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# Production and Characterization of Hybrid Polymer Composites Based on Natural Fibers

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Additional information is available at the end of the chapter

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## Abstract

In this chapter, a review is made on the processing and properties of hybrid composites based on a polymer matrix and a blend of different natural (lignocellulosic) fibers. In particular, the processing methods are described and comparisons are made between the general properties with a focus on physical, mechanical and thermal properties. A discussion is presented on the effect of the polymer and fiber types, as well as reinforcement content. Properties improvement is also discussed using fiber surface treatment or the addition of coupling agents. Finally, auto-hybrid composites are presented with conditions leading to a positive deviation from the rule of hybrid mixture (RoHM) model.

**Keywords:** hybrid composites, polymer matrices, natural fibers, fiber concentration, mechanical properties

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## 1. Introduction

Composites are materials containing at least two constituents, each one with different chemical composition. Their combination provides a new material with better functional properties than each of the components separately [1].

The main component in the composite is the matrix, which can be a metal, ceramic or polymer, while the other part is a reinforcement which can be in particulate, laminate, short fiber or long fiber form [2]. Composite materials are widely used in construction, aerospace, aircraft, medicine, electrical and automotive industries [2–5]. Here, a focus is made on fiber reinforced composites made from a polymer matrix reinforced with fibers having a natural origin [6].

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2. Natural fibers

Natural fibers are biosourced materials extracted from plants (lignocellulosic) or animals [7]. Lignocellulosic fibers are produced by plants for which, on a dry basis, the cell walls are mainly composed of cellulose, with hemicelluloses, lignins, pectins and extractives in lower amounts. Chemical composition and distribution mostly depend on fiber source and varies within different parts even of the same type or family [7, 8]. According to their source, lignocellulosic fibers can be classified as bast fibers, leaf fibers, fruits-seeds fibers, grass-reed fibers and wood fibers [7, 9–12]. **Table 1** presents some examples of each category [13].

Fiber type	Characteristics	Examples
Bast	High cellulose content, flexible, obtained from plants phloem	Kenaf, hemp, flax
Seed	Fibers that have grown around seeds	Cotton, kapok
Fruit	Obtained from fruit shells	Coir, oil palm
Stalk	Cereal stalks byproducts	Wheat and corn straw
Grass	Obtained from grass plants	Bamboo, wild cane, esparto grass
Leaf	Obtained by decortication of plants leaves	Banana, sisal, pineapple, agave
Wood	Extracted from flowering and conifers trees	Maple, pine

**Table 1.** Lignocellulosic fibers classification [13].

Due to natural fibers’ strength, stiffness, availability, low cost, biodegradability and lower density (1.2–1.5 g/cm<sup>3</sup>) compared to synthetic fillers such as talc (2.5 g/cm<sup>3</sup>) and glass fiber (2.5 g/cm<sup>3</sup>) [14–16], they can be effectively used in lightweight composites production [8, 9, 17].

3. Natural fiber composites

Natural fiber composites are materials based on a polymer matrix reinforced with natural fibers [9]. The polymer matrix can be a thermoplastic or a thermoset, the main difference being that once thermoplastics are molded they can be remelted and reprocessed by applying heat and shear, while this is not the case for thermosets [14, 15]. But thermoset matrices generally provide higher rigidity and are more chemically stable. This is why they are more difficult to recycle. The main thermoset matrices used for natural fiber composite production are polyester, vinyl ester, phenolic, amino, derived ester and epoxy resins. Thermoset composites are commonly processed via resin transfer molding (RTM), sheet molding compound (SMC), pultrusion, vacuum-assisted resin transfer molding (VARTM) and hand lay-up. All these manufacturing processes do not need high pressure requirements. Another advantage of thermoset matrices is that fiber loading can be higher than for thermoplastics since the resin

is initially in a liquid form. So, lower viscosity improves fibers introduction and dispersion via different mixing equipment [18–22]. Fiber orientation as well as fiber content might improve mechanical properties in thermoset composites. Grass, leaf and bast fibers are more effective to increase the matrix mechanical properties, while surface treatment improves interfacial interactions. **Table 2** summarizes some work on natural fiber thermoset composites with their manufacturing process, fiber content, fiber treatments and fiber source, as well as the main results obtained from each work.

Matrix	Natural fiber source	Manufacturing process	Fiber content (%)	Fiber treatment	Mechanical properties					References
					E (GPa)	TS (MPa)	FM (GPa)	FS (MPa)	IS (J/m)	
Epoxy	Banana	Hand lay-up	10	NaOH solution	0.6–1.4	12.1–33.6	15–34	26–69	2–12	[23]
	Recycled cellulose	RTM	19, 28, 40, 46	–	–	–	0.5–5.5	60–140	5–22	[21]
	Flax	RTM	40–50	–	17.3–33.6	–	–	–	–	[19]
		Hand lay-up	50	–	8.6	–	–	–	–	[24]
		Compression molding and pultrusion	40	NaOH solution	2.7–32	50–283	8–27	0.4–4.1	–	[25]
	Oil palm	Compression molding	5, 10, 15, 20	NaOH solution	–	11–17	–	–	–	[26]
	Hemp	Hand lay-up	30	H <sub>2</sub> PO <sub>3</sub> solution NH <sub>4</sub> OH Geniosil GF-9 Toluene solution aminosilane	3–4.8	49.1–66.5	3–5.2	69–92.8	–	[27]
	Date palm	Hand lay-up	10	NaOH solution	1.5–2.5	10–40	–	–	–	[28]
	Sansevieria cylindrical leaf	Molding	1, 5, 7, 9	NaOH solution	–	98.3–114.9	–	17–26	–	[29]
Polyester	Jute	Hand lay-up	NA	–	–	–	–	–	3.8–4.1	[30]
	Macadamia nut shell	Hand lay-up	10, 20, 30, 40	–	–	–	4.1–4.6	26–38	–	[31]
	Flax	VARTM	20	–	15.3–20.3	188.6–230.7	2.1–2.3	16.3–17.5	–	[32]
	Curaua	RTM	0–40	–	–	–	0.1	–	20–190	[33]
	Wild cane grass	Hand lay-up	0–40	NaOH solution KMnO <sub>4</sub> solution	–	–	1.8–7	–	–	[34]
	Sisal	Mixing and compression molding	10, 20, 30, 40	NaOH solution	–	–	1.49–2.68	–	–	[35]
	Typha leaf	Compression molding	7.3, 10.3, 12.6	NaOH solution Sea water	–	–	3.5–6	25–70	–	[36]

Matrix	Natural fiber source	Manufacturing process	Fiber content (%)	Fiber treatment	Mechanical properties					References
					E (GPa)	TS (MPa)	FM (GPa)	FS (MPa)	IS (J/m)	
	Rice husk	Mixing and compression molding	57	GMAMAHSAH solutions	0.4–1.6	2.5–19	0.1–1.9	3–42	9.5–40	[22]
	Elephant grass	Hand lay-up	30.4, 31.3, 31.5	NaOH KMnO <sub>4</sub> solutions	0.6–2.2	31.5–118.1	–	–	–	[37]
	Bamboo	Mixing and compression molding	NA	H <sub>2</sub> O <sub>2</sub> +DTPA +Na <sub>2</sub> O <sub>3</sub> Si +NaOH solution, IEM +DBTDL	–	39–65	–	75–105	–	[38]
	Coir	Hand lay-up	NA	NaOH solutions	–	17.9–23.6	–	18.7–48	–	[39]
			10, 20, 30	–	–	10.6–15.6	–	25.9–38.5	25.6–161.9	[40]
Polyurethane	Kraft cellulose	Compression molding	5, 10, 15, 20	–	0–0.2	–	–	–	–	[41]
Phenolic	Bagasse	Compression molding	17.6	HClO <sub>2</sub> solution Furfuryl alcohol	–	–	–	–	17–28	[42]
	Curaua	Compression molding	17.6	HClO <sub>2</sub> solution Furfuryl alcohol	–	–	–	–	39–88	[42]
	Cellulose from eucalyptus	Molding	1, 3, 5, 7	NaOH solution, propyl-trimethoxy-silane	0.7–0.9	9.5–16.5	5.1–1.0	18.5–28.0	–	[43]
	Ramie	Compression molding	40.4	–	3.3, 1.2	72.3, 158	–	90–145	–	[44]
	Jute	Pultrusion	N/A	–	–	25–38	–	28–63	–	[45]
Vinyl ester	Bamboo	Compression molding	15	–	21.2–30.1	–	–	210–320	–	[46]
	Silk	Hand lay-up	0–15	–	0.9–1.3	40–71	–	–	–	[47]
	Cellulose	VARTM	20, 30, 40, 50	–	3–7	–	–	40–160	–	[20]
	Sisal	RTM	10, 15, 20, 25, 30	NaOH solution	1.7–2.9	38–75	2.1–4.5	75–180	–	[48]
	Kenaf	Pultrusion	40	–	9–12.5	135–145	1.6–1.9	150–190	–	[49]
	Pineapple leaf	Molding	20	NaOCl solution	–	–	1.9–3.9	68–119	19–105	[50]

E: Tensile modulus; TS: tensile strength; FM: flexural modulus; FS: flexural strength; IS: impact strength; GMA: glycidyl methacrylate; MAH: maleic anhydride; SAH: succinic anhydride; DTPA: diethylenetriaminepenta-acetic acid; IEM: isocyanatoethyl methacrylate; DBTDL: dibutyltin dilaurate.

**Table 2.** Mechanical and thermal properties of natural fiber composites based on thermoset matrices.

The most common thermoplastic matrices used for natural fiber composites production are the different grades of polypropylene (PP) and polyethylene (PE), as well as polycarbonate (PC), nylon (PA), polysulfones (PSU), polyethylene terephthalate (PET) and polystyrene (PS). More recently, biopolymers such as polylactic acid (PLA) have gained interest to produce 100% biosourced materials [51–55]. Typical manufacturing processes for these composites are extrusion, injection, calendering, compression molding and thermoforming. Some advantages of using thermoplastic matrices are their recyclability and the production can be continuous [56–61]. Depending on the matrix, fiber and additives content, fiber treatment and manufacturing process, the mechanical and thermal properties of these composites can be adjusted as presented in **Table 3**, with the main results obtained.

The main objective of adding natural fibers in polymer matrices is to increase mechanical properties regardless of polymer and fiber type [21, 26, 31, 40, 52, 54, 55, 61–68]. Since natural fibers have lower density (1.2–1.5 g/cm<sup>3</sup>) compared to synthetic/inorganic reinforcement such as glass fibers (2.5 g/cm<sup>3</sup>), lightweight composites can be produced [28, 69, 70]. Nevertheless, lignocellulosic fibers are hydrophilic and polar which causes some incompatibility with the most common polymer matrices which are hydrophobic and nonpolar. This effect leads to poor mechanical properties due to a lack of interfacial adhesion between the fibers and the matrix. Furthermore, the high amount of hydroxyl groups available on the fiber surface is increasing water absorption, even when inside a composite [65, 71, 72]. These problems can be resolved by modification of the fibers surface such as mercerization (treatment in sodium hydroxide solution to remove lignins and hemicellulose) with subsequent addition of coupling agents [22, 73–75]. There is also the possibility to combine thermomechanical refining with coupling agent addition [71, 72]. More recently, fiber treatment with a coupling agent in solution has been proposed [76].

Matrix	Fiber source	Processing	Fiber content (%)	Fiber surface treatment	Additive		Mechanical properties					TD (°C)	References
					CA	BA	E (MPa)	TS (MPa)	FM (MPa)	FS (MPa)	IS (J/m)		
HDPE	Flax	Injection molding	0, 15, 30–		–	ACA	220–470	14–24	500–1600	15–26	60–230	–	[66]
	Wood	Compression molding	0–40	Thermo-mechanical refining	MAPE	ACA	–	–	0.9–3.9	–	–	–	[72]
	Wood	Extrusion	20, 30, 40	–	MAPE	–	2300–2900	–	1900–3400	–	–	–	[56]
	Wood	Extrusion	50, 60, 70, 80	–	MAPE	–	3130–4600	11.1–30.2	2470–3370	25.0–58.8	–	–	[77]
	Wood	Injection molding	40	Ethanol and toluene extraction NaClO <sub>2</sub> treatment NaOH solution	MAPE	–	3570–4940	23.8–48	–	–	–	–	[78]

Matrix	Fiber source	Processing	Fiber content (%)	Fiber surface treatment	Additive		Mechanical properties					TD (°C)	References
					CA	BA	E (MPa)	TS (MPa)	FM (MPa)	FS (MPa)	IS (J/m)		
	Wood	Injection molding	25, 35, 45	–	–	–	1200–2000	18.5–27.5	1200–2700	27.5–43	–	–	[59]
	Oil palm	Compression molding	30, 40	–	MAPP	–	650–1050	10–15	–	–	–	–	[65]
	Hemp	Compression molding	0–40	–	–	ACA	–	–	1093–1634	18.8–23	–	–	[55]
	Agave	Injection molding	0–20	–	–	ACA	225–550	15–24	1–2.7	–	–	–	[79]
	Hemp	Compression molding	40	Thermo-mechanical refining	MAPE–MAH	–	–	–	2–2.6	–	–	–	[71]
	Argan nut shell	Injection molding	5, 10, 15, 20, 25	NaOH solution	–	–	1136–1795	27.2–29.3	–	–	–	–	[80]
	UHMWPE	Wood powder molding	0–30	–	–	–	195–280	–	650–1260	–	–	–	[67]
LMDPE	Agave	Rotomolding	5, 10, 15–	–	–	–	255–440	13–18.8	495–590	12.5–16.5	0.9–7.5	–	[81]
	Agave	Rotomolding	15	Solutions of: MAPE NaOH Aldehyde Acrylic acid Methyl methacrylate Silane	–	–	167–217	13–18	420–520	13–17.8	83.8–148.5	–	[76]
	Hemp	Injection molding	30	Solutions of: MAPE NaOH MAPE	MAPE–	–	241–668	13.1–17.9	–	–	–	–	[73]
LLDPE	Maple wood	Rotomolding	0–20	–	–	ACA	26–184	3–16.4	119–680	–	–	–	[52]
	Wood	Injection molding	47	–	MAPP	–	–	30.2	–	–	–	–	[82]
	Agave	Compression molding	0–40	Solutions of: NaOH MAPE	–	–	224–381	10–22	389–1027	14–31	123–260	–	[75]
PS	Agave	Compression molding	10, 20, 30	–	–	ACA	3345–4929	30–62	–	–	–	400	[83]
	Wood fiber	Extrusion	10, 20, 30, 40	MAPS	–	–	–	31–49	–	54–94.5	–	–	[84]
	Wood flour	Extrusion	10, 20, 30, 40	MAPS	–	–	–	31–41.5	–	55–68	–	–	
PP	Argan nut shell	Injection molding	0–30	–	SEBS–g-MA	–	1034–1593	26.5–30	–	–	–	339.4–350	[85]
	Flax	Compression molding	10, 20, 26, 30	–	MAPP–PPAA	–	1000–3200	–	–	–	–	–	[86]



Matrix	Fiber source	Processing	Fiber content (%)	Fiber surface treatment	Additive		Mechanical properties					TD (°C)	References	
					CA	BA	E (MPa)	TS (MPa)	FM (MPa)	FS (MPa)	IS (J/m)			
PLA	Abaca	Injection molding	10, 15, 20, 25	Benzene diazonium treatment, NaOH solution	–	–	800–2700	24.5–31	800–3100	43–55	22.5–50	–	[87]	
	Coir bagasse	Injection molding	5, 10, 15, 20, 25, 30	NaOH solution	–	–	1100–1700	27.5–34.7	1400–2000	35–53	–	–	[88]	
	Wood	Compression molding	10, 20, 30, 40	–	MAPP	–	600–1600	–	2100–2400	44–52	10–17	–	[89]	
	NNC	Compression molding	1	–	MAPP	–	450–663	32.3–39.1	1809–2238	–	–	–	[90]	
	Sisal	Injection molding	10, 20, 30	NaOH solution	MAPP	–	500–1100	23–28	–	–	–	363.2–434.5	[91]	
	Pine cone	Injection molding	5, 10, 15, 20, 25, 30	NaOH solution	SEBS-g-MA SBS	–	1020–1550	21–27.5	–	–	–	321–355	[92]	
	Wood cotton	Compression molding	10, 20, 30	–	MAPP	–	–	28–50	–	37–152	–	–	[93]	
	Flax	Injection molding	15, 25, 40	–	–	–	–	–	2500–6000	–	–	282–340	[54]	
	Maple wood	Injection molding	15, 25, 40	–	–	–	–	–	2400–5900	–	–	282.3–342.7	[62]	
	Maple wood	Injection molding	5, 10, 15, 20, 25	–	–	–	1250–1890	59.8–61.5	3650–5260	96.6–107	21.7–34.3	250–360	[94]	
Post consumer PP+HDPE	Wood	Injection molding	20, 30, 40, 50, 55, 60, 65	–	–	–	–	5270–10300	56.8–64.6	5400–1088	77–91.8	–	–	[58]
	Cotton	Injection molding	10, 20, 30, 40, 50	–	–	–	–	1260–2500	58.1–62.6	3690–8220	97.9–106.2	17.5–24.3	250–360	[94]
	Agave Coir Pine	Injection molding	10, 20, 30	–	–	–	–	1242–1865	43–60	2300–3110	55–96	30–49	–	[95]
	Wood flour	Compression molding	0–40	–	MAPP-MAPE	–	247–394	12.7–15.3	950–1889	–	38–65.6	–	–	[61]
Post consumer HDPE	Wood flour	Compression molding	0–40	–	POE-MAPP-MAPE	–	–	–	1073–1958	16.6–22.4	–	–	–	[96]
	Flax	Injection molding	30	–	MAPP-EO-g-MAH	–	608–579	–	3090–2921	–	–	–	–	[97]
					–	–	332–608	–	1114–3090	–	–	–	–	[98]
														[99]
Post consumer HDPE	Pine wood	Compression molding	30	–	MAPE-CAPE	–	–	21.4–30.6	–	–	–	341.3–342.4	[60]	



Matrix	Fiber source	Processing	Fiber content (%)	Fiber surface treatment	Additive		Mechanical properties					TD (°C)	References	
					CA	BA	E (MPa)	TS (MPa)	FM (MPa)	FS (MPa)	IS (J/m)			
Post consumer PP					TDM									
	Bagasse	Compression molding	30	–	MAPE–CAPE	–	22.3–36.1	–	–	–	–	348.5–353.3	[60]	
					TDM									
	Wood	Compression molding	50, 60	–	MAPE–	–	9–18	–	–	20–35	–	[100]		
	Wood	Extrusion	–	–	MAPP–	450–490	27.3–29.8	2230–2940	43–51	–	285–499	[101]		
	Oil palm	Extrusion	–	–	MAPP–	340–380	18.7–19	1870–2150	30.1–33.8	–	268–495	[101]		
CA: coupling agent; BA: blowing agent; TD: thermal degradation; ACA: Azodicarbonamide; MAPE: Maleic anhydride-grafted polyethylene; MAPP: maleic anhydride-grafted polypropylene; MAH: maleic anhydride, SEBS-g-MA: styrene-(ethylene-octene)-styrene triblock copolymer grafted with maleic anhydride; PPAA: acrylic acid grafted polypropylene; POE: ethylene-octene copolymer; EO-g-MAH: maleic anhydride grafted ethylene-octene metallocene copolymer; CAPE: carboxylated polyethylene; TDM: titanium-derived mixture.														

Table 3. Mechanical and thermal properties of natural fiber composites based on thermoplastic matrices.

Coupling agents are usually copolymers containing functional groups compatible with the fibers (hydroxyl groups) and the polymer matrix [74]. These reactions (chemical or physical) are increasing interfacial adhesion leading to improved mechanical properties and water absorption reduction [22, 65, 71–73, 75, 76, 99, 102, 103]. Coupling agents can be mixed with the polymer matrix by extrusion previously to fibers addition [65, 74, 92] but can also be added during composite compounding, i.e. mixing the matrix, fiber and coupling agent all together [55, 72, 83, 90, 97–99, 102–104]. Likewise, natural fibers can be functionalized by treating them with a coupling agent in solution, to increase compatibility with the polymer matrix [22, 71, 73–76].

Since natural fibers start to degrade at lower temperature (150–275°C) than most polymer matrices (350–460°C) [60, 63, 74, 83, 105], fiber mercerization and coupling agent addition were shown to improve the thermal stability of the fibers and therefore of the final composites [24, 29, 73, 75, 85, 91, 92].

4. Hybrid composites

To improve on the properties of natural fiber composites and/or overcome some of their limitations such as moisture absorption, thermal stability, brittleness and surface quality, the concept of hybrid composite was developed. The idea is to combine natural fibers with other fibers or particulate reinforcements, which can be of natural or synthetic origin such as glass fibers or rubber particles [15, 51, 63, 106–109]. The main purpose of blending different reinforcements is to obtain a material with better properties than using a single reinforcement. Assuming there is no chemical/physical interaction between each type of fibers, the resulting

properties of hybrid composites ( $P_H$ ) should follow the rule of hybrid mixtures (RoHM) given as [106, 110, 111]:

$$P_H = P_{C1}V_{C1} + P_{C2}V_{C2} \quad (1)$$

where  $P_{C1}$  and  $P_{C2}$  are the properties of composite C1 and C2, respectively, while  $V_{C1}$  and  $V_{C2}$  are their respective volume fractions such that:

$$V_{C1} + V_{C2} = 1 \quad (2)$$

Naturally, the model can be generalized for more than two types of reinforcement.

Natural and synthetic reinforcements combination has showed to improve several composite characteristics such as thermal stability [106, 112–114], impact strength [63, 115–117] and water uptake [70, 112–114, 118, 119]. But the combination of two different types of lignocellulosic fibers was shown to control water absorption [53, 103, 110] and increased impact strength [103, 120], especially when using coupling agents.

The final properties of hybrid composites depend are function of different factors [53, 74, 104, 120], and **Table 4** summarizes some of the most important mechanical and thermal properties of hybrid composites based on thermoset matrices. The effect of fiber and matrix type, as well as fiber surface treatment is reported with their mechanical properties and thermal degradation temperature. Similarly, **Table 5** reports the corresponding information for hybrid composites based on thermoplastic matrices. In general, it is observed that combining natural fibers with inorganic reinforcements leads to improved thermal stability and impact strength, as well as higher flexural and tensile moduli. Moreover, **Table 6** shows that water uptake decreases by combining two natural fibers from different sources, or using natural fibers with inorganic reinforcements in hybrid composites based on thermoplastics matrices.

Matrix	Fibers	Manufacturing process	Fiber treatment	Mechanical properties					TD (°C)	References
				E (GPa)	TS (MPa)	FS (MPa)	FM (GPa)	IS (kJ/m <sup>2</sup> )		
Polyester	Hemp/wool	Pultrusion	–	16.84	122.12	180	11	–	–	[18]
	Palmyra palm leaf/jute	Compression molding	NaOH solution	2.3–5.1			15.3–19.3	24.7–36.4	–	[121]
	Banana/sisal	Hand lay-up + compression molding	–	1.1–1.5			2.7–4.2	~16–37	–	[122]
	Coir/silk		NaOH solution		11.4–17.4		37.4–42		–	[123]

Matrix	Fibers	Manufacturing process	Fiber treatment	Mechanical properties					TD (°C)	References
				E (GPa)	TS (MPa)	FS (MPa)	FM (GPa)	IS (kJ/m <sup>2</sup> )		
Epoxy	Oil palm/glass	Compression molding	–	~2.5–5.5	~20–75	~30–138	~1.5–8	~7–16	–	[124]
	Banana/kenaf	Hand lay-up	Solutions of: NaOH SLS	–	45–139	75–172.2	–	~15–28	–	[125]
	Ramie/cotton	Compression molding	–	–	24.2–118	–	6.3–27.4	–	–	[126]
	Sisal/roselle	RTM	–	–	30.1–58.7	48.4–63.5	–	1.39–1.41	–	[127]
	Sisal/glass	Hand lay-up	–	–	~78–95	~70–265	~2.1–11	~66–88	–	[128]
	Sisal/jute/glass	Hand lay-up	–	–	111.2–232.1	214.1–308.6	–	–	–	[118]
	Hemp/glass fibers	Hand lay-out + compression molding	NaOH solution	–	–	–	–	–	345	[107]
	Banana/jute	Hand lay-up + compression molding	–	0.6–0.7	16.6–19	57.2–59.8	8.9–9.1	13.44–18.23	376.5–380	[108]
	Banana/sisal	Hand lay-up	–	0.6–0.7	16.1–18.6	57.3–62	8.9–9.3	13.2–17.9	–	[129]
	Jute/bagasse	Hand lay-up	NaOH HCl solution	0.3–0.7	0.6–1.7	6.9–15.9	0.6–1.7	6.9–15.9	438.2–475.9	[109]
Polyurethane	Jute/coir	Hand lay-up	NaOH Cyclohexane/ethanol Furfuryl alcohol	~0.3–0.7	~8.5–35	~39–37	~0.5–1.5	–	–	[130]
	Banana/silica	Hand lay-up	–	6.5–9.1	–	–	–	–	–	[111]
	Sisal/silica	Hand lay-up	–	4.7–6.1	–	–	–	–	–	[111]
Polyurethane	Hemp/wool	Pultrusion	–	18.91	122.66	~142	~12	–	–	[18]
Vinyl ester	Hemp/wool	Pultrusion	–	15.27	112.54	~143	~13	–	–	[18]
	Jute/ramie	VARTM	–	6.7–6.8	6.2–6.7	–	–	18–19	–	[131]
	Coconut/sisal/glass	Molding	–	–	–	–	–	1993–16373	–	[117]

Matrix	Fibers	Manufacturing process	Fiber treatment	Mechanical properties					TD (°C)	References
				E (GPa)	TS (MPa)	FS (MPa)	FM (GPa)	IS (kJ/m <sup>2</sup> )		
	Vetiver/glass	Hand lay-up	NaOH solution	1–2.4	53.2–69.8	97.3–131.9	2–3.6	–	–	[116]
	Jute/vetiver	Hand lay-up	NaOH solution	1.7–1.9	63.3–71.7	114.8–133.1	2.9–3.6	–	–	[116]

E: tensile modulus, TS: tensile strength, FS: flexural strength, FM: flexural modulus, IS: impact strength, TD: thermal degradation.

**Table 4.** Mechanical and thermal properties of natural fiber hybrid composites based on thermoset matrices.

Manufacturing process	Composite	Coupling agent	Filler content (%)	Filler surface treatment	Mechanical properties					TD (°C)	References
					E (MPa)	TS (MPa)	FS (MPa)	FM (GPa)	IS (J/m)		
	PP-glass/ flax fibers	MAPP (5%)	40 (vol)	–	522–629	21.9–25.5	–	–	37.9–49.6	–	[106]
	MAPE-GTR rubber/hemp fiber	–	10, 30, 50, 60	–	120–243	9.8–14.3	–	363–781	139.6–294–239.8	–	[63]
	PP-Kenaf/coir/MMT	MAPP (5%)	30	–	300–360	11–12	–	–	–	–	[132]
	PP-NNC/Maple fibers	MAPP (2%)	21	–	444.9	25.4	–	1735.2	–	–	[104]
	PP-wood/SiO <sub>2</sub>	MAPP (4.5%)	50	–	–	32–45	48–65	2400–3540	–	348	[133]
	PP-wood/CaCO <sub>3</sub>										
	PP-wood/milled glass fibers										
	PP-sisal/glass fibers	MAPP (1%, 2%, 3%)	30	–	41.75–55.1	970–1686	47.4–67.5	1900–2800	59.3–81.6	346–384	[70]
	PP-jute/flax fibers	MAPP (19.12%)	25.96%	PP/jute and MAPP/flax woven fabrics were treated with NaOH solution	29.7–42.6	2437.3–2852.4	50.1–68.8	1399.7–2331.8	–	–	[134]

Manufacturing process	Composite	Coupling agent	Filler content (%)	Filler surface treatment	Mechanical properties					TD (°C)	References
					E	TS	FS	FM	IS		
					(MPa)	(MPa)	(MPa)	(GPa)	(J/m)		
	LDPE-banana/coir fibers	MAPP (5%)	15	Solutions of: NaOH Acetylation bleaching with H <sub>2</sub> SO <sub>4</sub>	36.2–50		29.5–52.4		9.3–13.6	473	[135]
	HDPE-coir/Oil palm fibers	MAPE (2%, 4%)	40	Hot water and soap	8–13.5	550–630	17–27	1570–2380	–	–	[120]
	HDPE-kenaf/pineapple leaf fibers (PALF)	–	40	–	27–30	550–680	23–28	1700–2100	–	–	[110]
	PS-banana/glass fibers	–	20	Solutions of: NaOH Benzoyl chloride PSMA	29–38.8	1462.2–1558.3	7.9–11.3	489.7–698.8	–	–	[136]
Injection + compression	PP-SBR rubber/birch wood	MAPP (3%, 5%)	0–40	–	10.5–25	520–1560	–	–	–	–	[51]
Injection molding	PP-sisal/glass fiber	N/A (3.5%)	10, 20, 30	Boiled in methanol and benzene mixture and with NaOH solution	–	–	–	–	100	190–230	[112]
	PP-sisal/glass fibers	MAPP (3%)	30	–	29.2–31.6	2330–2430	66.7–68.8	4.03–4.14	16.7–20	331.3–464.7	[113]
	RPP-date palm wood/glass fiber	–	30	–	19.5–21	1100–1300	–	–	–	361.8–479.4	[114]
	PP-hemp/glass fibers	MAPP (5%)	40	–	52.5–59	3800–4300	97–101	5000–5400	49–55.4	360–474	[57]
	PP-wood flour/glass fiber	MAPP PP-g-MA POE-g-MA	40	–	28–45.4	–	39.7–62.8	2680–3497	–	345–363	[137]

Manufacturing process	Composite	Coupling agent	Filler content (%)	Filler surface treatment	Mechanical properties					TD (°C)	References
					E	TS	FS	FM	IS		
					(MPa)	(MPa)	(MPa)	(GPa)	(J/m)		
		SBS-g-MA (3%, 6%)									
	PP-wood/kenaf fibers	MAPP (1%)	40	–	39–44	2771–3008	–	–	–	–	[138]
	PLA-kenaf/corn husk	–	30	NaOH solution Sodium lauryl sulfate solution Silane and potassium permanganate	–	1547	–	–	–	–	[139]
	PLA-banana/nano-clay	–	33	–	67	4965–5577	105–108	7715–7725	119–120	295–397	[140]
	HDPE-Pine/agave fibers	MAPE (3%)	20, 30	–	20.5–26.5	415–650	24–32	670–1180	37–47	–	[53]
	HDPE-coir/agave fibers	MAPE (3%)	20, 30	NaOH solution	19.5–25.9	355–500	23.3–31.9	890–1190	42–68	–	[103]
	HDPE-sisal/hemp	MA solution (10%)	25, 30	NaOH solution	15.7–19.2	–	–	–	–	–	[141]
	PP-coir shell/coir fibers	SEBS-g-MA (8%)	20	NaOH solution Benzoyl peroxide solution	26.5–29.5	1050–1300	–	–	–	344–349	[74]
	PLA-banana/sisal fibers	–	30	–	57–79	1700–4100	91–125	4200–5600	–	–	[142]
	PLA-hemp/lyocell	–	40	–	41.4–71.5	4643–7035	–	–	–	–	[143]
	PLA-hemp/kenaf fibers	–	40	–	34.4–61	4920–7039	–	–	–	–	
	HDPE-wood/hollow	–	50	–	26.2–31	3300–3600	–	–	–	–	[119]

Manufacturing process	Composite	Coupling agent	Filler content (%)	Filler surface treatment	Mechanical properties					TD (°C)	References
					E	TS	FS	FM	IS		
					(MPa)	(MPa)	(MPa)	(GPa)	(J/m)		
Extrusion	glass microspheres										
	HDPE-wood/bast fibers	–	60	Vinyl triethoxysilane	42–44	650–700	73–77	4900–5250	–	–	[144]
	HDPE-wood/Kevlar	–	60	Allyl and 3–trimethoxy silyl-propyl	13.8–19.8	3050–4100	24.5–3600	2200–3400	–	–	[145]
Extrusion calendering	PP-jute/glass	–	20, 30, 40	–	42–63	4660–7170	72.8–102.5	3550–5950	–	–	[69]

MAPP: maleic anhydride-grafted PP; MAPE: maleic anhydride-grafted PE; GTR: ground tire rubber; LDPE: low density polyethylene; HDPE: high density polyethylene; PS: polystyrene; SBR: styrene butadiene rubber; RPP: recycled polypropylene; PP-g-GMA: glycidyl methacrylate-grafted PP; POE-g-MA: maleic anhydride-grafted ethylene-octene copolymer; SEBS-g-MA: maleic anhydride-grafted hydrogenated styrene-butadiene-styrene; PLA: polylactic acid.

Table 5. Mechanical and thermal properties of hybrid composites based on thermoplastic matrices.

Matrix	Reinforcements	Observations	References
MAPE	GTR rubber/hemp fiber	GTR decreases water uptake	[63]
PP	Kenaf/coir/MMT	Water uptake is reduced by hybridization	[132]
	Wood/SiO <sub>2</sub>	SiO <sub>2</sub> , CaCO <sub>3</sub> and milled grass decreased water uptake	[133]
	Wood/CaCO <sub>3</sub>		
	Milled glass fibers		
	Hemp/glass fibers	Glass fiber reduced water uptake	[57]
	Wood/glass fibers	Increasing fiber glass weight ratio, water uptake was reduced.	[146]
HDPE	Pine/agave fibers	Pine fiber decreased water uptake in hybrid composites	[53]
	Coir/agave fibers	Coir reduced water uptake in hybrid composites	[103]

PP: polypropylene; HDPE: high density polyethylene; MAPE: maleic anhydride-grafted polyethylene; GTR: ground tire rubber; MMT: montmorillonite.

Table 6. Water uptake in hybrid composites using thermoplastic matrices.

5. Auto-hybrid composites

Composites reinforced with two sizes of the same type of reinforcement are referred to as auto-hybrid composites. As these composites only have a single type of reinforcement, they are



easier to recycle. But most importantly, these materials were shown to exhibit a positive deviation from the RoHM depending on fiber concentration, weight ratio, size and type [64, 102, 147]. Nevertheless, the auto-hybridization effect seems to be more influenced by the total fiber content than coupling agent addition [64, 147]. However, coupling agent addition is always important to improve tensile strength [102]. As total fiber content, fiber type and coupling agent content, all affect the level of deviation from the RoHM, and optimization of these parameters is a new challenging field of research to develop better composite performances. **Table 7** summarizes the limited amount of work on auto-hybrid composites using natural fibers as reinforcement.

Processing	Composite	Coupling agent	Fiber diameter (μm)	Fiber content (%)	Crystallinity index (%)	Main results	References
Injection	PP-hemp fibers	MAPP (3%, 5%)*	Fiber: 300–710 Powder: 45–180	20, 30	–	Hybridization more effective at 20 wt.% reinforcement Optimum weight ratio of 20/80 (powder/fibers) 3% of coupling agent was more efficient Ductility and impact strength decreased with fiber content Tensile and flexural modulus increased with fiber content	[147]
	HDPE-pine fibers	MAPE (3%)	Short fiber: 40–105	10, 20, 30	56.2–61.1	Coupling agent increased tensile strength, and decreased tensile modulus, flexural strength and impact strength of auto-hybrids	[102]
	HDPE-agave fibers		Long fiber: 300–425		53.3–57.4	Total fiber concentration affected hybridization being more effective at 20 and 30 wt.% Higher values of mechanical properties were obtained at 30/70 (short/long) weight ratio (without coupling agent) in auto-hybrids Crystallinity index decreased with coupling agent addition	
	PP-pine fiber – PP-agave fibers	–	Short fiber: 50–212	10, 20, 30	–	Hybridization did not affect flexural and tensile strength Hybridization was more effective at 30/70 (short/	[64]

Processing	Composite	Coupling agent	Fiber diameter (μm)	Fiber content (%)	Crystallinity index (%)	Main results	References
			Long fiber: 300–425			long) and 50/50 (short/long) weight ratio Positive hybridization effect was higher at 20 and 30 wt.% fiber content Impact strength was higher at 20 wt.% with a 30/70 (short/long) weight ratio Water absorption was not affected by fiber size	
Compression molding	LLDPE-maple fibers	MAPE (3%)	Short fibers: 0–45 Medium fibers: 125–250 Long fibers: 355–450	5, 10, 15, 20	13–32	Positive deviation of RoHM at 30/70 (smaller/longer) weight ratio, regardless of fiber size 20 wt.% showed higher RoHM positive deviation and auto-hybridization was more effective Positive deviation of RoHM is affected by fiber size and total fiber content Tensile and flexural modulus increased with fiber content, but not with fiber size Impact strength and torsion modulus of hybrid composites are affected by fiber weight ratio	[148]

\*MAPP was not used in auto-hybrid composites.

Table 7. Overview of the different investigations on auto-hybrid composites based on natural fibers.

6. Conclusion

Natural fibers are now interesting alternative to replace synthetic fibers due their good specific properties (per unit weight). They have been used to develop different composites based on thermoset and thermoplastic matrices. As for any composite, their mechanical, thermal and physical properties are function of the properties of the matrix and the reinforcement, as well as fiber loading, fiber source and manufacturing process. Nevertheless, interfacial conditions are always important to optimize the general properties.

The main disadvantages of using natural fibers are water uptake, low thermal stability, as well as low mechanical properties due to fiber agglomeration and poor interfacial adhesion, especially at high concentration. The problem is usually more important in thermoplastics than

thermosets due to their difference in initial resin viscosity. But most of the limitations associated to natural fiber composites can be controlled or overcome by the addition of coupling agents and/or fiber surface modifications.

Finally, another possibility to improve the properties of natural fiber composites is to add a second reinforcement to produce hybrid composites. These materials were shown to have improved mechanical and thermal properties over neat natural fiber composites as they follow the rule of hybrid mixture (RoHM) regardless of the matrix, manufacturing processing and fiber combination. Based on this concept, different class of materials was also developed such as all natural fiber hybrid composites (combination of two different natural fibers) and auto-hybrid composites (combination of two different sizes of the same fiber). The latter is highly interesting as positive deviations from the RoHM were reported. This is usually the case around 20 wt.% of total fiber content with around 30/70 short/long fiber ratio regardless of coupling agent addition, fiber type and processing method. This opens the door to a new field of investigation as several parameters can be controlled to optimize the final properties of the materials and to design new applications for these multi-functional composites.

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