazquez
A. Degradation of sisal fibre/mater
Bi-Y biocomposites buried in soil.
Polymer Degradation and Stability.
2006;91(12):3156-3162

[33] Rutkowska M et al. Biodeterioration of mater-Bi Y class in compost with sewage sludge. Polish Journal of Environmental Studies.2004;13(1):85-90 [34] Scaffaro R et al. Degradation of mater-Bi®/wood flour biocomposites in active sewage sludge. Polymer Degradation and Stability. 2009;**94**(8): 1220-1229

[35] Huda M et al. "Green" composites from recycled cellulose and poly (lactic acid): Physico-mechanical and morphological properties evaluation.
Journal of Materials Science.
2005;40(16):4221-4229

[36] Plackett D et al. Biodegradable composites based on L-polylactide and jute fibres. Composites Science and Technology. 2003;**63**(9):1287-1296

[37] Kaplan D. Biopolymers from Renewable Resources: Macromolecular Systems-Materials Approach. Berlin: Springer-Verlag; 1998

[38] Dilnawaz F et al. Dual drug loaded superparamagnetic iron oxide nanoparticles for targeted cancer therapy. Biomaterials. 2010;**31**(13):3694-3706

[39] Rouilly A, Rigal L. Agro-materials: A bibliographic review. Journal of Macromolecular Science, Part C: Polymer Reviews. 2002;**42**(4):441-479

[40] Mousavi SM et al. Octadecyl amine functionalized Graphene oxide towards hydrophobic chemical resistant epoxy Nanocomposites. ChemistrySelect. 2018;**3**(25):7200-7207

[41] Bastioli C. Handbook of Biodegradable Polymers. United Kingdom: iSmithers Rapra Publishing; 2005

[42] Van de Velde K, Kiekens P. Biopolymers: Overview of several properties and consequences on their applications. Polymer Testing. 2002;**21**(4):433-442

[43] Shibata M et al. Biocomposites made from short abaca fiber and biodegradable polyesters. Macromolecular Materials and Engineering. 2003;**288**(1):35-43

[44] Meinander, K., et al. Polylactidesdegradable polymers for fibres and films. in Macromolecular Symposia. San Francisco, CA, USA: Wiley Online Library; 1997

[45] Klöpffer W. Life-Cycle Based Methods for Sustainable Product Development. Springer; 2003

[46] Nampoothiri KM, Nair NR, John RP. An overview of the recent developments in polylactide (PLA) research. Bioresource Technology. 2010;**101**(22):8493-8501

[47] David C, Fornasier R. Utilization of waste cellulose. 7. Kinetic study of the enzymatic hydrolysis of spruce wood pretreated by sodium hypochlorite. Macromolecules. 1986;**19**(3):552-557

[48] Paillet M, Peguy A. New biodegradable films from exploded wood solutions. Journal of Applied Polymer Science.1990;40(3-4):427-433

[49] Bledzki A, Reihmane S, Gassan
J. Properties and modification methods for vegetable fibers for natural fiber composites. Journal of Applied Polymer Science.
1996;59(8):1329-1336

[50] Hon DN. Chemical modification of lignocellulosic materials: Old chemistry, new approaches. Polymer News. 1992;**17**(4):102

[51] Ugbolue S. Structure/property relationships in textile fibres. Textile Progress. 1990;**20**(4):1-43

[52] Kucharyov M. Properties and modification methods for vegetable fibers for natural fiber composites. Tekst Prom. Journal of Applied Polymer Sciences. 1993;**23**:8-9

[53] Wuppertal E. Die Textilen Rohstoffe. Frankfurt: Dr. Spohr-Verlag; 1981

[54] Narayan R. Emerging technologies for materials and chemicals from biomass. In: ACS Symposium Series 476. American Chemical Society; 1992

[55] Varma D, Varma M, Varma I. Coir fibers: Part I: Effect of physical and chemical treatments on properties. Textile Research Journal. 1984;**54**(12):827-832

[56] Mukherjee P, Satyanarayana K. Structure and properties of some vegetable fibres. Journal of Materials Science. 1986;**21**(1):51-56

[57] Bledzki A, Gassan J. Natural Fiber Reinforced Plastics. New York: Marcel Dekker, Inc; 1997. pp. 787-809

[58] Varma D, Varma M, Varma I. Thermal behaviour of coir fibres. Thermochimica Acta. 1986;**108**:199-210

[59] Helfenbein D. Development of Recycling of PLA Sheet into PLA Pellets without Undergoing Chemical Process. Atlanta, GA: Atlanta; 2011

[60] Pillin I et al. Effect of thermomechanical cycles on the physicochemical properties of poly (lactic acid). Polymer Degradation and Stability. 2008;**93**(2):321-328

[61] Żenkiewicz M et al. Characterisation of multi-extruded poly (lactic acid). Polymer Testing. 2009;**28**(4):412-418

[62] Badia J et al. Material valorisation of amorphous polylactide. Influence of thermo-mechanical degradation on the morphology, segmental dynamics, thermal and mechanical performance. Polymer Degradation and Stability. 2012;**97**(4):670-678 [63] Ohya Y, Maruhashi S, Ouchi
T. Preparation of poly (lactic acid)-grafted amylose through the trimethylsilyl protection method and its biodegradation.
Macromolecular Chemistry and Physics.
1998;199(9):2017-2022

[64] Ouchi T, Kontani T, Ohya Y. Modification of polylactide upon physical properties by solutioncast blends from polylactide and polylactide-grafted dextran. Polymer. 2003;44(14):3927-3933

[65] Park K, Xanthos M. A study on the degradation of polylactic acid in the presence of phosphonium ionic liquids. Polymer Degradation and Stability. 2009;**94**(5):834-844

[66] Gang W, Aimin L. Thermal decomposition and kinetics of mixtures of polylactic acid and biomass during copyrolysis. Chinese Journal of Chemical Engineering. 2008;**16**(6):929-933

[67] De Jong S et al. New insights into the hydrolytic degradation of poly (lactic acid): Participation of the alcohol terminus. Polymer. 2001;**42**(7):2795-2802

[68] Ahmann D, Dorgan JR.
Bioengineering for pollution prevention through development of biobased energy and materials state of the science report. Industrial Biotechnology.
2007;3(3):218-259

[69] Lagaron JM, Lopez-Rubio A. Nanotechnology for bioplastics: Opportunities, challenges and strategies. Trends in Food Science & Technology. 2011;**22**(11):611-617

[70] Vilaplana F, Strömberg E, Karlsson
S. Environmental and resource aspects of sustainable biocomposites.
Polymer Degradation and Stability.
2010;95(11):2147-2161 [71] Arias JL et al. Development of iron/ethyl cellulose (core/shell) nanoparticles loaded with diclofenac sodium for arthritis treatment.
International Journal of Pharmaceutics.
2009;382(1-2):270-276

[72] Álvarez-Chávez C et al. Sustainability of bio-based plastics: General comparative analysis. Journal of Cleaner Production. 2012;**23**:47-56

[73] Álvarez-Chávez CR et al. Sustainability of bio-based plastics: General comparative analysis and recommendations for improvement. Journal of Cleaner Production. 2012;**23**(1):47-56

[74] Mousavi SM et al. Development of Clay Nanoparticles toward Bio and Medical Applications. Bioresources. Intech open; 2018. pp. 167-191

[75] Hubbe MA et al. Cellulosic nanocomposites: A review. BioResources. 2008;**3**(3):929-980

[76] Eichhorn SJ et al. Current international research into cellulose nanofibres and nanocomposites. Journal of Materials Science. 2010;**45**(1):1

[77] Mousavi S et al. Investigation of physical, mechanical and biodegradation properties of nitrile butadiene rubber by natural polymers and Nano-silica particles. American International Journal of Research in Formal, Applied and Natural Sciences. 2014;5:110-117

[78] Farazi Z, Abdolrasoul O, Mojtaba MS, Alireza HS. Preparation of LDPE/EVA/PE-MA, Nano clay blend composite in the stage potassium Sorbate (KS) and garlic oil (GO) as an antimicrobial substance. Polymer Sciences. 2018;4:1-12

