



# Multicomponent Polymer Systems Based on Agro-Industrial Waste

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This work is dedicated to the memory of Prof. Cristofor I. Simionescu (1920–2007) on the occasion of the commemoration of the centenary of his birth.

## Abstract

Nowadays concerns over global warming favored research to focus on the environmental pollution and waste accumulation and on the possibility to recycle and up-cycle the waste. A particular class of such waste consists of natural remains and by-products from agriculture and industries, such as forestry, pulp and paper, furniture, and food industry. There are various methods to upcycle and capitalize the agro-industrial waste: (1) anaerobic digestion for energy production; (2) thermal conversion for the resulting liquids and char that may be further used in obtaining other materials, bioplastics included; (3) industrial processing to obtain natural fibers, long or short, able to be employed as reinforcing materials in biocomposites (multicomponent systems where the matrix consists of polymers or polymer blends, either synthetic or natural; they may also contain emulsifiers, compatibilization agents, antioxidants, as well as inorganic, organic, metal, or metal oxides micro- and/or nanoparticles); and (4) composting. Composites based on agro-industrial waste displayed a level of performance high enough to grant their use in a wide variety of applications:

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furniture and civil constructions; interior design, fencing, and decking; automotive and aerospace industry; food and general packaging. To obtain such materials, it is necessary to consider specific issues, the most important being the compatibility between matrices and reinforcing material. Characteristics of the final material (mechanical, thermal, water sorption, weather resistance) strongly depend on a good compatibility. A critical assessment of the selected methodology is necessary as it is reflected in the final cost of production, which must be limited in relation to the envisaged application.

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**Keywords**

Multicomponent systems · Agricultural waste · Industrial waste · Applications

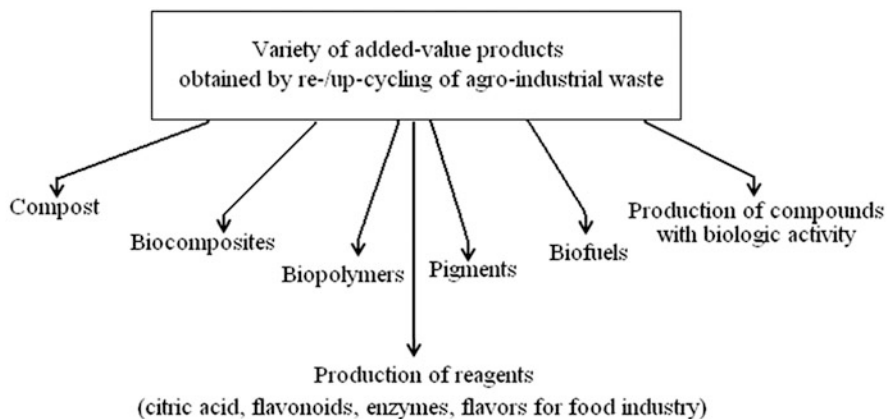
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## 18.1 Introduction

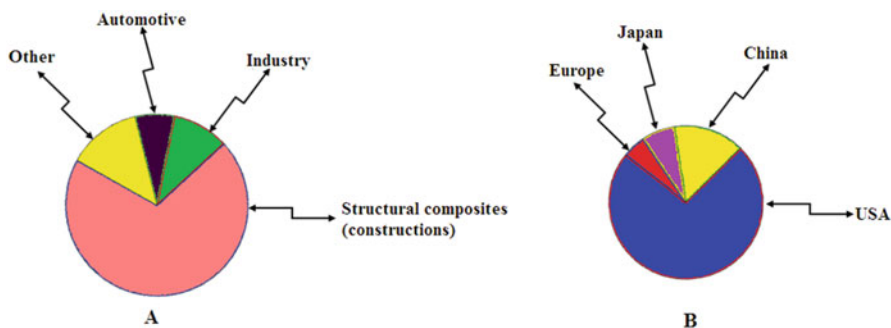
Modern days concerns over the global warming favored research to focus on the environmental pollution and waste accumulation on one hand and, on the other hand, on the possibility to recycle and up-cycle the waste, in order to response to the societal needs and to replace, at least partially, the classic materials based on fossil resources. One very particular class of re- and up-cyclable waste consists of natural remains and by-products (wood chips and sawdust, straw, husk, stalk and leaves, pulp, etc.) from agriculture and various industries, such as forestry, pulp and paper, furniture, food industry, etc. The chemical composition of such waste varies in correlation with their nature (animal origin—proteins: silk fibroin, wool fibers, egg shells; plant origin—carbohydrates: cellulose, lignin, hemicellulose; polyphenols; etc.) and location (marine organisms—carbohydrates: chitin, chitosan, etc.; proteins—seashells).

There are various methods to up cycle and capitalize the agro-industrial waste: (1) the anaerobic digestion—used mainly for energy production (gas); (2) thermal conversion—the resulting liquids and char may be further used in obtaining other materials, bioplastics included; (3) industrial processing to obtain natural fibers, long or short, able to be employed as reinforcing materials in biocomposites (multicomponent systems where the matrix consists of polymers or polymer blends, either synthetic or natural; they may also contain emulsifiers, compatibilization agents, antioxidants, as well as inorganic, organic, metal or metal oxides micro- and/or nanoparticles); and (4) composting. The wide variety of added-value products (Fig. 18.1) strongly depends on the type of approach, degree of transformation, technology used for processing, range of applications, possibility to include these processes into circular economic loops (recovery and reuse) in order to limit and/or eliminate waste disposal, etc.

In this regard, a more responsible selection and disposal of agro-industrial waste, the employment of knowledge-based, and sometimes state-of-the-art, technologies (single or integrated in synergic combinations), and committed local policies inspired from the regional strategies for the environment protection and preservation



**Fig. 18.1** Methods to upcycle and capitalize the agro-industrial waste



**Fig. 18.2** Thermoplastic biocomposites market shares: (a) in applications; (b) in different countries. (Adapted from Zaaba and Ismail 2019)

are key factors. The market indicators confirmed that structural composites (building panels, railing, fencing, decking, roofing, siding, interior compartmenting) recorded the largest share, based on high volume sales, while industry (automotive industry included) comes the second, despite the knowledge-based development of new products with high added value (Fig. 18.2a). These conclusions are in good concordance with the data for the market shares of different countries (Fig. 18.2b) (Zaaba and Ismail 2019).

Another problem related to this subject is the land management, and it originated in the increasing demand for food and natural fibers. Maintaining a wise balance between production and demand, between land depletion and sustainable agriculture, and all the time preserving the whole ecosystem, may offer tools to provide for both social and economic needs, without sacrificing this natural resource—agricultural land—for the future generations.

This chapter offers an overview of some of the most recent literature data concerning the composites based on polymer matrices and agro-industrial waste as

reinforcement, using the type of polymer matrix as classification criterion. Specific problems related to the manufacture of polymer composites based on agro-industrial waste will be presented, as well as some advantages and disadvantages of these composites.

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## 18.2 Manufacture of the Polymer Composites Based on Agro-Industrial Waste

Polymer composites based on agro-industrial waste displayed a level of performance high enough to grant their use in a wide variety of applications: furniture and civil constructions; interior design, fencing, and decking; automotive; sportswear; food and packaging; and adsorbents and filtration media (Berthet et al. 2016; Leao et al. 2010; Pandey et al. 2010; Thakur et al. 2014; Sanyang et al. 2016; Tanasă et al. 2020a). To this purpose, it is necessary to take under consideration some specific issues. The compatibility between matrix and reinforcing material (whether it is fibers, particles or powder) is of utmost importance as it determines the dimensions of the interphase and type and strength of interactions at the interface between components, hence the compatibility and adhesion of components. The characteristics of the final material (mechanical strength, thermal properties, water sorption, UV and biologic resistance) strongly depend on these parameters. Therefore, all aspects of manufacturing these materials must be controlled so as to enable all factors to converge to the desired end products.

Thus, uniformity, consistency, and predictability of composite properties may be reached by using particles/fibers of a specific particle size and dimensional dispersity, but for each type of composite particle size must be wisely considered as some applications require a narrow particle size distribution, while others benefit from the mixture of particles/fibers of different size and sometimes different shapes (Kumar et al. 2017, 2020). Associated to this issue, harvesting, transport, storage, and preliminary preparation steps (e.g., dusting, drying etc.) must also be taken into consideration because these operations will be reflected in the final cost of production.

Another problem that must be addressed is the nature of the polymer matrix employed in these composites manufacture. Literature of the last decades abounds in reports on various composite formulations specifically designed for agro-industrial waste reinforcement. Be they thermoplastics (polyethylene PE, polypropylene PP, polyamides PA, etc.) or thermosets (urea-formaldehyde UF, phenyl-formaldehyde PF, methylene diphenyl diisocyanate MDI, etc.), synthetic or natural or naturally derived (starch, lignin, poly(lactic acid) PLA etc.), and organic or inorganic (gypsum, Portland cement, etc.), the main issue is to choose the most suitable matrix for the selected reinforcement and the envisaged application. Subsequently, the technology must be adapted to the specifications for each type of polymer and reinforcement, whether the end products are particleboards or fiberboards (Abba et al. 2013).

In close relation with these technological aspects are, at the same time, the compatibilization strategies that have to be adopted in a discriminative manner.

Typically, the compatibilization methods may be applied to (1) the polymer matrix (Zănoagă and Tanasă 2011; Zanoaga et al. 2016), (2) reinforcement (Li et al. 2012; Singha and Rana 2012), or (3) both matrix and reinforcement (Pracella et al. 2010; Teaca et al. 2018; Zaaba and Ismail 2019). The surface functionalization of agro-industrial waste intended for polymer composites can be achieved by different means and methods: chemical, physical, mechanical, radiative, etc. (Thakur et al. 2015; Collins et al. 2019; Ferreira et al. 2019; Tanasă et al. 2020b), each of them with advantages and drawbacks, but all aiming at reducing the specific hydrophilicity of natural fibers versus the hydrophobic character of polymer matrices and, by consequence, increasing the dimensions of the interphase and improving the interfacial interactions. Hence, a better compatibility of components and a higher level of performance of composites can be achieved.

Nevertheless, properties other than mechanical ones can be also improved by using different additives in composite formulation. Thus, resistance toward water and humidity can be achieved by particle/fiber functionalization, as well as by adding natural (rosin, waxes) or synthetic resins, which may act at the surface of the composite or in-depth (Zach et al. 2014; Brzyski et al. 2017; Cichosz and Masek 2019). Hydrophobization has proven to be important not only for composites used in outdoors applications, but indoors too, as it grants composites dimensional stability due to a limited adsorption of water. Thus, crack formation, and subsequently mechanical failure, is significantly delayed. Even more, composites with increased resistance toward water and humidity have, at the same time, a good resistance to weathering and biologic attack when used in outdoor applications. Flame retardants may also be included in these composites formulation in order to render them improved thermal stability and flame resistance. Some of these additives are inorganic compounds (Sain et al. 2004); others are organic products or polymers (Subasinghe and Bhattacharyya 2014; Toldy et al. 2017; Elsabbagh et al. 2018). Depending on the chemical structure of these additives and their heteroatom, they can act in different stages of the burning process and in different phases as well (gas, liquid, or solid). Halogen-containing compounds act as gas with flame inhibiting effect, while phosphorus-based agents reduce the formation of flammable vapors by increasing the char formation upon pyrolysis (Sain et al. 2004).

Weathering, photochemical degradation, and biologic attack must be also considered when it comes to manufacturing composites made of polymers and agro-industrial waste. Their behavior upon UV radiation and under complex weathering conditions (variation of temperature, seasonal or freeze–thaw cycles, humidity, pH, pollutants, ozone, microorganisms, wind, solid particles in air) is of high importance in terms of functional reliability and service life (Zanoaga and Tanasa 2016; Teaca et al. 2019). After addressing these problems, multicomponent polymer systems with complex composition result at the end of the technological process. The wise selection of components and processing approach enables them to act in synergy as to respond to requirements of the envisaged applications.

## 18.3 Synthetic Polymer Matrices

Synthetic polymers reinforced with agro-industrial waste still retain the interest of scientists in both fundamental research and industrial applications because these materials offer certain possibilities for the waste management and allow energy and material recovery. The most commonly used synthetic polymers for the manufacture of such composites are thermoplastics (polyethylene PE, polypropylene PP, polyamides PA, polystyrene PS) and thermosets (urea-formaldehyde UF, phenyl-formaldehyde PF, methylene diphenyl diisocyanate MDI, epoxydic resins, unsaturated esters). Some illustrative data regarding the main characteristics of thermoplastics as compared to thermosetting polymers are summarized in Table 18.1.

### 18.3.1 Thermoplastic Matrices for Composites with Agro-Industrial Waste Reinforcement

#### 18.3.1.1 General Considerations

Thermoplastic polymers are highly appreciated as matrices for a wide variety of composites and biocomposites, but thermoplastics with low melting temperatures are particularly preferred for composites with natural fillers or reinforcements, such as wood chips, shives of flax flakes, cereal straws, corn and cotton stalks, olive wet/dry husk and pits, tomato peels, grape stalks and pomace, textile fiber waste, etc. (Kozłowski and Władyska-Przybylak 2004; Thakur et al. 2014; Navas et al. 2015; Zaaba and Ismail 2019). Typically, polyolefins (PP, PE) and polyamides (PA,

**Table 18.1** Main properties of thermoplastics versus thermosets<sup>a</sup>

Polymer	Density (g/cm <sup>3</sup> )	Melt temperature (°C)	Tensile strength (MPa)	Elastic modulus (GPa)	Elongation at break (%)
Thermoplastic polymers					
PE	0.91–0.96	105–140	15–78	0.055–1.5	2–800
PP	0.90–0.92	160–176	26–41	1.0–1.8	15–700
PS	1.04–1.06	110–135	25–69	4.0–5.0	1–3
PVC	1.40	100–260	41–52	2.8	40–80
Copolyamide <sup>b</sup>	1.11	125–135	20	1.1	300
Thermosetting polymers					
Polyester	1.0–1.5	90–250	40–90	2.0–4.5	< 2.6
Vinylester	1.2–1.4	86	69–86	3.1–3.8	4–7
Epoxy	1.1–1.6	100–200	28–100	3.0–6.0	1–6
Phenolic	1.29	210–360	35–62	2.8–4.8	1.5–2.0
Polyurethane	1.04–1.46	–	41–90	2.1–4.4	1–3

<sup>a</sup>Adapted from Ku et al. (2011), Beyler and Hirschler (2001), Holbery and Houston (2006), Bogoeva-Gaceva et al. (2007)

<sup>b</sup>Zanoaga and Tanasa (2002)

mainly aliphatic or mixed aliphatic-aromatic PA, with low melting point, and having properties as hot-melt adhesive, if possible (Zanoaga and Tanasa 2002; Zanoaga et al. 2016)), polyvinyl chloride (PVC) (Wang et al. 2008; Zainudin et al. 2009), polystyrene (PS), or even polymer blends (Clemons 2008) are widely considered. In this regard, the most sensitive issue is the processing temperature which usually has to be maintained under 200 °C to prevent the thermal degradation of the natural fillers (Väisänen et al. 2016). Compared to thermosets, thermoplastics offer some benefits, such as the possibility to recover and re-use them (up to a certain extent and with minimum property decay) (Bourmaud and Baley 2007). Even more, in certain cases the matrix is made of polymer waste, if appropriate, from the very beginning (Zanoaga and Tanasa 2011). In terms of mechanical properties, thermoplastic matrices afford tensile strength values comparable with the thermosets, but the elasticity of these composites is definitely superior. Given the hydrophilic nature of agro-industrial waste and their ability to absorb water even when included in plastic composite formulations, the poor compatibility with the matrices is obvious. Compatibilization strategies were designed and successfully applied, as follows:

- Functionalization—(1) surface functionalization of fillers and (2) polymer functionalization—refers to various methods to chemically and/or physically alter one or both components of the composite (Dányádi et al. 2010; Moghadamzadeh et al. 2011; Tanasa et al. 2020b);
- Use of coupling agents which are low molecular/oligomeric reagents used to promote chemical bonds formation between matrix and filler, as well as hydrogen bonding, and polymer entanglement; in the case of polyolefins, the most commonly used are maleated (maleic anhydride MA) oligomers (PP-*g*-MA, PE-*g*-MA); other coupling agents are monofunctional isocyanates, *m*-phenylene bismaleimide, etc. (Mohanty et al. 2006; Väisänen et al. 2016; Zanoaga et al. 2016; Tanasa and Zanoaga 2018).

From technological point of view, most industrial equipment and technologies employed for processing thermoplastics can be used to manufacture thermoplastic composites with natural fillers. Among other parameters, the mixing regime has to be carefully considered as it can negatively influence the properties of the final product, as follows:

- The use of one screw mixer/blender might be ineffective in terms of distribution of filler inside the matrix, even at high speeds, because it may cause secondary agglomeration of the filler and cannot mobilize the material stuck in the blind spots of the machine.
- If a twin-screw (co-rotating or counter-rotating) extruder is employed, the speed must be preset so that no mechanical degradation of polymers will occur due to the high values of the shear force created between screws (especially for the counter-rotating screws).
- Although the fact that co-rotating extruders provide better mixing than the counter-rotating extruders is still under debate, it was experimentally proved the counter-rotating twin-screw extruders generate a higher-pressure build-up,

so they may be preferred over co-rotating twin-screw extruders for profile extrusion (Shah and Gupta 2004).

As a direct result of the combined effects of all the above-mentioned factors, the main properties of the final composites get improved.

**Mechanical properties:** Typically, natural particles and fibers have a reinforcing effect so that an increase in tensile strength and elastic modulus is evident (El-Saied et al. 2012). Tensile strength is more sensitive to the polymer matrix, while the elastic modulus depends mainly on the reinforcement. Therefore, strong interfacial interactions (adhesion and cohesion forces) are the condition for the composite to reach an increased tensile strength, along with orientation of fibers (when required) and a low concentration of stress centers. On the other hand, higher values of tensile modulus are achieved at high amounts of filler and when the polymer matrix is wetting very well the filler particles, and a large interphase results. The aspect ratio of particles/fibers strongly influences the fracture properties of the composite: the higher the ratio, the better the fracture behavior (Nourbakhsh and Ashori 2010; Viksne et al. 2010).

In principle, there is a minimum critical length of the particles/fibers for the composite to reach high performance mechanical properties. If the length of the filler is less than that, the interphase is not strong enough as the particles cannot create a co-continuous phase within the matrix, and debonding occurs regardless the nature of the thermoplastic. When the length exceeds the critical length, the composite may become brittle (especially at high amounts of reinforcement) because the matrix cannot wet satisfactorily the particles/fibers and the system becomes rigid (Tanasă et al. 2014). Secondary alteration of fibers (curling, bending) may also occur (Väisänen et al. 2016). At this point, the impact strength of the composite is compromised.

**Water sorption:** The amount of water absorbed by a composite reinforced with agro-industrial waste strongly depends on the type and nature of the reinforcement. Thus, eggshells and seashells (Owuamanam and Cree 2020) will cause a much lower water sorption than feathers (Bullions et al. 2006) and wood fibers or particles, rice husk, or kenaf fibers (Tajvidi et al. 2006). When referring to natural fiber-reinforced thermoplastics, an almost linear correlation was found between the amount of filler and the water sorption of the composite (George et al. 1998), but the fiber size is important as well, as it contributes to transfer water molecules from the surface to interior and inside the composite by diffusion from fiber to fiber (Migneault et al. 2008). This is, in fact, the dominant mechanism of water sorption in composites with high load of reinforcement, when large bundles of filler stick together. When the filler content is low, then the mechanism is percolation (Wang et al. 2006).

**Thermal characteristics:** The thermal degradation of polymers reinforced with natural fillers evolves in stages. It starts with the loss of water linked physically and chemically onto the filler surface, continues with the decomposition of the natural filler, and the final stage being the thermal degradation of the polymer matrix (Monteiro et al. 2012; Stokke et al. 2014). When thermal degradation of filler is initiated during the service time, the decay of other properties takes place in avalanche: mechanical properties weakened, the exterior aspect is no longer esthetic,



bad odors develop under biologic attack, etc. Moreover, the thermal stability and flame resistance of these composites must be considered from different standpoints, such as: (a) type and nature of filler—for example, flax fibers have long ignition time and reach the flashover point very slowly (Manfredi et al. 2006); (b) addition of flame retardants (Chapple and Anandjiwala 2010).

Some of the most recent reports on thermoplastic composites with agro-industrial reinforcements will be reviewed in the following section, using as criterion the nature of the polymer matrix.

### 18.3.1.2 Thermoplastic Matrices for Composites with Agro-Industrial Waste

**Polyolefins:** PP and PE are the most commonly used thermoplastics in composite formulations. In any form, either, freshly prepared polymers or recovered waste, or high density or low density polymers, with or without a coupling agent, or employed in either structural or non-structural, indoors or outdoors applications, PP and PE showed a versatility that makes them suitable candidate for agro-industrial waste for reinforcement. Some illustrative examples are summarized in Table 18.2.

**Polystyrene (PS):** PS is a synthetic, inexpensive, relatively low melting point aromatic polymer, with poor barrier properties toward oxygen and water vapor; it is non-biodegradable, but easy to process by extrusion (Styrofoam), molding, and vacuum forming. Composites based on PS and agro-industrial waste showed good mechanical and thermal properties, as presented in Table 18.3.

**Polyvinyl chloride (PVC):** PVC is the world's third most widely produced synthetic plastic polymer (after PE and PP); it comes in two basic forms: rigid (RPVC—used for pipes and in profile applications, such as doors and windows) and flexible (by the addition of plasticizers). In composites with cotton or linen, it is used for the manufacture of canvas. Composites with agro-industrial waste as reinforcement were prepared using various additives, such as compatibilizers, impact modifiers, and lubricants, as summarized in Table 18.4.

**Polyamides (PA) and co-polyamides:** Polyamides and co-polyamides are a viable alternative to polyolefins as matrices in composites reinforced with agro-industrial waste (Taj et al. 2007). Their lower hydrophobic character as compared with polyolefins favors the matrix-filler interfacial bonding, even without adding compatibilizers. Still, there are other processing difficulties due to their higher melting temperatures, so the composite manufacturing must be adapted as to avoid the thermal degradation of natural reinforcement. Some examples are presented in Table 18.5.

**Thermoplastic polymer blends:** Composites with agro-industrial waste were produced using thermoplastic polymer blends as matrix as well. Various combinations were considered, sometimes even incompatible polymers, and applied with satisfactory results, as summarized in Table 18.6.

Most noticeable improvements were recorded for the mechanical properties of composites, but thermal stability and flame retardancy and resistance to water increased as well upon the addition of coupling agents and/or other selected

**Table 18.2** Thermoplastic composites with polyolefins as matrix

Polymer matrix	Agro-industrial waste/additive	Manufacturing method	Results/observation	References
Polyethylene (PE)				
Recycled PE	<ul style="list-style-type: none"> <li>• 20–39% wood sawdust</li> <li>• 3 wt% Exxelor VA 1803 as compatibilizer.</li> </ul>	Extrusion and compression method	The wood sawdust in the PE matrix decreased the thermal stability of the blends	Constantinescu and Cazacu (2007)
HDPE	<ul style="list-style-type: none"> <li>• 20, 40, and 60 wt% wood flour</li> <li>• 20, 30, 40, and 60 wt% paper sludge</li> <li>• 2 and 3 wt% coupling agent (PE-g-MA)</li> </ul>	Co-rotating twin-screw extruder, granulated and injection-molded samples	Tensile modulus and flexural properties of the composites increased by the addition of the sludge and MAPE, while water absorption and thickness swelling decreased	Hamzeh et al. (2011)
HDPE	<ul style="list-style-type: none"> <li>• 30 wt.% wood flour</li> <li>• 0, 2, and 4 wt.% modified montmorillonite</li> <li>• 3 wt.% PE-g-MA as coupling agent</li> </ul>	Injection molding	The addition of nanoclay provided indirect protection against weathering, little decrease in modulus of elasticity due to low water absorption, and reduced oxidative action	Eshraghi et al. (2016)
RecycledPE	<ul style="list-style-type: none"> <li>• 30–40% waste: cacao husks, beet pulp, husks of buckwheat, rice, millet and sunflowers, and potato and maize pulp (maximum size of the filler particles was <math>\leq 150\mu\text{m}</math>)</li> <li>• 2% bentonite as dispersing agent</li> <li>• 2% activated carbon as sorbent</li> </ul>	Extrusion and compression molding	Composites with cacao husks, beet pulp, and rice husks showed good mechanical properties	Kirsh and Chutkina (2011)
LDPE	<ul style="list-style-type: none"> <li>• 4 wt% rice husk</li> <li>• 6 wt% MAPE</li> <li>• 0–6 wt% MMT</li> </ul>	Extrusion blown films	Good mechanical properties for a	Majeed et al. (2017)

(continued)

**Table 18.2** (continued)

Polymer matrix	Agro-industrial waste/additive	Manufacturing method	Results/observation	References
			variety of food packaging	
LDPE	<ul style="list-style-type: none"> <li>• 2, 5, and 7 wt.% rice husk</li> <li>• 2 wt% MMT</li> </ul>	Extrusion blown films	<ul style="list-style-type: none"> <li>– Increased tensile modulus</li> <li>– Decreasing tensile and tear strength</li> </ul>	Majeed et al. (2017)
LDPE	<ul style="list-style-type: none"> <li>• Rice husk</li> <li>• MAPE</li> <li>• 0–6 wt% MMT</li> </ul>	Extrusion blown films	Improved tensile strength and tear strength, O <sub>2</sub> barrier properties, and thermal stability	Majeed et al. (2014a)
LDPE	<ul style="list-style-type: none"> <li>• 5 wt% rice husk flour</li> <li>• 3 wt% MMT</li> <li>• 0–8 wt% MAPE</li> </ul>	Extrusion blown films	Composite films showed improved tensile strength and oxygen barrier properties	Majeed et al. (2014b)
Polypropylene (PP)				
PP	<ul style="list-style-type: none"> <li>• 40–69 wt % wood dust</li> <li>• 2–6 wt % Cloisite<sup>®</sup> 15A</li> <li>• 4 wt % MAPP</li> </ul>	<ul style="list-style-type: none"> <li>– Extrusion into a counter-rotating twin-screw extruder</li> <li>– Compression</li> </ul>	<ul style="list-style-type: none"> <li>– Low production costs</li> <li>– Limits the accumulation and disposal of wood industry waste</li> </ul>	Madhoushi et al. (2014)
PP	<ul style="list-style-type: none"> <li>• 0, 10, 20, 30 wt. % wood chips</li> <li>• 0, 1, 3, and 5 wt. % Cloisite 20A</li> <li>• 0, 3, 5, and 10 wt. % MAPP</li> </ul>	Melt blending	<ul style="list-style-type: none"> <li>– Improved thermal stability of composites</li> <li>– Increased tensile modulus</li> </ul>	Tanasa et al. (2014)
PP	<ul style="list-style-type: none"> <li>• 10, 20, 30, 40, 50 wt%, respectively— wood flour treated with the dopamine hydrochloride</li> <li>• PP and MAH-g-PP (PP/MAH: E-43 Epolene) = 80/20)</li> </ul>	Mixer mixing, secondly mixing into torque rheometer mixer, injection molding under pressure	The use of polydopamine-treated wood flour for preparing WPCs can be an efficient way to improve the water resistance of WPCs	Lanh and Thien (2016)
PP	<ul style="list-style-type: none"> <li>• 0–60 wt% wood flour</li> <li>• 60 to 0 wt% charcoal powder</li> <li>• 3 wt% MAPP</li> </ul>	Rotary drum blender, hot press molding	The swelling and water sorption considerably decreased along with the	Ayrilmis et al. (2015)

(continued)

**Table 18.2** (continued)

Polymer matrix	Agro-industrial waste/additive	Manufacturing method	Results/observation	References
			increasing charcoal content, while the strength and bending significantly improved	
PP	<ul style="list-style-type: none"> <li>• 30 wt% biomass wastes (rice husk, coffee husk, coarse wool, landfill wood)</li> <li>• 24 wt% biochar</li> <li>• 4 wt% MAPP</li> </ul>	Melt blending followed by injection molding	Wood and biochar composite exhibited the highest values of tensile/flexural properties with a relatively low peak heat release rate. Drastically reduced susceptibility toward fire	Das et al. (2018)
PP	<ul style="list-style-type: none"> <li>• 40% filler corn stalk waste/flour</li> <li>• 6 wt% Epolene<sup>®</sup> G3015 as coupling agent (maleic anhydride-grafted polypropylene MAPP)</li> </ul>	Mixing components in an internal mixer, palletizing, injection-molding	Tensile strength and modulus of the composites could be improved by using MAPP coupling	Flandez et al. (2012)
PP	<ul style="list-style-type: none"> <li>• 0, 10, 20, 30, 40% corn cob (CC)</li> <li>• 3 wt.% MAPP</li> </ul>		Tensile strength and elongation at break of biocomposites decreased with increasing corn cob loading, while modulus of elasticity increased	Salmah et al. (2014)
PP	<ul style="list-style-type: none"> <li>• 10, 30, and 50 wt % buckwheat husk</li> <li>• PP-g-MA</li> </ul>	Injection molding	Mechanical properties do not differ significantly from those obtained with wood flour. Improved rheological characteristics	Andrzejewski et al. (2019a, b)

(continued)

**Table 18.2** (continued)

Polymer matrix	Agro-industrial waste/additive	Manufacturing method	Results/observation	References
PP	• 20/20 and 10/30% filler (talc/eggshell)	Blending with an internal mixer, palletized, pressed	The Young's modulus ( $E$ ) improved with the increment of eggshell content	Toro et al. (2007)

additives. Since all components of the system are acting in synergy, the overall effect is a result of the combined effects of each of them interacting with the others.

### 18.3.2 Thermoset Polymer Matrix Biocomposites

As polymer matrices, thermosets, used alone or blended with other materials, undergo a cross-linking reaction during the curing process to form irreversible chemical bonds. Thermoset plastics always remain in a permanent solid state after an initial heat treatment, and therefore they are no longer affected by additional heat exposure. As a consequence, they cannot be melted, reshaped, recycled, and re-molded as part of a conventional recycling process. Thermoset plastics significantly improve the chemical and heat resistance, structural integrity, and mechanical properties like strength, hardness, and moldability of the resulted materials. Thermoset components that require tight tolerances, superior strength-to-weight ratio and performance, outstanding dielectric strength, low thermal conductivity, and resistance to corrosion are extensively used in high-heat applications such as electronics, appliances, automotive, electrical, lighting, and energy markets, and they are often used for sealed products due to their resistance to deformation. At the same time, thermosets present certain disadvantages such as low initial viscosity of materials which leads to supplementary operations, low tensile strength and ductility that need designs with thick walls, significant variation and less consistency from lot-to-lot in batch processes, poor thermal conductivity for housing replacements, and high rigidity resulting in product failure when used in high vibration applications (Väisänen et al. 2016).

Based on the two thermoset molding compound processes, namely reaction injection molding (RIM) and resin transfer molding (RTM), there are specific types of thermoset polymers, such as polydicyclopentadiene, polyurethanes and polyureas (RIM), polyesters, polyimides and vinyl ester (RTM), bismaleimide, epoxy, fluoropolymers, melamine, phenolics, polytetrafluoroethylene, polyvinylidene fluoride, silicone, and urea-formaldehyde. Thermoset composites consist in a large variety of formulations. Usually, matrix is a thermosetting resin (unsaturated polyesters, epoxies, vinyl esters, polyurethanes, etc.) that binds the reinforcement/filler (particles, chips, short or continuous fibers) together for an efficient transfer of load between them. Some properties of typical thermosets have been already illustrated in Table 18.1. The basic fabrication methods to prepare

**Table 18.3** Thermoplastic composites with PS matrices

Polymer matrix	Agro-industrial waste/additive	Manufacturing method	Results/observation	References
Expanded PS waste	<ul style="list-style-type: none"> <li>Extruded PS waste</li> <li>10, 20, 30, and 40 wt.% of wood flour</li> <li>2 wt% coupling agent—a poly (styrene-co-maleic anhydride) oligomer with 30 wt.% of maleic anhydride groups</li> </ul>	Processed in a co-rotating twin-screw extruder	The use of poly (styrene-co-maleic anhydride) as a coupling agent improved the compatibility between the wood flour and polystyrene matrix and the mechanical properties subsequently improved. The mechanical properties decreased as the wood flour loading increased.	Poletto et al. (2011)
Recycled PS	<ul style="list-style-type: none"> <li>Recycled PS</li> <li>20 wt% wood flour</li> <li>2 wt% three different styrene-co-maleic anhydride (SMA) oligomers as coupling agents</li> </ul>	Processed in a co-rotating twin-screw extruder	The compatibilizers increased composite thermal stability and activation energy of composites.	Poletto et al. (2012)
Expanded PS waste	<ul style="list-style-type: none"> <li>80 and 40 wt% expanded PS waste</li> <li>20 and 40% wt% wood flour</li> <li>2 wt % poly (styrene-co-maleic anhydride) as compatibilizer</li> </ul>	Processed in a twin-screw co-rotating extruder	The mechanical and dynamic mechanical properties decreased with the wood flour loading, for composites without coupling agent. When the coupling agent was used, all these properties increased.	Poletto (2017)
Expanded PS waste	<ul style="list-style-type: none"> <li>Expanded PS waste</li> <li>20 w/w% of wood flour, with and without 1, 2, and 4 wt% of expanded PS</li> <li>Styrene maleic anhydride as coupling agents, with different content of maleic anhydride groups, 30%, 25%, and 20% (w/w) and levels of 1, 2, and 4% in composites</li> </ul>	Processed on a twin-screw co-rotating extruder	The addition of styrene maleic anhydride increased the wood plastic composites mechanical properties. The improvement in composite mechanical properties using SMA as coupling agent was strongly dependent on the amount of maleic anhydride.	Poletto (2016)
Expanded PS	<ul style="list-style-type: none"> <li>Expanded PS</li> <li>60% hemp shive particles</li> </ul>	Preformed by cold mold pressure	The thermoforming pressure improved the mechanical properties of the composites.	Almusawi et al. (2017)

**Table 18.4** Thermoplastic composites with PVC matrices

Polymer matrix	Agro-industrial waste/additive	Manufacturing method	Results/observation	References
PVC	<ul style="list-style-type: none"> <li>• 0, 10, 20, 30, and 40 wt % rice husk powder</li> <li>• MA as compatibilizer</li> </ul>	Mixed in a mixer grinder followed by compression molding	Mechanical properties of composites increased along with the amount of filler, while arc resistance decreased. Thermal and mechanical properties improved even more upon addition of MA.	Chand and Jhod (2008)
Un-plasticized PVC (PVC-U)	<ul style="list-style-type: none"> <li>• 10–40 phr rice husk</li> <li>• Acrylic impact modifiers</li> </ul>	Using a two-roll mill at temperature of 165 °C	Flexural strength for both unmodified and modified PVC-U composites increased up to RH loading of 20 phr. The tensile and impact strength of PVC-U composite decreased with RH loading.	Ahmad et al. (2010)
Plasticized polyvinyl chloride (PPVC)	<ul style="list-style-type: none"> <li>• 0, 20, 30, and 40% rice husk</li> </ul>	Molding on a hot press	Composites with 20% rice husk loading had the best mechanical properties compared to 30 and 40% rice husk loading due to high tensile and flexural strength.	Rahman et al. (2014)
PVC	<ul style="list-style-type: none"> <li>• 16.7, 28.6, and 37.5 wt% wood sawdust</li> </ul>	Extrusion	Tensile strength decreased with increasing moisture content up to 1–2% and then unexpectedly increased at higher moisture contents. Impact strength of the composites was considerably improved with	Sombatsompop and Chaochanchaikul (2004)

(continued)

**Table 18.4** (continued)

Polymer matrix	Agro-industrial waste/additive	Manufacturing method	Results/observation	References
			moisture content at low sawdust contents (16.7 wt %) and was independent of the moisture content at higher sawdust contents (28.6 and 37.5 wt%). A decrease in decomposition temperature with an increase in polyene content was evidenced with increasing moisture content.	
PVC	<ul style="list-style-type: none"> <li>• 25 and 50% bamboo (<i>Bambusa vulgaris</i>) particle</li> <li>• Lubricant melting as additives</li> </ul>	Blending with hot-cool mixing, granulation via counter-rotating extrusion, consolidation by compression molding	It was found that the thermal stability of composites depended on the particle size/loading and the amount of lubricant used for processing. The oxidative induction time revealed that processing with bamboo particles does not contribute to degradation of PVC composites	Bahari et al. (2017)

thermoset composites include injection molding, compression molding, liquid molding (RTM), hand lay-up and spraying, filament winding, and pultrusion (Yashas Gowda et al. 2018). The manufacturing of thermosetting composites requires the mixing of dispersed phase and matrix at high temperatures, above the reaction temperature of the polymeric matrix.

### 18.3.2.1 Epoxy Resin-Based Biocomposites

Epoxy polymers, used as matrix resins for composites, have applications in electronics, aerospace, adhesives, insulators, and metal coatings owing to their desirable



**Table 18.5** Thermoplastic composites with PA matrices

Polymer matrix	Agro-industrial waste/additive	Manufacturing method	Results/ observation	References
PA 6, PA 6,6 (Technyl A302 V25), PA 6,6 (Technyl A502)	– 20 and 30% flax ( <i>Linum usitatissimum</i> ) and jute ( <i>Corchorus capsularis</i> )	Mixing in an internal mixer with two Banbury-like rotors, then injection molded	– Natural fibers were effective in reinforcing high-performance plastics such as polyamide 6 and polyamide 6,6 – An increase in both strength and modulus has been observed for all the fibers with respect to the unreinforced matrices	de Arcaya et al. (2009)
PA 6	– Flax fiber fabric and Kraft pulp cellulose – C20, a bifunctional activator – Aminopropyl-triethoxysilane (APS) and NaOH—for the surface modification of fibers	Anionic in situ ring-opening polymerization (ROP) using a special vacuum-assisted resin infusion process	– A pretreatment of the fibers was necessary in order to minimize the severe intrinsic inhibitive effect of cellulosic fibers on the ROP reaction – Optimum mechanical properties were obtained at a polymerization temperature of 150 °C	Kashani Rahimi and Otaigbe (2019)
PA 6	Recycled woven jute coffee bags (thickness of approx. 0.5 mm) coated with flexible epoxy resin	Compression molding	– The flexible epoxy coating (5 wt%) was able to improve the thermal degradation resistances – Flexural properties improved – SEM revealed strong fiber/matrix interfacial bonding for both coated and uncoated jute fabric composites	Thitithanasarn et al. (2012)
PA 6	– Up to 20 wt.% mixture of flax,	Melt compounding,	– Minor changes in the thermal	Kiziltas et al. (2016)

(continued)

**Table 18.5** (continued)

Polymer matrix	Agro-industrial waste/additive	Manufacturing method	Results/ observation	References
	kenaf, and hemp fibers	followed by injection molding	properties of the composites – The storage modulus ( $e'$ ) increased as the natural fiber content increased	
Anionic polyamide-6 (APA-6)	<ul style="list-style-type: none"> <li>• Textile-ramie fibers</li> <li>• Initiator—caprolactam magnesium bromide</li> </ul>	Reactive processing	<ul style="list-style-type: none"> <li>– Severe polymerization-inhibition and fiber-discoloration phenomena occurred</li> <li>– Initiator caprolactam magnesium bromide significantly suppressed inhibition and discoloration</li> </ul>	Kan et al. (2015)
PA 6	– 30 wt. % raw and treated pineapple leaf fibers	Compounding by twin-screw extruder, followed by injection molding	<ul style="list-style-type: none"> <li>– Treated fibers had superior interfacial adhesion over the untreated ones</li> <li>– Improved mechanical and thermal properties</li> </ul>	Panyasart et al. (2014)
Co-polyamide 6/6.6/6.10	• 30–70 wt.% wood chips	Extrusion and hot pressing	<ul style="list-style-type: none"> <li>– The tensile and bending strength increased by an increase in wood content, compared to wood-polyolefin composite materials</li> <li>– The thermal stability of composites was superior to that of wood</li> </ul>	Zanoaga et al. (2004)

properties including superior thermal stability, low shrinkage, high stiffness, and excellent chemical resistance to environmental degradation. The poor mechanical properties and fragility of the epoxy matrix, as well as longer cure schedules and high monomer cost, have restricted its use in the automotive industry (Mittal et al.

**Table 18.6** Thermoplastic composites with polymer blends matrices

Polymer matrix	Agro-industrial waste/additive	Manufacturing method	Results/observation	References
<ul style="list-style-type: none"> <li>• Recycled HDPE (rHDPE)</li> <li>• Polyethylene terephthalate (rPET)</li> </ul>	<ul style="list-style-type: none"> <li>• 75:25% rHDPE/rPET</li> <li>• 40, 50, 60, 70, and 80 wt.% rice husk</li> <li>• MAPE as a coupling agent</li> <li>• EGMA as a compatibilizer</li> </ul>	Extrusion process and compression molded	Mechanical properties significantly improved with the increasing amount of filler.	Ghani et al. (2014)
<ul style="list-style-type: none"> <li>• Recycled polyethylene (rPE)</li> <li>• Recycled copolyamide 6/6.6/6.10 (r-coPA)</li> </ul>	<ul style="list-style-type: none"> <li>• 3–70% wood chips</li> </ul>	Extrusion and compression molding	SEM confirmed the close contact between the wood and polymer matrix at their interface, which was evidence of good polymer–wood adhesion interactions.	Mamunya et al. (2006)
<ul style="list-style-type: none"> <li>• Recycled polyethylene (rPE)</li> <li>• Recycled copolyamide 6/6.6/6.10 (r-coPA)</li> </ul>	<ul style="list-style-type: none"> <li>• 60% wood chips</li> </ul>	Extrusion and compression molding	Improved bending strength, a better dispersion of the wood chips in matrix.	Zanoaga et al. (2016)
<ul style="list-style-type: none"> <li>• PVC</li> <li>• Thermoplastic polyurethane (TPU)</li> </ul>	<ul style="list-style-type: none"> <li>• 20, 30, and 40 wt.% kenaf (<i>Hibiscus cannabinus</i>) bast fiber</li> </ul>	Melt mixing	<ul style="list-style-type: none"> <li>– Lower tensile strength and strain, and impact strength along with increased fiber content</li> <li>– Tensile modulus increased with the increased fiber content</li> <li>– High-impact strength was observed for 40% fiber content</li> </ul>	El-Shekeil et al. (2014)

2018). Epoxy is a thermoset typically produced by reacting epichlorohydrin and a bis(epoxide) monomer, such as diglycidyl ether bisphenol A, with hardeners like anhydrides or amines, usually triethylenetetramine (Rahimi and Garcia 2017). Advanced composite materials with unique properties, cost effectiveness, and environment-friendly nature have been prepared using agro-industrial waste as reinforcement in epoxy matrix as an alternative to wood- and plastic-based components. Introducing coir pith and rice husk hybrid particulates obtained from waste agricultural residues into epoxy matrix produced customer value-added products (Prithvirajan et al. 2015). These composites, prepared by compression molding technique and particulate weight content ranging from 10 to 50%, exhibited

considerably increased mechanical properties due to the attractive physical and mechanical properties of bioparticulates.

The microstructural, physical, mechanical, and thermal properties of epoxy polymer composites in the presence of oil palm boiler ash (OPBA) as reinforcement were studied using different filler loadings (10–50 wt.%) and different particle sizes (50–150 $\mu\text{m}$ ) (Rizal et al. 2020). The experimental results showed that the incorporation of OPBA (53 wt.% silica content) in composites improved the physical, mechanical, and thermal properties of the epoxy matrix with maximum values recorded at 30 wt.% loading and size of 50 $\mu\text{m}$ . The utilization of this agro-industrial waste as filler in the structure of the composite material would benefit the palm oil industry by reducing waste load disposed to landfills and solid waste disposal cost and enhancing sustainability.

Particles of wood waste of *Pinus* sp. and teak (*Tectona grandis*) species as particleboard reinforcement phase and residual ink resin-based epoxy were applied to produce and to add value to new composites. The companies can use these panels for their economic and environmental advantages instead of disposing them off (Souza et al. 2018). Epoxy-based ink-waste melts at elevated temperatures and works as a possible binder in the consolidation of wood panels. As a consequence, these epoxy resins can be used as appropriate adhesives to reduce the demand of formaldehyde-based resins (urea-formaldehyde, melamine-formaldehyde, etc.) resulting in positive environmental and social aspects by decreasing in air releases of free formaldehyde in the production of panels. Economic benefits, especially for suppliers of wood waste regarding energy consumption for processing wood, can be gained in the production of panels from particles of wood waste of *Pinus* sp. and teak (*Tectona grandis*) species. Parameters, such as low-, medium-, and high-density panel classifications; epoxy resin content (20, 30, and 40%); and mixture of *Pinus* and teak wood waste particles (0, 25, 50, 75, 100%) were varied to manufacture and test the resulted particleboards. High density (up to 0.80 g/cm<sup>3</sup>), 30–40% epoxy-based ink-waste adhesive, and 50–100% teak wood particles promoted a satisfactory performance in terms of physical and mechanical properties.

### 18.3.2.2 Polyurethane-Based Biocomposites

Among various applications, such as fibers and fabrics, adhesives, and sealants, rigid and flexible foams, coatings, elastomers, and polyurethanes are also used as polymer matrix in composites. Polyurethane-based composites are widely used as components, such as caravans, bumpers, covers, flaps, rear ends, hoods, and roof modules for commercial vehicles in automotive engineering due to their low-density, superior mechanical properties, and easy processability (Abdel-Hamid et al. 2019). Polyurethane composites present superior properties such as abrasion resistance, damping ability, high flexibility, and high elongation at breaking point, good weathering resistance and weathering durability, anti-aging, high impact strength, biostability, and low-temperature flexibility. Their applications are restricted by some drawbacks, such as high flammability, poor adhesion to metal surfaces, poor electrical and thermal conductivity, poor tensile strength, and thermal stability. The composites technology, in which particulate fillers, such as

nanoparticles and natural fibers, are added to the polymers, may provide a good method to produce polyurethane hybrid nanocomposites and to solve the above-mentioned problems (Soundhar et al. 2019).

Polyurethanes are synthetic resins or plastics mainly synthesized by a polyaddition reaction between polyols and polyisocyanates. Vegetable oils such as soybean, palm oil, linseed oil, sunflower oil, and castor oil are renewable materials intensively employed in formulating polyurethane thermosets due to their good availability and high degree of functionalization possibility in manufacturing (Furtwengler and Avérous 2018). Several studies reporting the use of polyurethane resin as a binder for particleboards proved that the resulted materials meet the requirements of the international standards for physical and mechanical properties (Gava et al. 2015; Battistelle et al. 2016; Garzón et al. 2016; Varanda et al. 2018). The production technology of particleboards from alternative raw materials emerged as an interesting strategy to replace wood and dampen the deforestation. Introducing lignocellulosic wastes, such as corn cob (Akinoyemi et al. 2016), flax and hemp (Sam-Brew and Smith 2015), topinambour and cup-plant (Klímek et al. 2016), poppy husks (Keskin et al. 2015), sorghum (Khazaian et al. 2015), coconut and sugarcane bagasse (Fiorelli et al. 2018), and bamboo and sugarcane bagasse (Nakanishi et al. 2018) to produce particleboards could be an economic, social, and environmental strategy. These panels are typically manufactured from wood particles bonded by an adhesive or other synthetic binder, and the resulted aggregation is hot pressed till the curing of the resin.

The physical, mechanical, thermal, and aging properties of multilayer particleboards produced with green coconut (one inner layer, IL) and sugarcane bagasse fibers (two outer layers, OL) bonded with castor oil polyurethane resin were assessed (Fiorelli et al. 2019). The polyurethane resin was the bi-component castor oil polyurethane (50% polyol and 50% isocyanate), and its amount (by mass) for the OL and IL was 15% and 12%, respectively. Variations in the density profile across the thickness of the particleboards were evidenced by X-ray densitometry. Smaller fiber size and higher resin content led to a densification of the OL of the particleboards. All the particleboards survived a fatigue life of 40,000 cycles at stress levels equal to 25% of the breaking load as the fatigue tests indicated. Degradation of the particleboards under accelerated aging test indicated that the exposure conditions (temperature, humidity, UV irradiation) caused permanent damage to the physical (water absorption) and mechanical (tensile strength) properties of the materials.

Particleboards produced from sugarcane bagasse (*Saccharum officinarum*), jute (*Corchorus capsularis*), and curaua (*Ananas erectifolius*) fibers and their thermal, physical, and mechanical properties and microstructural characteristics were investigated (Fiorelli et al. 2018). The resin was the bi-component castor oil polyurethane adhesive prepared from 50% polyol and 50% isocyanate. By comparison with conventional wood particleboards, similar or superior physical and mechanical properties were observed when sugarcane bagasse (OL) and jute and curaua (IL) vegetable fibers of lower density were used as reinforcements for the multilayer particleboards.

The particleboards using vegetable fibers had higher values of their mechanical properties than those of the panels using only sugarcane bagasse with the same density, but the presence of the vegetable fiber did not influence the thermal conductivity. Addition of jute and curaua fibers induced higher porosity in the IL panels due to the geometry of the particle with a high value of aspect ratio ( $L/D$ ). Although a higher porosity contributes to higher water absorption and lower values of internal bond, introducing jute and curaua fibers did not weak the particleboards. Particleboards containing only sugarcane bagasse and only jute fibers presented no significant variations in their physical and mechanical properties when subjected to accelerated aging tests.

Balsa wood (*Ochroma pyramidale*) waste and castor oil polyurethane resin in 11 and 15% content were used in the production of oriented strand boards (OSB) with different densities ( $300$  and  $400 \text{ kg/m}^3$ ) and 10 mm thickness, and their thermo-physical-mechanical properties were evaluated (Barbirato et al. 2020). OSB panels agglomerated with 15% of castor oil polyurethane resin and a density of  $400 \text{ kg/m}^3$  had better physical and mechanical properties compared to the other formulations having lower density or lower resin content. Concerning the thermal behavior, all the treatments proved their potential in applications that require insulating properties. Modular panels made of reforested wood (*Eucalyptus* sp.), sugarcane bagasse, and bicomponent polyurethane resin based on castor oil were used in lateral closing for construction of crowding pens and chutes for cattle handling facilities (de Lucca et al. 2015). These modular panels, evaluated by the soft body impact testing, had physical and mechanical properties demanded by the normative documents for structural use. As a consequence, they can be used in cattle handling facilities because they showed structural performance necessary to withstand impact loads demanded by their usage.

Medium-density particleboard (MDP) panels were produced from coconut husk and *Pinus oocarpa* wood, and their properties (internal bond, the modulus of rupture and elastic modulus in static bending, moisture content, thickness swelling, and water absorption after water immersion for 2 and 24 h) were investigated (Narciso et al. 2020). *Pinus oocarpa* substituted with various content of coconut husk (25, 50, 75, and 100%), and 11% and 7% urea-formaldehyde adhesive (based on the weight of the particles) on the faces and in the core, respectively, were used to produce panels with a nominal density of  $650 \text{ kg/m}^3$  and relation face/core/face ratio of 20:60:20. Water absorption properties (screw holding and thickness swelling) increased after 2 and 24 h, and mechanical properties (modulus of rupture, modulus of elasticity in static bending and internal bond) decreased by increasing the amount of coconut husk in the MDP panels. These experimental results met the values determined by standards, being allowed to state that it was possible to produce MDP panels only with coconut fiber.

New particleboards were produced from sugarcane bagasse in two different fiber lengths (5 and 8 mm) of the same density ( $1.0 \text{ g/cm}^3$ ) as a raw material and castor oil-based two-component polyurethane adhesive (Fiorelli et al. 2016). Their properties (density, thickness swell, absorption, modulus of elasticity, modulus of rupture in static bending and internal bond) were evaluated. Based on the fiber

length, a significant difference between particleboards was revealed. The spaces between particles are filled with polyurethane adhesive, as evidenced by scanning electron microscopy, which contributed to improve the physical and mechanical properties of these particleboards. Waterproofed particleboards could be used in moist environments, as proved by a durability assessment based on accelerated aging tests.

Particleboards using industrial sawmill waste from tropical hardwoods (a mix of species) and sugarcane bagasse with castor oil-based bicomponent polyurethane resin were fabricated, and the influence of the incorporation of sugarcane bagasse (0, 10, 20, 30, 40, and 50%) on the physical and mechanical properties of the composites was evaluated (Yano et al. 2020). Some of the particleboards met standardized requirements, and the formulation 50% sawdust and 50% bagasse showed better performance, indicating the possibility for indoor use in dry conditions. The addition of sugarcane bagasse increased dimensional stability of particleboards when compared with panels manufactured with timber residue.

### 18.3.2.3 Amino Resin-Based Biocomposites

Amino resins are thermosetting polymers largely used in the furniture and construction industry; in coatings, laminates, and molding compounds; and in the production of wood-based panels, such as particleboards, fiberboards, and plywood. Urea-formaldehyde (UF), melamine-formaldehyde (MF), and melamine-urea-formaldehyde (MUF) are the main commercially used amino resins. Some other formaldehyde derivatives, such as phenol-formaldehyde (PF), tannin-formaldehyde (TF) and phenol-resorcinol-formaldehyde (PRF), and polymeric diphenylmethane diisocyanate (pMDI) are also used in manufacturing wood-based panels (Silva et al. 2015).

Urea-formaldehyde resin is widely used in the composites industry, especially in the forest products industry in the production of particleboard, medium-density fiberboard, and hardwood plywood, due to its special advantages, such as hardness, low flammability, noticeable homogeneity, good thermal properties, absence of color in the cured polymer, and adaptability of the resin to a variety of curing conditions. The main disadvantage of UF resin is its hydrolytic degradation in the presence of moisture and/or acids, which can be overcome by introducing other components like melamine into the UF resin molecules. MUF resin can replace UF resin especially to improve the properties of wood-based panels under high humidity conditions, because of its lower contribution to photochemical oxidation, environment, and human toxicity. By increasing melamine content in the MUF resin, formaldehyde air emission decreases. Free formaldehyde emission, one of the key environmental problems of formaldehyde-based resins, takes place from residual formaldehyde in the resin, during resin manufacture (when formaldehyde is generated by condensation reaction), and by hydrolytic degradation of cured resin during wood panel production. Free formaldehyde emissions are toxic to human health and potentially carcinogenic. There is a global trend toward biodegradable, non-pollutant, and renewable products. Therefore, numerous studies on finding new

products to replace the above adhesives, such as castor oil-based adhesive, have been reported.

Waste biomass obtained after orchard pruning (plum and apple tree branches) was used to produce three-layered composite materials consisting in wood-chips bonded by UF resin (Kowaluk et al. 2020). The physical (thickness swelling, water absorption) and mechanical (internal bonding strength, screw withdrawal resistance, density profile and machining properties during milling and drilling) properties of the resulted particleboards were investigated. Modulus of rupture and modulus of elasticity fulfill the mechanical requirements for panels of furniture production and interior equipment purposes. A good performance of the composite materials elaborated from waste biomass expressed by the experimental results offers a potential revalorization of the two undervalued by-products.

A core layer made of mixture sugar beet pulp (SBP) and industrial wood particles aggregated with (UF) resin and a hardener (a 10% water solution of  $\text{NH}_4\text{Cl}$ ) were used for production of three-layer particleboards (Borysiuk et al. 2019). Physical (density and density profile, thickness swelling, water absorption) and mechanical (modulus of rupture, modulus of elasticity, internal bond, screw holding) properties of the boards were studied. Investigation of the three-layer particleboards containing various proportions of SBP (10, 20, 30, 50, and 100%) in the core glued with UF resin showed that an increase in SBP content resulted in a decrease of modulus of rupture, modulus of elasticity, and screw holding and preserved the internal bond. An increase in SBP content in the core modified the density profile alignment and increased absorption and swelling of boards. Boards with 30% SBP met the requirements for furniture boards used in dry conditions.

The possibility of using agricultural residues (hazelnut husk, peanut hull, cotton stalk, corn stalk and licorice root) by using UF adhesives in manufacturing three-layer particleboards was investigated (Güler 2015). The UF resin was applied 8–9% for the core layer and 10–11% for the face layers based on oven dry weight. All the produced panels tested for mechanical (modulus of rupture, modulus of elasticity, and internal bond strength) properties complied with the minimum requirements in the standards for the general grade particleboards with the exception of thickness swelling and water uptake. Cup-plant (*Silphium perfoliatum* L.), sunflower (*Helianthus annuus* L.), and topinambour (*Helianthus tuberosus* L.) were used as raw materials for particleboards produced at a conventional density of  $600 \text{ kg/m}^3$  (Klímek et al. 2016). Particleboards were glued with two different adhesives, pMDI and UF resins. It was shown that the obtained particleboards had acceptable performances, though some properties (modulus of rupture, thickness swelling, and water absorption) were below those obtained from conventional spruce particleboards. Even so, the requirements for general purpose particleboards used in dry conditions were met and the pMDI-bonded particleboards produced from agricultural residues were found to be a feasible alternative to conventional UF-bonded particleboards made from spruce.

The potential of poppy (*Papaver somniferum* Linnaeus) husk to manufacture wood-based particleboards was investigated, and the results claimed that it was possible to produce three layers of particleboards from poppy husk and pine wood



using UF adhesive (Keskin et al. 2015). Particleboards were produced using poppy husk (up to 25%), 8% of UF resin for the core layer, and 10% for the outer layer, and 1% ammonium chloride was added to harden the adhesive. The physical and mechanical properties, formaldehyde emission, and limitation of oxygen index analysis met the requirements for interior fitments to use under dry conditions in the normative documents (EN 312). Formaldehyde emission is reduced and the feature of fire retardant for the manufactured particleboards is increased because of high limitation of oxygen index.

*Miscanthus x giganteus* stalks were studied as a possible replacement for wood in particleboards using 4 and 6% methylene diphenyl diisocyanate resin (Klímek et al. 2018). Modulus of rupture, modulus of elasticity, and internal bonding strength had lower values than those of spruce-made particleboards. Particleboards from *Miscanthus* showed collapsed cell regions in the soft parenchyma, with no obvious adhesive failures after internal bonding testing, suggesting a connection with the reduced mechanical properties. In contrast, particleboards made with spruce revealed much smoother fracture surfaces with structural failures running through cell walls and possibly also through gluelines. The *Miscanthus*-made particleboards exhibited higher thickness swelling, but lower water absorption in comparison with spruce-made particleboards. For *Miscanthus*, no effects of higher pMDI adhesive dosages on modulus of rupture, modulus of elasticity, and internal bonding strength were observed.

The efficiency of some particleboards manufactured using particles from sugarcane bagasse and eucalyptus residues of the pulp industry was investigated using castor oil-based polyurethane resin (PU-Castor) and UF resin (Sugahara et al. 2019).

When 60% wood particles and 40% bagasse and a 10% adhesive dose based on the dry mass of particles composed the particleboards, their physical (apparent density, moisture content, thickness swelling after 24 h) and mechanical (modulus of rupture, modulus of elasticity to static bending, and internal bond strength) properties were investigated. The experimental results proved the potential of eucalyptus and sugarcane bagasse residues in the production of high-density particleboards. Greater efficiency showed the panels produced with PU-Castor, and their physical and mechanical properties met the requirements necessary for their use in dry conditions.

Rice husk without any further treatment was processed into particleboards using PF resin as binder (Ciannamea et al. 2017). Processing parameters, pressure and binder content on the density, water absorption, thickness swelling, modulus of rupture, and modulus of elasticity were investigated. Particleboards with 11% of binder content met the minimum requirements of modulus of rupture and modulus of elasticity recommended for commercial use, while particleboards with 14% binder content also accomplished the requirements for industrial use, finding a resourceful use for this agricultural waste.

## 18.4 Multicomponent Polymer Systems Comprising Biopolymers from Renewable Resources and Agro-Industrial Waste

The society generates large amount of waste—both non-degradable (e.g., petroleum-derived) and degradable (e.g., from agriculture and food industry). A sustainable approach to decrease the resulted amount of waste may include all aspects related to the manufacture of bio-derived products from producing chemicals through complex biomass capitalization to introducing feasible polymerization strategies and further assembly into bioplastics with environmental friendliness and whole recyclability features. Up-to-date information (De Corato et al. 2018) emphasized that the issues of utilization and application of integrated biorefinery from remaining parts resulted from agricultural and industrial processes and agro-waste are somewhat underrepresented and they require additional attention in the near future. Among renewable biomass, the first remaining parts are produced in the plains, while the indirect wastes resulted from crop processing. Such agricultural residues, as well as food waste, may be transformed for varied and valuable chemical production by implementing suitable biorefining strategies (De Corato et al. 2018; Pleissner et al. 2017).

A durable progress in society is also attained through designing eco-friendly and versatile plastic materials. Furthermore, it is essential that such materials be recyclable to a large extent, being produced based on a lasting use of the both fossil and green carbon sources. For a long time, the plastic materials derived from petroleum and bio-derived sources will coexist and bring an outstanding contribution in the creation of viable and competitive products, thus fulfilling the increasing demands in the contemporary communities (Mülhaupt 2013) where polymer materials are designed in order to exhibit stiffness, softness, rubber behavior, conductance, or insulation properties, as well as transparency or opacity, permeability or impermeability, and stability or biodegradability. The significant potential afforded by the polymers derived from agricultural sources mainly those that are biodegradable like starch was from a long time established (Averous and Halley 2009). The influence of sustainable starch materials on the macro- or nano- structural level and further processing, thermo-mechanical behavior, and efficiency of plasticized starch-based materials were previously presented in a comprehensive review (Averous and Halley 2009).

Polymers from agro-resources are obtained mostly from plant species and have compostable and renewability features, at the same time manifesting some ordinary behavior, such as the high water affinity. Majority of such polymers can be engineered by a direct route as fillers, either through plasticization or chemical modification. Accurately, polyoses, proteic compounds (e.g., gluten or zein), and aromatic polyphenolic polymer, namely, lignin, are such polymers. The available in large quantities class of polymers from agro-resources is represented by polyoses, with various provenience sources and chemical structural features, such as cellulose, chitin, and starch. Biopolymers and some of their applications are schematically presented in Fig. 18.3. Some biopolymers, namely, starch, PLA, and lignin, used as



**Fig. 18.3** Biopolymers from agro-industrial resources and some of their applications

matrix in composites with agro-industrial waste reinforcements will be presented in the following sections.

#### 18.4.1 Starch

Starch, a relatively low-cost polyose obtained from renewable sources, was studied to a large extent to be used as an effective alternative for plastic materials derived from petroleum sources mainly as thermoplastic starch (TPS) which results under high temperature and shear conditions (Averous and Halley 2009; Pushpadass et al. 2009; Chang et al. 2010). Its chemical structure is formed by amylose, a linear component, and amylopectin, a highly branched component, with crystalline and amorphous regions in alternating layers, their ratio significantly influencing the properties of the resulted bio-based materials. Due to their better properties (increased thermal stability, superior mechanical strength, reduced water affinity),

the amylose-rich materials are usually preferred than amylopectin-rich materials. There was a previous study (Pang et al. 2014) which investigated the interesting performance behavior of the thermoplastic starch/polyolefin mixtures where starch is derived from agricultural waste presenting different ratios for the structural components (amylose/amylopectin), the amylose content being lower than in a normal pristine starch. The characteristics of starch, like high temperature value for glass transition, strong inter- and intra-molecular hydrogen bonding, reduced water resistance, and reduced flowing ability in granular form, make the processing methodology to be difficult which therefore determines a limited applications area. All these above-mentioned drawbacks can be solved through suitable approaches such as addition of plasticizers, blending with polymers and nanofillers, chemical modification, or an appropriate combination of the former listed (Mekonnen et al. 2013; Teacă and Bodîrlău 2015; Bodîrlău et al. 2014; Teacă et al. 2014; Xie et al. 2014).

Plasticizers like water, glycerol, sorbitol, fructose, mannose, glycol, and fatty acids (some of them even in combination like water and glycerol) are largely used for starch plasticization which is the most applied methodology for improving starch properties. In spite of the fact that the chemical modification can generate harmful by-products and implies relatively high costs, starch can be successfully modified for enhancing both its properties and polymer systems comprising the modified starch (Zamudio-Flores et al. 2010; Bodîrlău et al. 2012). The use of waste derived from agriculture practices as a source of starch for production of bioplastics may be advantageous for local farmers which can valorize such waste not only as nutrients for animal feeding. Furthermore, the lower cost of such agricultural waste enriched in starch is more conveniently in comparison with that of native sources of starch. In this context, significant starch amounts can be found in maize (corn), rice, and wheat, alongside with other sources (Gupta 2011) such as potatoes, sweet potatoes, cassava (known as yuca or manioc), banana (mainly peels), barley, buckwheat, rye, and taro (*Colocasia esculenta*, a tropical Asian plant grown primarily as a vegetable food for its edible corms which are very rich in starch). Another important source of starch is represented by sugar palm (*Arenga pinnata*) which alongside its fibers is largely considered as a significant agro-industrial waste in the agricultural industry from tropical Asian countries with potential use for biocomposites production (Sahari et al. 2013; Sanyang et al. 2016, 2017; Edhirej et al. 2017a, b; Ibrahim et al. 2020). The sugar palm tree can be an attractive source for a starch suitable for plasticization with beneficial effects upon properties of materials resulted through blending it with other polymers or fibers. The fibers extracted from the sugar palm trees contain a significant amount of cellulose as major chemical polymer component (66.49%) which confers them significant mechanical properties (Sanyang et al. 2016). In Table 18.7, some examples of applications of starch in multicomponent polymer systems comprising agro-industrial waste, where it is usually present as thermoplastic polymer matrix, are presented.

**Table 18.7** Use of starch in multicomponent polymer systems comprising agro-industrial waste

Type of starch	Type of agro-industrial waste	Type of polymer system and processing	References
Corn starch	By-products from cassava and ahipa starch extraction (cassava and ahipa peels and bagasse)	Corn starch plasticized using glycerol and mixed with agro-residues; thermo-compression	Versino et al. (2020)
Tapioca starch	Water hyacinth fibers (WHF)	Tapioca starch plasticized using glycerol and mixed with WHF's water solution Solution casting method	Abral et al. (2018), Asrofi et al. (2018)
Corn starch	Cornstalk Fiber (csf)	Corn starch thermoplasticized using fructose as plasticizer; solution casting and dehydration technique	Ibrahim et al. (2019)
Corn starch	Corn stalk Sugar palm fibers (spf)	Corn stalk/sugar palm fiber reinforced corn starch biopolymer hybrid composites Solution casting method	Ibrahim et al. (2020)
Sugar palm starch	Sugar palm fibers (SPF) <i>Eucheuma cottonii</i> seaweed waste	Seaweed/SPF reinforced Sugar palm starch/agar bio-hybrid composite; using glycerol as plasticizer; melt-mixing and hot pressing	Jumaidin et al. (2017a)
Sugar palm starch	<i>Eucheuma cottonii</i> seaweed waste	Thermoplastic sugar palm starch/agar (TPSA) blend containing seaweed waste as biofiller; using glycerol as plasticizer; melt-mixing and hot pressing	Jumaidin et al. (2017b)
Sugar palm starch	Woody fibers from sugar palm tree for starch extraction	Thermoplastic sugar palm starch/agar blends; using glycerol as plasticizer; melt-mixing and hot pressing	Jumaidin et al. (2016)
Commercial thermoplastic starch	Sugarcane bagasse fibers	Starch-based biopolymer reinforced with sugarcane bagasse fibers	Jiménez et al. (2016)
Sugar palm starch	Sugar palm fibers from which nanocrystalline cellulose was extracted using delignification and mercerization	Sugar palm nanocrystalline cellulose reinforced sugar palm starch bionanocomposites; sorbitol and glycerol as plasticizers; solution casting method	Ilyas et al. (2018)
Corn starch	Blueberry agro-industrial waste	Thermoplastic corn starch/blueberry powder films; using glycerol as plasticizer Solution casting method	Luchese et al. (2017)

(continued)

**Table 18.7** (continued)

Type of starch	Type of agro-industrial waste	Type of polymer system and processing	References
Cassava starch	Blueberry agro-industrial waste as pomace in powder form	Cassava starch films incorporated with blueberry pomace; using sorbitol as plasticizer; solution casting method	Luchese et al. (2018)
Corn starch	Oil palm frond (OPF) fiber	Oil palm fiber-reinforced thermoplastic starch composites; OPF pretreatment with ionic liquids; corn starch plasticization using glycerol as plasticizer; compression molding technique	Mahmood et al. (2017a, b)
Native cassava starch from cassava tubers	Sugar palm fibers	Sugar palm fibers reinforced thermoplastic cassava starch films; using fructose as plasticizer; solution casting method	Edhirej et al. (2017a, b)

### 18.4.2 Polylactic Acid (PLA)

*Polylactic acid (PLA)* is an aliphatic-type thermoplastic polyester, biodegradable, and recyclable, different than most thermoplastic polymers as it is derived from renewable resources, namely, agricultural residues, waste, and by-products like corn starch, tapioca (roots, chips or starch), or sugarcane (Mekonnen et al. 2013). From these sources, the lactic acid (LA) can be produced through suitable biotechnology routes as a platform chemical and precursor for production of biodegradable, biocompatible LA polymers (e.g., either by condensation of lactic acid or ring opening polymerization of lactide which is the diester of lactic acid). These polymers are very promising thermoplastic polymers mostly used for high-end applications (Drumright et al. 2000), ranging from technical textile fiber applications, furnishing, and agricultural products to pharmaceuticals and drug release systems as well as medical textile industry, namely, also biotextile, which include suture materials, implantable matrices, and organ reconstruction (Gupta et al. 2007). The sustainable use of polylactic acid in these applications is based not only on its biodegradability and its provenience from renewable resources but also on its ease processing and well behavior under technological conditions providing excellent properties with low expense.

A recent review (Djukić-Vuković et al. 2019) focused on the sustainable production of lactic acid using residues and wastes derived from agricultural practices through different methodologies considering all aspects involved. The characteristics of PLA-based polymer matrix can be accordingly modified through complex interrelations occurred when using varied fillers, both organic and

inorganic type, including by-products and wastes. Reinforcement and toughness effects can be thus determined by the rigidity of used particles and the structural changes in PLA macromolecular chain (Battegazzore et al. 2014a, b, c; Barczewski et al. 2020). Another interesting way to modify and develop enhanced characteristics in PLA for its use as an alternative sustainable biopolymer in applicative end-uses including automotive and electronics ones is addition of nanofillers—nanofibers and nanoparticles (Raquez et al. 2013)—in order to overcome the inherent shortcomings of PLA (reduced thermal stability, decreased crystallization rate), while properties such as flame retardancy, conductance, resistance against UV radiation, and microorganisms and the barrier ones are essential for many other end-use areas. Some examples for use of PLA in multicomponent polymer systems comprising agro-industrial waste are presented in Table 18.8.

### 18.4.3 Lignin

Lignin represents an aromatic biopolymer of polyphenolic type, constituent of the lignocellulosic materials, which is responsible for the mechanical resistance of plants being intertwined between the polysaccharide components, namely, cellulose and hemicelluloses, conferring rigidity to the plant cell walls. Lignin has the most complex structure as derived from oxidative coupling of p-hydroxycinnamoyl alcohols (monolignols) and related compounds. The processes for assembling structural units and their activation toward lignin polymerization are under strict biological control; even these occur via simple combinatorial chemistry pathways (Tobimatsu and Schuetz 2019). Comparatively with the use of cellulose and hemicelluloses, lignin is still the most under-utilized of the biopolymers from lignocellulosic biomass despite its large potential for generation of renewable fuels and value-added chemical compounds in relation with many progresses made in research areas such as bioengineering, biorefinery, and catalysis (Rinaldi et al. 2016).

Actual approaches for introducing lignin in latest designed materials focus on its combination with other polymer matrices, either natural or synthetic, through appropriate covalent modification and blending strategies. The significance of lignin in technology applications and focused attention on its sustainable blending and copolymerization strategies for use in a large applicative area were previously reviewed (Chung and Washburn 2013). For example, the blending of two different kinds of biopolymers derived from renewable resources, lignin, a tough polymer matrix and biodegradable starch, has great potential when considering the major drawback of the starch which exhibits a pronounced hydrophilic behavior in various environmental conditions. In this context, lignin confers additional features to starch matrix when result films and foams with better properties thus becoming potentially attractive for a broad applicative area. As example, starch-lignin blends with enhanced properties were obtained through use of chemical modification of starch polymer matrix and adequately its appropriate plasticization strategy (Spiridon et al. 2011).

**Table 18.8** Uses of PLA in multicomponent polymer system comprising agro-industrial waste

Types of polymer system based on PLA	Type of agro-industrial waste	Type of processing	References
Biocomposites	Hemp hurd (as chips and powder), alfalfa, and grape stem	Melt blending	Battegazzore et al. (2019)
Films	Water-soluble extracts from banana pseudo-stem as functional additives	Solution blending, casting, and thermo-compression method	Sango et al. (2019)
Biocomposites	Cellulose extracted from rice husk	Melt blending	Battegazzore et al. (2014d)
Fully bio-based and biodegradable composites	Almond shell flour	Maleinized linseed oil as a reactive compatibilizer during the extrusion process; melt compounding	Quiles-Carrillo et al. (2018)
Biocomposites	Bagasse agave tequilana fibers pretreated using alkali and enzymatic stages	Extrusion and press molding	Huerta-Cardoso et al. (2020)
Composites	Chestnut shell waste	Melt blending without hydrolytic degradation of the bio-polyester PLA Matrix	Barczewski et al. (2018)
Bio-nanocomposite films	Cellulose nanocrystals derived from coffee silverskin	Melt compounding: Extrusion and press molding	Sung et al. (2017)
Composites	Cork-wood hybrid Filler system	Melt compounding: Extrusion/injection molding process; evidencing the potential of using cork fillers to improve the dimension stability of composites in the presence of moisture	Andrzejewski et al. (2019a, b)
Composite films	Cellulose nanocrystals from waste cottonseed hulls	Solvent casting method; using maleic anhydride ma as both the compatilizer and crosslinker	Zhou et al. (2018)
Biocomposites	Algae waste (red, brown, and green) as filler	Melt mixing; algae can be used as filler in thermoplastic composites up to at least 40 wt% loading, since the stiffness of materials can be maintained	Bulota and Budtova (2015)
Composite films	Corn Stover organosolv lignin from biorefinery	Melt compounding: Extrusion and press molding	Gao (2017)
Composites	Coffee silverskin (CS) as natural filler being	Melt blending	Ghazvini (2018)

(continued)



**Table 18.8** (continued)

Types of polymer system based on PLA	Type of agro-industrial waste	Type of processing	References
	by-product of coffee roasting process		
Biocomposites	Rice husk as source of silica	A semi-industrial process of extrusion	Battegazzore et al. (2014c)
Natural filler-based composites	Cellulose fibers, wood sawdust, hazelnut shells, flax fibers, corn cob, and starch from the Italian agricultural production	Melt blending	Battegazzore et al. (2014a)
Biocomposites	Natural additives from hazelnut skin and cocoa by-products through a selective and serial extraction to be used as plasticizers for PLA, antioxidants for PP, and reinforcement fillers for both polymers	Melt blending	Battegazzore et al. (2014b)

An effective valorization of lignin implies an attentive consideration of both multiple upstream (e.g., bioengineering approaches, extraction methodology, conversion by catalysis) and downstream (e.g., lignin depolymerization and upgrading) processing phases. Such strategy is a prerequisite for achievement of a sustainable economy and to reduce the carbon footprint. There are many potential uses of lignin for high-value products ranging from fuels and chemical platforms to filler materials, namely, plant-derived plastics and composites (engineering plastics and thermoplastic elastomers, polymeric foams and membranes), as well as low cost carbon fibers (Azadi et al. 2013; Ragauskas et al. 2014; Rinaldi et al. 2016; Becker and Wittmann 2019; Collins et al. 2019). Some examples for the use of lignin in multicomponent polymer systems comprising agro-industrial waste are illustrated in Table 18.9.

## 18.5 Conclusion and Future Trends

The use of composite materials based on agro-industrial waste has been expanding in the recent decades. This trend is encouraged by industry and agriculture, on one hand, because it helps reduce the demand of the fossil resources as raw materials, limits the waste disposal, and supports the recovery and re-/up-cycling of waste, contributing thus to the joint effort to protect the environment. On another hand, the properties of these composites are satisfactory, although not at the same level of performance as typical plastic composites, but their lightweight and low cost, in addition to their other advantages, make them highly attractive for the consumers. For example, composites panels made of polymer matrices and various types of

**Table 18.9** Use of lignin in multicomponent polymer systems comprising agro-industrial waste

Type of polymer system based on lignin	Type of agro-industrial waste	Type of processing	References
Thermoplastic cassava starch-based composites	Kraft lignin and esterified lignin from	Mixing, compression molding	Kaewtatip and Thongmee (2013)
Chemically modified starch microparticles (using malic acid)/ glycerol plasticized corn starch/lignin filler composite films	Industrial Kraft lignin extracted by precipitation in the lignoboost process	Solution casting	Bodirlău et al. (2013)
Chemically modified starch microparticles (using adipic acid)/ glycerol plasticized corn starch/lignin filler composite films	Industrial Kraft lignin extracted by precipitation in the lignoboost process	Solution casting	Spiridon et al. (2011)
Blends with polylactic acid	Lignin extracted from almond shells by organosolv process and acetylated	Extrusion	Gordobil et al. (2014)
Lignin–formaldehyde matrices for biobased composites reinforced with sisal fibers	Lignin isolated through an industrial organosolv acid hydrolysis process from sugarcane bagasse (Dedini rapid hydrolysis DHR)	Mixing and compression molding (the presence of lignin in both fiber and matrix Greatly improved adhesion at the fiber–matrix interface)	Ramires et al. (2010)
Fiberboards	Sugarcane bagasse Lignophenolic prepolymer and fibers	Mixing, compression molding	Hoareau et al. (2006)

agro-industrial waste have been successfully employed in structural applications because they showed adequate mechanical strength and dimensional stability, good screw holding capacity, and low water sorption.

Agro-industrial wastes such as rice husk, bagasse, palm oil, wood chips and flour, cereal straws, eggshells and seashells, feathers, plant stalks, natural fibers, etc. are able to provide much higher strength per unit mass than some inorganic fillers (carbon black, calcium carbonate, talc, zinc oxide). They also content a high amount of silica which may have a negative effect on the processing equipment. Since agriculture waste depends on localization and season, agriculture practice and harvesting, transport, storage, and processing, it is easy to understand that their characteristics vary within large limits for the same species. In order to counterbalance these drawbacks, the other elements of the multicomponent systems must be wisely chosen: the nature and type of polymer matrix; type of pretreatment,

functionalization method, or compatibilization strategy; number and nature of additives, in correlation with the application; processing methods and parameters; and type of equipment. As shown in this chapter, agro-industrial waste was successfully used as reinforcement in various polymer matrices: synthetic thermoplastics and thermosets, and biopolymers. The thermoset biocomposites have significantly improved chemical and thermal stability, structural integrity, mechanical properties (strength, hardness), and moldability, but they cannot be melted, reshaped, recycled, and re-molded in order to be introduced in a new production cycle. By comparison, the thermoplastic biocomposites showed an enhanced processability, increased elastic properties, and a high degree of post-consumer recovery, due to the low melting point of the polymer matrix. As for the composites based on biopolymers (starch, lignin, PLA) as matrix, they are promising materials due to their high added-value and biodegradable character, despite their low rate of recovery.

The literature of the recent decades is abundant in reports on multicomponent polymer materials based on agro-industrial waste, with an emphasis on the variety of formulations and procedures. Unfortunately, only a small amount of data is effectively transferred to industry and applied, although constant progress is evident and encouraged. The wide range of inexpensive and locally available raw materials represent a potential leverage that may help industrial and individual consumers to switch their preference from classic plastic materials to the new bio-based materials as they can provide promising benefits and contribute to a sustainable development. Constant concerns over the climate changes and global food security have been motivating the development of bio-based materials. This trend may be regarded as a challenge to both academic and industry R&D scientists and policy makers, because manufacture of bio-based materials, including the valorization of agro-industrial waste, requires land and crops which are used less for food.

Nevertheless, future research must focus on the development of new high added-value products and methods, and on up-grading the existing materials, in close relation with the local availability of materials and processing facilities. The design of new procedures or hybrid methods may be approached simultaneously with the use of the already existing ones in various combinations. Another very important aspect, which has been so often neglected, is the efficient, low-cost production without generating secondary pollution (waste that cannot be further recycled). Still, this field of research is very active, so it is reasonable to expect new and exciting developments.

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