

Progress and challenges in sustainability, compatibility, and production of eco-composites: A state-of-art review

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Abstract

Owing to economic and environmental benefits, new generations of materials/commodities follow “from waste to wealth” strategy. Recently, there has been a huge upsurge in research on the development of eco-composites using recycled plastic polymers and agro-residues because the eco-composites satisfy the stringent environment regulations and are cost-effective. Herein, we present a detailed review on the potential use of several types of natural fillers as an efficient reinforcement for recycled plastic polymers. In particular, the characterization of different categories of eco-composites according to their morphological, physical, thermal, and mechanical properties is extensively reviewed and their results are analyzed, compared, and highlighted. Furthermore, a framework to produce functional eco-composites, which includes functionalization of ingredients, critical issues on microstructural parameters, processing, and fabrication methods, is outlined and supported with sufficient data from the literature. Finally, the review outlines the emerging challenges and future prospects of eco-composites to be addressed by interested researchers to bridge the gap between research and commercialization of such a class of material. Overall, the acquired knowledge will guide researchers, scientists, and manufacturers to plan, select, and develop various forms of eco-composites with enhanced properties and optimized production processes.

KEYWORDS

agro-residue, characterization, eco-composite, functionalization, natural fibers, polymers, processing

1 | INTRODUCTION

Eco-composites are newly emerging class of bio-composites, which are produced by mixing recycled plastics and cellulose waste to obtain enhanced materials.^{1,2} Developing new eco-composites, which consist of recycled polymers and agro-residue reinforcing fillers, with better understanding of overall performance will increase their aggregated values and potential applications. The eco-composites reduce

harmful environmental impact as waste materials and are useful in closing the carbon cycle and producing green composites.^{3–5} To gain a better understanding on eco-composites, the material properties are studied according to international standards and manufacturing requirements. Several eco-composites have been successfully developed and qualified for use in a broad range of non-load bearing products. For instance, recent studies show that using recycled polymers as a matrix for manufacturing

wood–plastic composites (WPCs) is a viable alternative to traditional matrices, which can produce high performance eco-composites as well as help in reducing the recycling and disposal cost.^{6–8} Two types of produced waste can be classified as matrix component. First, the municipal solid waste (MSW), which is generated by from offices, government institutions, hospital, and schools. Second, the non-municipal solid waste, which is generated by industries and factories. Effective utilization of solid wastes helps in reducing waste hazards and environmental risks, which will eventually lead to better quality of air, water, and soil. Therefore, considering the advantages of recycled plastics and produced waste from agriculture residues, these materials can be utilized in developing cost-effective and eco-friendly products.^{9,10} Bio-based products offer a lighter environmental imprint and enhanced performance. Product lines focus on indoor applications such as automotive interiors and household accessories, and outdoor applications such as decking and fences. Eco-composites continue to offer sustainability, durability, and cost effectiveness in the consumer sector. These materials will certainly continue to address challenges through the development of new products in the foreseeable future. At this stage, it is worthy to mention that recent studies have confirmed that eco-composites behave similar to the traditional plastics. No major equipment is required in the production of eco-composites since existing technologies for thermoplastic processing is sufficient. Only low processing temperature is required for biofiller protection against degradation. Therefore, eco-composites provide a strong base for reducing harmful emissions and may reduce the need for extracting fossil fuels.

The newly developed class of eco-composites distinctly combine the qualities that are suitable for accommodating producer and consumer preferences in competitive markets. They provide a sustainable option of using agro-residues with virgin, recycled or renewable plastics. Recently, plastic manufacturers have become concerned about offering more sustainable products to different market segments and customers including agriculture and energy. Eco-composite markets differ in comparison to such markets because the emerging industry lacks government support. Instead, the responsibility of eco-composite manufacturers is to provide strategic solutions for cost cutting and remain competitive. To achieve this, well-established industries may need to heavily change their infrastructure and systematically develop new technologies, which require a massive investment. However, eco-composites are made to function within the existing production systems and manufacturing equipment, which can result into major cost savings. In general, the other most encouraging aspect of investing in eco-composites

is that they can possibly replace traditional virgin plastics in future. Plastics are a vital part of manufacturing—demand and markets are already well established. Indeed, there are number of factors that contribute to the improvement of the utilization and sustainability of eco-composites, thus making the investments toward their development valid. Some of these major factors are: degradability, renewability, recyclability, increasing market demand, wide range of products and applications, maturity of supply chain logistics, social and governmental concerns, cost-effectiveness, ability to meet the quality standards, ease and readiness of processing and production, new trends for economic growth, and affordable start-up investment with high rate of return.

The structure of this review is organized as follows: Section 2 reviews the technical sustainability of eco-composites. It provides extensive data and findings on the characterization of different types of eco-composites in terms of morphological, physical, thermal, and mechanical properties. Section 3 discusses the critical issues of eco-composite performance such as compatibility, treatment, functionalization of ingredients, and factors relating to high-performance production. Section 4 outlines challenges and future perspectives, and concluding remarks are provided in Section 5.

2 | CHARACTERIZATION OF MATERIAL PROPERTIES

In general, eco-composites are characterized by the type of filler and recycled polymer. Further, prior to application in industrial products, eco-composites are evaluated against several aspects. Table 1 summarizes different types of successfully developed eco-composites. This section discusses major research works reported in literature to characterize eco-composites according to their morphological, physical, thermal, and mechanical properties.

2.1 | Morphological observations

In principle, natural fiber type and geometry, base recycled polymer, chemical additives, and fabrication processes are the major contributing factors to the overall behavior of eco-composites. The irregular geometry of natural fibers will generally lower the strength of eco-composite due to weak interfacial regions, poor compatibility, and wetting between the polar plant fibers and non-polar polymers.⁵⁴ In addition, filler content should be optimized to achieve desired output properties. A high content of natural fiber embedded in polymer can initiate dominant fiber agglomeration, causing poor fiber dispersion and excessive voids or cavities in eco-composites.³

TABLE 1 List of eco-composites successfully developed by previous studies

Recycled resin	Natural filler
Recycled low density polyethylene (rLDPE)	Cocoa (C) ¹¹
	Corn Husk Fibers (CHF) ¹²
	Eggshell Nanoparticles ¹³
	Rice Husk (RH)/Nano Clay (NC) ¹⁴
	Rice Husk (RH)/Nano Silica (NS) ¹⁴
	Rice Husk (RH) ¹⁵
	Uncarbonized bagasse particles (UBp); Carbonized bagasse particles (CBp) ¹⁶
Recycled high density polyethylene (rHDPE)	Wheat Straw (WS) ¹⁷
	Banana Fiber (BF)/Fly Ash Cenospheres ¹⁸
	Banana Fiber ¹⁹
	Chestnut Cupula (CC) ²⁰
	Date Palm Mesh (DPM) ²¹
	Durian Peel (DP) ²²
	Flax Fibers (FF) ^{23,24}
	Hemp Fiber (HF) ²⁵
	Kapok Fiber (KAF) ²⁶
	Piassava Fibers (PF) ²⁷
	Reed Fibers (RF) ²⁸
	Rice Husk (RH) ^{29–31}
	Rice Straw (RS) ³²
	Sisal Fibers (SF) ^{33,34}
Sugarcane Bagasse (SB) ^{35–39}	
Water Hyacinth Fiber (WHF) ⁴⁰	
Recycled polypropylene (rPP)	Almond Tree Leaves (ATL) ⁴¹
	Banana Fiber (BF) ⁴²
	Corn Straw (CS) ⁴³
	Curaua Fiber (CUF) ⁴⁴
	Date Palm Fiber (DPF)/Glass Fibers ⁴⁵
	Date Palm Fiber (DPF) ^{46,47}
	Jute Fibers (JF) ⁴⁸
	Kenaf Fiber (KF)/recycled Carbon Fiber (rCF) ⁴⁹
	Kenaf Fiber ⁵⁰
	Kenaf Fiber/Nano-CaCO ₃ /Fire Retardant ⁵¹
	Kenaf Fiber/Graphene nanoplatelets ⁵²
	Mengkuang Leaf Fiber (MLF) ⁵³
	Oil Palm Trunk (OPT); Oil Palm Frond (OPF); Empty Fruit Bunches (EFB) ^{54,55}
	Pine Nutshell (PN) ⁴³
	Pineapple Fiber (PAF) ⁵⁶
	Quill from Chicken Feathers ⁵⁷
	Rice Husk ³¹
	Sisal Fibers ^{58,59}
	Snail Shell (SS) ⁶⁰
	Wheat Straw/Inorganic filler ⁶¹

(Continues)

TABLE 1 (Continued)

Recycled resin	Natural filler
Recycled polyethylene terephthalate (rPET)	Date Palm Fiber (DPF) ⁶²
	Empty Fruit Bunch Fibers (EFB) ⁶³
	Rice Husk (RH) ⁶⁴
Recycled ternary polyolefin polymer blends (PP/HDPE/LDPE) (rTPB)	Date Palm Fiber (DPF) ^{65,66}
	Kenaf Fiber ⁶⁷
rPP/rPET	Rice Husk ³
Unsaturated polyester from recycled polyethylene terephthalate (UP-rPET)	Coconut Fibers (CF) ^{68,69}
	Kenaf Fibers ⁷⁰

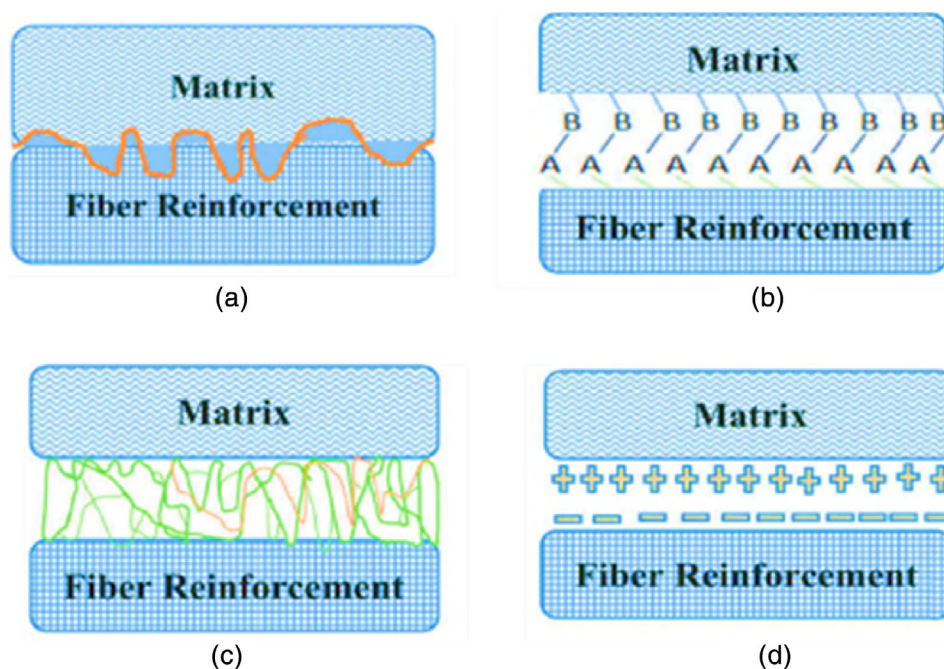


FIGURE 1 Forms of interfacial bonding: (a) mechanical interlocking, (b) chemical bonding, (c) molecular inter diffusion, and (d) electrostatic bonding.⁷³ [Color figure can be viewed at wileyonlinelibrary.com]

Coupling agents assist in reducing gaps, voids, and fiber agglomeration. Moreover, they enhance the filler encapsulation by the matrix. Therefore, homogeneity of fiber/particle distribution in the matrix determines the overall strength and quality of the produced eco-composites. Optical microscopy and scanning electron microscopy (SEM) are used to investigate the distribution of fillers and other microstructural properties. In the case of rLDPE matrix, morphology analysis suggests that the fiber loading (content) has major influence on the microstructure of the eco-composites.¹² Poor homogeneity and compatibility between corn husk fibers and rLDPE were observed as the fiber loading increases beyond 20% owing to fiber malleating with rLDPE. Agunsoye and Aigbodion¹⁶ reported that the agglomeration and

clustering of bagasse particles occur when their concentration exceeds 40 wt%. In addition, Amieva et al.⁵⁷ reported that quill particles were wetted by rPP and no voids were observed. At higher quill loadings, the surface of filler becomes less ductile and more irregular, because the rPP matrix cannot adequately encapsulate the quill particles. In contrast, chemically treated fibers in rPP-based eco-composites exhibited no significant fiber agglomeration or micro voids when fractured specimen were tested using SEM.

The use of nanofillers in nanocomposites has demonstrated an improvement in their electrical, magnetic, dielectric as well as mechanical properties. The augmented filler-polymer interactions influenced the role of conductive and magnetic fillers in upgrading the

FIGURE 2 Comparison between the microstructures developed in the fracture regions of tensile tested eco-composites.⁴⁶ [Color figure can be viewed at wileyonlinelibrary.com]

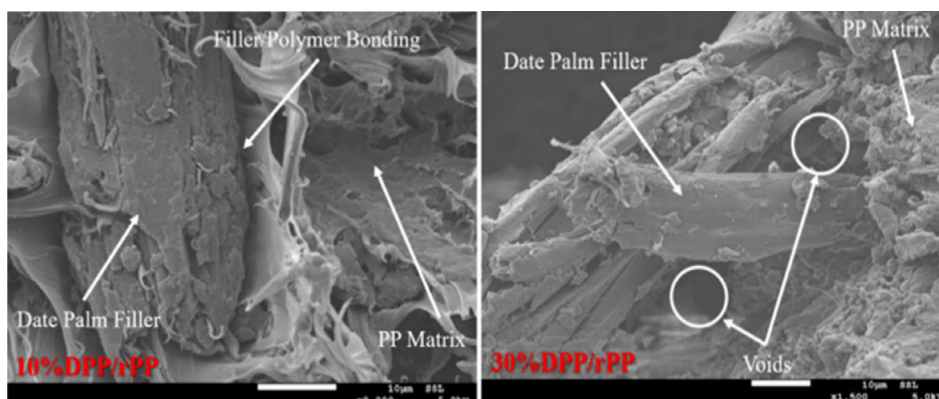
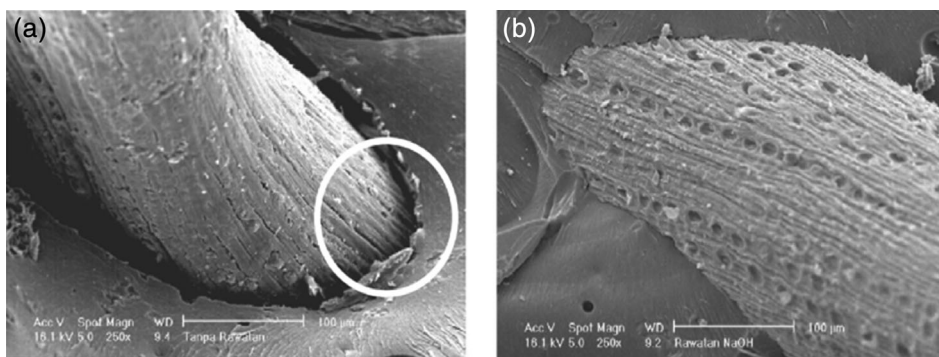


FIGURE 3 SEM micrographs of the tensile fractured surfaces of (a) empty-fruit-bunch-reinforced composites, (b) untreated composite.⁶³



characteristics of polymer matrix.⁷¹ The realized strength of hybrid nanocomposites is owed to the extremely small size and increased surface area of additive particles. A higher number of particles can be accommodated in thin films forming stronger interfaces.⁷²

The above findings support that the overall eco-composites characteristics are strongly affected by their microstructures and interactions among all constituents. In particular, the compatibility between a filler and surrounding matrix or the bonding of a fiber with an encapsulated matrix is critical to the mechanical behavior of synthesized material. Generally, interfacial bonding can occur in four different forms: (a) mechanical interlocking, (b) chemical bonding, (c) molecular inter diffusion, and (d) electrostatic bonding, as shown in Figure 1.⁷³

Fiber pull-out from the matrix is the result of a secondary failure such as fiber breakage and delamination, followed by a primary failure of the whole eco-composite.⁷⁴ Hence, proper treatment of natural fibers confirms the removal of low molar mass impurities including wax and oil, which make the fiber surface rougher, resulting in better bonding between the fibers and the surrounding matrix. Chianelli-Junior et al.³³ observed that untreated sisal fibers have poor interaction with their hosting rHDPE matrix; however, no fiber pull-out was observed for the eco-composites containing alkalinized and acrylate fibers.^{21,25,37,68,69} Farahani

et al.⁷⁰ reported that the kenaf fibers are not completely encapsulated with their matrix due to poor adhesion between the untreated fibers and matrix, as a gap between the two components was shown by SEM micrographs. However, an enhancement in the mechanical properties was observed because of better interlocking between the filler and recycled matrix due to filler treatment.⁶⁴ In fact, filler treatment and polymer grafting can make the interacting surfaces rougher. Figure 2. confirms the degradation of the tensile strength of DPP/rPP composites, which was caused by poor filler–polymer interactions at the surfaces.⁴⁶ The microstructural investigations of the fractured surfaces of eco-composites have exhibited good bonding between UP-rPET and coconut fiber owing to the porous and fibrillated nature of the treated fibers.^{68,69} Furthermore, Tan et al.⁶³ reported that the treatment of empty fruit bunch fibers increases the interfacial adhesion between the fibers and UP-rPET, as shown in Figure 3. SEM analysis have shown fiber pull-out and debonding between cleaned banana fibers and rHDPE, thus indicating a weak interfacial characteristics.¹⁹ An improved adhesion in microstructure was observed for untreated rice husk (RH) fiber mixed with functionalized rHDPE.³⁰ This explains the support of the RH fibers to the rHDPE matrix during the load transfer process. Fávares et al.³⁴ also noticed that alkalinized sisal fiber and oxidized rHDPE adversely affect the microstructure interactions,

which is responsible for poor mechanical properties of the produced eco-composite. Moreover, SEM fractography suggested to use steam explosion for treating fibers because it relatively reduces void content and the spacing between microfibrils in a fabricated eco-composite.⁴¹ From different perspective, adding nanoparticles to eco-composites enhances the interaction among ingredients and performance of hybrid eco-composites. In rice husk-fiber-based eco-composites, nano silica and nano clay decrease the numbers of interfacial cavities, reinforcing the pulled-out resistance of the fiber.¹⁴ Morphological images of tensile fractured surfaces of kenaf/graphene nanoplatelets/rPP nanocomposites revealed lowered micro-holes and fibers aggregation signifying enhanced interfacial adhesion among ingredients due to kenaf flour treatment.⁵²

A strong correlation between the length and orientation of a fiber and the tensile strength exists; a higher tensile strength is achieved in the case of long fibers.⁷⁵ However, the aspect ratio of the natural fiber is one of the most influential factors for the mechanical strength of polymer composites. It was reported that for effective reinforcement, the aspect ratio should exceed 10.²⁸ In contrast, short fibers are easier to handle and process, and maintain a better dispersion in the corresponding matrix. For example, Spinace et al.⁴⁴ reported that the composites fabricated with short fibers exhibit good dispersion compared to those filled with long fibers.

2.2 | Physical properties

In general, eco-composites have lower density compared to other types of composites. The density is highly

dependent on the internal bond strength, which increases the density and the overall eco-composite strength.¹² Amieva et al.⁵⁷ reported that the quill/rPP composite density remains constant if the quill content is in the range of 10%–15%, implying better interaction between rPP and quill. The density of cocoa/rLDPE eco-composite decreases (from 0.81 to 0.61 g/cm³) with an addition of lignocellulosic waste into the polymeric matrix.¹¹

Crystallinity of a substance is an indication of its atomic, ionic, or molecular order; the structure of crystallographic planes; and the number of amorphous phases. To understand crystallinity features, X-ray diffraction (XRD) is used. Figure 4(a),(b) show the XRD patterns of neat polymer and developed eco-composites, as reported by Rosa et al.⁴⁰ The coexistence of α -PPr ($2\theta = 17^\circ$) and β -PPr ($2\theta = 16.3^\circ$) crystals in the eco-composites, PPr-SAF01 and PPr-SAF05, with the formation of the β -phase is due to inhomogeneous nucleation of the steamed fibers, which can be assumed as a nucleating agent. The α -crystalline substance is sensitive to the volume of the nucleating agent, although the good quality bonding and distribution of fibers determine an extension of the nucleation impact. It seems that SAF provides better adhesion to the rPP matrix.

Water uptake (moisture absorption) is another important physical feature of the eco-composites. It is considered as the main obstacle for utilizing the eco-composites in various outdoor applications. During water uptake, the natural fillers stick on the surface, and then the water penetrates deep inside the material. This is due to the presence of lumens and hydroxyl groups in the natural fiber biomass, which negatively affect the hydrophobicity of the eco-composites.³⁵ Therefore, the eco-composites with high fiber

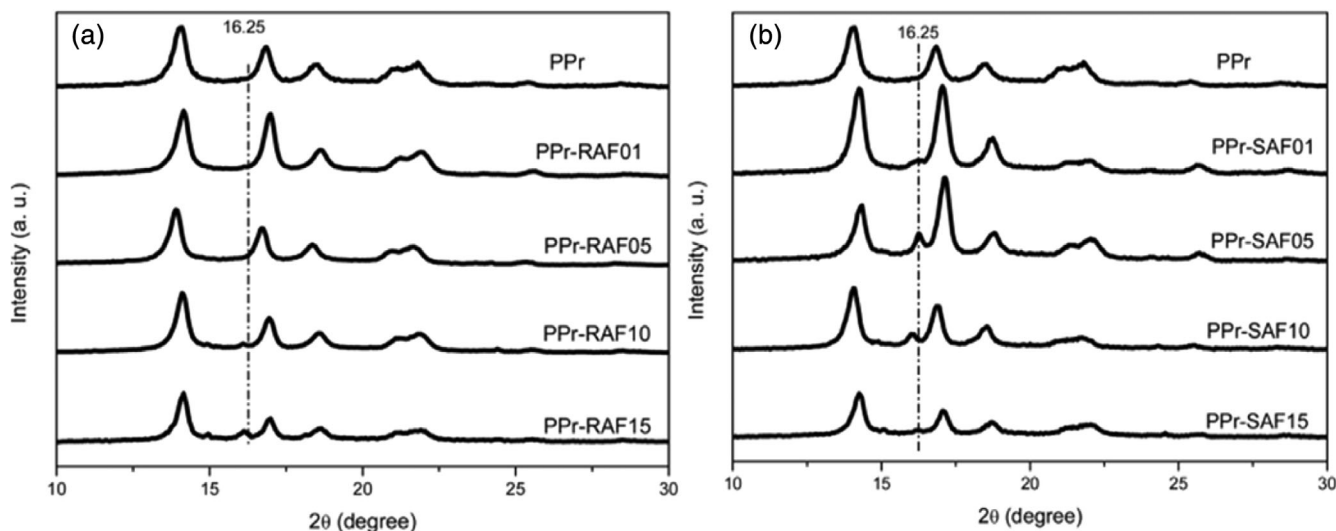


FIGURE 4 X-ray diffractograms of rPP eco-composites with the incorporation of: (a) untreated (raw) almond fiber (RAF) and (b) steam-exploded almond fiber (SAF).⁴¹

volume fraction are more vulnerable to water uptake.³⁷ Further, poor adhesion between the natural fibers and their hosting polymers is caused by the hydrophilic nature of the fibers. In particular, the poor wettability is responsible for higher percentage of water uptake.^{37,70} Water absorption in the eco-composites occurs through fine voids, gaps between the polymer and fiber, and microcracks in the polymeric matrix. Therefore, the polymer–filler interfacial strength significantly affects the water uptake of the eco-composites, as illustrated in Figure 5.⁷⁶ The exposure of the eco-composites to moisture eventually impels the natural fillers to swell. In addition, moisture diffusion into the interior parts of the eco-composites involves flowing of water from a high-concentration zone to a low-concentration zone. This diffusion is further intensified through poor moistening of fibers and surface flaws, thereby leading to mechanical fatigue.⁷⁷ This effect is prominent if the matrix has a high content of microcracks and less filler–polymer compatibility. The flow of water along the weak interface induces leaching of the water-soluble substances present in the filler, causing further fiber debonding from the matrix. Consequently, the strength and durability of the eco-composites degrade. The water absorption of wheat straw/rPP material at saturation was markedly increased by incorporating natural fillers to rPP. However, hybrid nanocomposites produced from wheat straw and inorganic nanofillers showed a cut in water absorption compared to wheat straw/rPP based eco-composites.⁶¹

To enhance the eco-composite quality for outdoor applications, it is recommended to modify the natural fiber using coupling agents and surfactants to enhance the fiber–matrix interaction.^{36,55,68,69} Similarly, chemical compatibilizers for

polymers improve water retardation capacity of eco-composites.⁴³ The modified polymer matrix covers treated fibers with strong bonding interphase promoting greater hydrophobicity and less significant water absorption and, consequently, avoiding degradation of mechanical performance.^{12,70} Hence, the eco-composites with these properties are appropriate for open-air applications such as in the field of construction or outdoor furniture.³

Viscosity is another important property that gives an indication of processability of eco-composites. Generally, melt flow index (MFI) of eco-composites is less compared to the neat polymer matrix. Particularly, in the case of large particle size, where viscosity of an eco-composite increases.^{28,45,54} It was reported that recycling increases the complex viscosity of LDPE and HDPE blends, which exhibit miscible behavior in molten state. However, intricate viscosity of eco-composites can be improved by ethylene propylene diene monomer compatibilizer.⁷⁸

Other studies indicate that the existence of fibers in the polymeric matrix may possibly act as a driving factor for the deterioration of the polymer, while confirming the dimensional stability of the eco-composites.²⁸ Kuang et al.¹⁷ reported that polyacrylate latex (PAL) and β -hydroxyethyl acrylate (HEA) improve the natural fiber–polymer interface, and hence reduce the thickness swelling. This is due to the presence of hydroxyl groups in HEA that promote the formation of an intramolecular hydrogen bond with the fiber. It should be noted that the mechanical properties, thickness swelling, and stability of the eco-composites were meeting the international standards for severe-duty load-bearing applications in humid conditions.

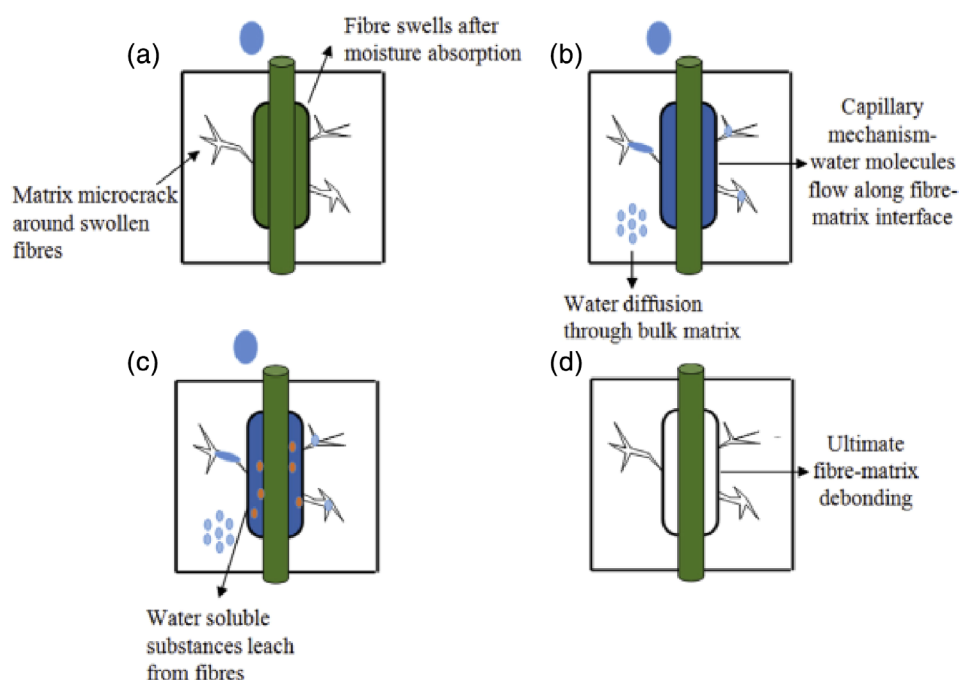


FIGURE 5 Mechanism of diffusion in eco-composites.⁷⁶ [Color figure can be viewed at wileyonlinelibrary.com]

2.3 | Thermal behavior

The thermal characteristics of the developed eco-composites such as thermal stability, coefficient of thermal expansion, thermal degradation, decomposition temperature, transition temperature, enthalpy, and crystallinity degree are evaluated using differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), and dynamic mechanical analysis (DMA). In general, both the thermal stability of cellulose and preservation of internal structure of microfibrils contribute to achieving enhanced mechanical properties.²⁷ Furthermore, thermal characteristics of the fibers are altered by applying proper chemical treatments and modifications. For example, Neto et al.³⁷ showed that acetylated fibers enhance the thermal stability of their lignocellulose structure, as compared to mercerized and unmodified fibers. The enhanced thermal stability of the acetylated fibers is achieved due to the removal of amorphous components, such as lignin and hemicellulose, and the addition of carbonyl groups, which increase the molar mass of the fibers. Moreover, fiber treatment reduces the lignin and hemicellulose contents during the chemical modification, as confirmed by TGA. Similarly, it was found that the thermal stability of the developed eco-composites is significantly increased with the addition of natural fibers to the recycled matrix.¹²

TGA and DTG analysis confirm that the decomposition temperature of date-palm-reinforced rPP eco-composites is not significantly different from that of neat polymer (Figure 6). In such case, two thermal degradation stages for the eco-composite were detected: the first stage occurred at a temperature significantly lower than the fiber degradation temperature, and in the second stage, the degradation occurs mainly in the polymer. The

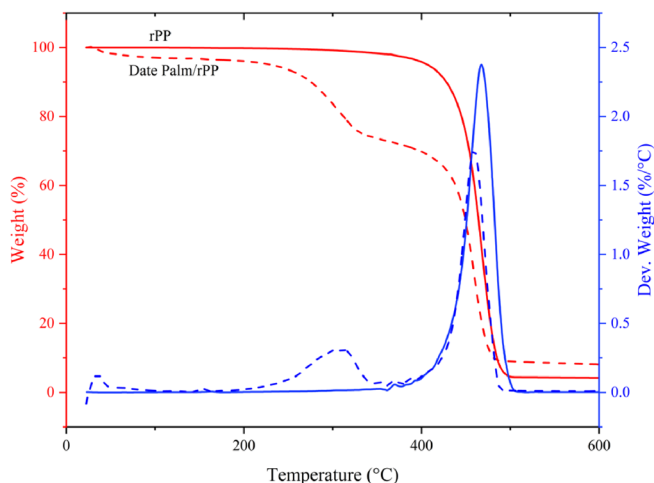


FIGURE 6 Thermal degradation of rPP and date-palm-filled rPP [Color figure can be viewed at wileyonlinelibrary.com]

coupling agents exhibited little influence on the thermal degradation of bio-composites.³⁹ In comparison to neat rHDPE, sugarcane bagasse/rHDPE eco-composites revealed similar melting temperature and less thermal stability. TGA also showed that the rHDPE-based eco-composites have lower thermal stability as compared to both virgin and recycled rHDPE.^{19,36} In contrast, the incorporation of natural fibers enhanced the thermal stability of rPP-based eco-composites.^{41,43} However, the thermal stability of rLDPE eco-composites was substantially enhanced with the addition of natural fillers. However, Agunsoye and Aigbodion¹⁶ concluded that the coefficient of thermal expansion was degraded due to the elastic–plastic incompatibility between the rLDPE and the bagasse flakes. DSC analysis demonstrated that the use of natural fibers in both the rHDPE and virgin HDPE resins resulted in a lowered peak of heat flow rate.³²

The transition temperature of rPP remained almost unaltered compared to that of virgin PP. The thermal stability was found to be strongly related to the quill content.⁵⁷ The thermal stability of the rPP eco-composites was slightly lower compared to the virgin PP composites. With the help of coupling agents, treated fibers improve the crystalline packing and crystallinity of rPP, as confirmed through XRD and DSC analyses.⁵⁰ The chopped RH may be viewed as a nucleating agent for rPP. Thus, it can promote the crystallization of rPP at its surface, leading to a composite with higher crystallization temperature.⁶⁴ In contrast, high proportion of date wood flour decreases the crystallinity of WPCs. Additionally, the DSC analysis showed that there was a slight difference in the melting temperature of the rPP eco-composite and rPP, and the crystallization temperature was slightly changed.⁴⁵ In particular, hybrid eco-composites comprising recycled carbon fiber, kenaf natural fiber, and rPP showed an increase in the crystallization degree with increasing content of recycled carbon fiber.⁴⁹ It is worth mentioning that while the incorporation of fibers in rPET increases the crystallinity degree, it has a negative effect of lowering the thermal stability of the developed eco-composites.⁶² Tan et al.⁶³ reported that the treated fiber composites showed elevated decomposition temperature and reduced residue compared to the untreated fiber composites. In rTPB blends, crystallinity increases with the increase in the content of date palm leaves due to heterogeneous nucleation effect in presence of maleic anhydride grafted polyethylene (MAPE). Mixing recycled polyolefin and date palm leaves exhibited high thermal stability, low thermal conductivity, and decent strength. Therefore, they are considered as attractive materials to be used in thermal insulation applications.⁷⁵

Finally, the flaming characteristics of the eco-composites showed lower rate of burning compared to the other types of composites. For example, an enhanced performance of rPP with the inclusion of treated sisal fiber was observed.⁵⁸ Similarly, Zadeh et al.⁶⁶ reported that the eco-composites containing high concentration of $Mg(OH)_2$ particles demonstrate good flame retardation and heat resistance.

2.4 | Mechanical performance

The overall mechanical properties of eco-composites are highly affected by the reinforcing fillers since they dictate the load transfer to the polymer matrix. In general, the strength of a natural fiber depends on the intensity of crystalline cellulose biomass. Moreover, lignin facilitates an efficient mixing between the fiber and matrix during the production of eco-composites.⁵⁴ The mechanical behavior of the eco-composites can be further addressed considering the following parameters of interest:

- Filler type and its chemical composition.
- Filler geometry.
- Chemical modification of fillers.
- Polymer properties.
- Manufacturing process.
- Coupling agents and additives.

Generally, voids are generated at high filler loadings. These voids can initiate high chances of failure in weak regions of the eco-composite, which further increases the probability of separation among the constituents. Agglomerates may turn into stress concentrators that accumulate stresses in composites faster than usual. This can cause immature rupture compared to non-reinforced polymers. In contrast, the chemical treatment of fibers includes leaching out of non-amorphous biomass materials and partial removal of wax, fatty, impurities, phenol components, and other substances. As a result, a better mechanical bonding between the filler and matrix can be achieved.^{27,79} Further discussions on fiber treatment are

provided in Section 4. Common recycled thermoplastics include LDPE, HDPE, PP, and PET. Globally, the quantity of the recycled plastic remains steady, and has even started to increase. Polyolefin thermoplastics dominate a large amount of plastic waste, and is generally sent for recycling or landfill.⁸⁰ The mechanical properties of common virgin polymers that can be easily recycled are summarized in Table 2.

Indeed, our literature review show that there is a limited number of studies dedicated to characterizing the mechanical properties of the rLDPE eco-composites. Among the few studies is the work of Youssef et al.¹² Their demonstrated that the tensile strength increases with the increase in fiber percentage up to 10% and then slightly decreases. However, the tensile strength remains higher than the strength of the neat rLDPE. The developed eco-composites using bagasse particles exhibited better properties with the fiber content in the range of 20–30 wt%. For optimum conditions, the content of carbonized bagasse particles should not exceed 30 wt%. At 10 wt% of fiber content, the material exhibited enhanced tensile properties, impact energy, and toughness. Nevertheless, as the fiber content is increased, several issues relating to microstructure are encountered causing a degradation in the mechanical properties.¹⁶ Owing to the hydrophilic nature of RH, the characterized strength of RH/rLDPE decreases with the increasing natural filler content.¹⁵ However, the incorporation of up to 6% of nanosilica and 4% of nanoclay is found to be optimal. Excessive amounts of nanoparticles can agglomerate, resulting in the developments of gaps and cracks in the prepared eco-composites.¹⁴ Whereas, incorporating the rLDPE with up to 40 wt% of cocoa waste degrades the strength and elongation, and the material rigidity is increased.¹¹ Bello et al.¹³ noted that when 10% eggshell nanoparticles content was incorporated into rLDPE, maximum tensile strength was accomplished. Likewise, a content of 12% eggshell nanoparticles developed 52% higher flexural strength of nanocomposite over that of standard composite.

Figures 7 and 8 show the maximum values of the tensile strength and Young's modulus, respectively, of natural-filler-reinforced rLDPE, as reported in the literature.

TABLE 2 Average mechanical properties of the commonly used polymers (S_{UT} : tensile strength; E: Young's modulus; S_F : Flexural strength; E_F : Flexural modulus; S_I : Impact strength; SI_Z : Izod impact strength)

Thermoplastic	S_{UT} (MPa)	E (MPa)	S_F (MPa)	E_F (MPa)	S_I (kJ/m ²)	SI_Z (J/m)
LDPE ^{81,82}	10.0	112.4	9	275.8	16.7	–
HDPE ^{32,34,83,84}	21.4	950	22.4	840	3.9	47
PP ^{81,85}	36.2	4483	44.8	1430.7	–	22
PET ^{86,87}	67.3	2467	98.6	2620	9.8	16

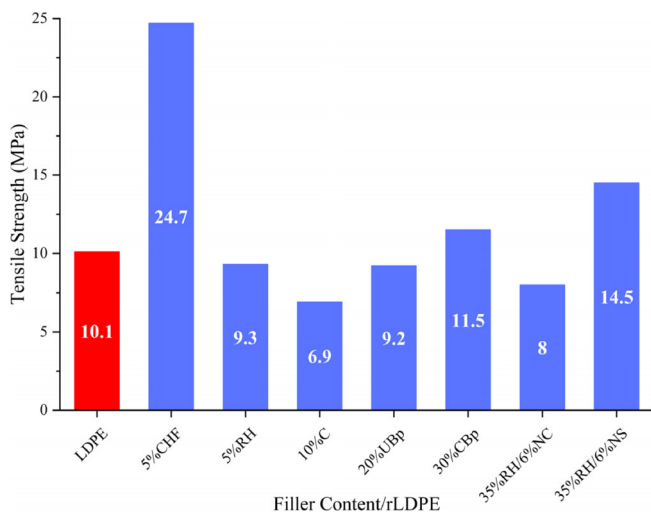


FIGURE 7 Maximum values of the tensile strength of previously reported natural-filler-reinforced rLDPE [Color figure can be viewed at wileyonlinelibrary.com]

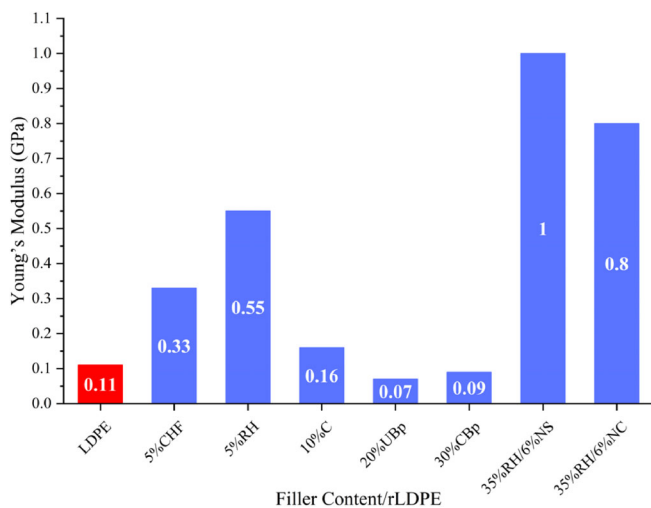


FIGURE 8 Maximum values of the Young's modulus of previously reported natural-filler-reinforced rLDPE [Color figure can be viewed at wileyonlinelibrary.com]

Results of tensile, flexural, and impact strengths showed that the mechanical behavior of the rHDPE-based eco-composites were improved compared to the neat HDPE in the presence of treated natural fillers and coupling agents. In these eco-composites, induced cracks do not initiate at the interface, but within the filler itself as the material undergoes tensile loading.³⁰ The alkali treatment (5 wt% of NaOH) had a favorable effect on the tensile and flexural properties of chestnut cupula/rHDPE.²⁰ Moreover, it was reported that hemp/rHDPE composites with 40% of fiber volume fraction exhibited excellent properties such as tensile strength, flexural strength, and elastic modulus.²⁵ The coupling agents

effectively improve the compatibility between the natural fillers and rHDPE, enhance the dispersion of fibers into the polymer matrix, and create chemical bonding's between the modified polymer and filler.^{18,30,39} The outcomes of the tensile tests showed enhanced tensile strength and modulus of elasticity for the compatibilized eco-composites compared to the un-compatibilized eco-composites. It was also found that at 6 wt% of PE-g-MAH, maximum tensile strength and Young's modulus were achieved.⁴⁰ A conflicting result for the mechanical properties of natural-fiber-reinforced rHDPE was observed.^{19,56} While the tensile strength has been degraded, a progression in the tensile modulus of developed banana/rHDPE eco-composites was observed as compared to the virgin and recycled polymers.¹⁹ Moreover, the banana fiber/fly ash/rHDPE blend increases the tensile strength, tensile modulus, flexural strength, flexural modulus, and hardness by approximately 17%, 188%, 38%, 159%, and 37%, respectively, as compared to the rHDPE matrix.¹⁸ In addition, increasing the RH fiber content in the eco-composites increases their tensile and flexural properties; however, it decreases their impact strength. The incorporation of the sisal fibers into the rHDPE matrix significantly increased the tensile and flexural moduli of the developed composites when prepared with 10 wt% of acetylated sisal fiber and original polyethylene matrix using oxidation.³⁰ In summary, improvement in the mechanical properties was driven by several aspects. First, the density of the rHDPE is marginally higher than that of the HDPE, which shows that the rHDPE has less branching, and hence, provide strong intermolecular interactions and tensile strength. Second, in the recycling process, the presence of additives can result in a better compatibility of natural filler with the rHDPE matrix, which consequently enhances the mechanical properties of the eco-composites.⁸⁸ The maximum values of the tensile strength and Young's modulus of previously reported natural-filler-reinforced rHDPE at different filler contents are shown in Figures 9 and 10, respectively.

The rPP-based eco-composites exhibited similar mechanical properties to the virgin PP-based bio-composites. Natural fillers demonstrated a remarkable compatibility with the polypropylene matrix owing to good dispersion and fiber-matrix bonding.^{50,57} Rosa et al.⁴⁰ reported that the eco-composite with 1% of almond tree leaves of steam-exploded fibers exhibited an increase in tensile strength, which implies that the leaves imparted a strengthening effect to the eco-composites. It was also found that the properties of rPP were enhanced using date palm wood flour as a reinforcing agent. The best eco-composite performance was achieved with the incorporation of as low as 5 wt% of glass fiber to wood-flour-reinforced rPP. It increased the tensile

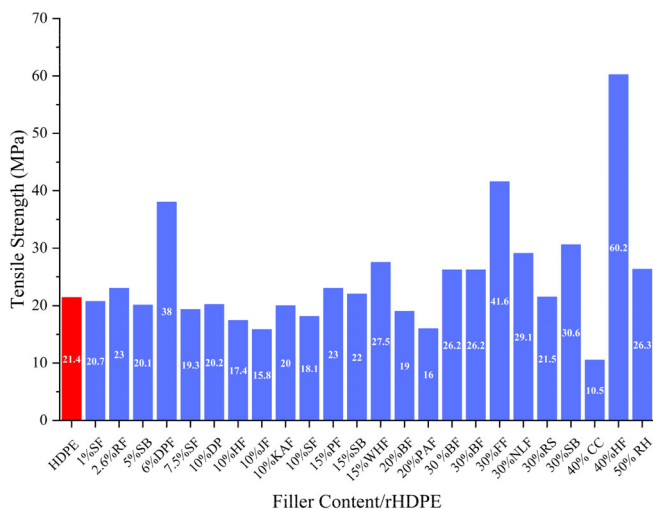


FIGURE 9 Maximum values of the tensile strength of previously reported natural-filler-reinforced rHDPE [Color figure can be viewed at wileyonlinelibrary.com]

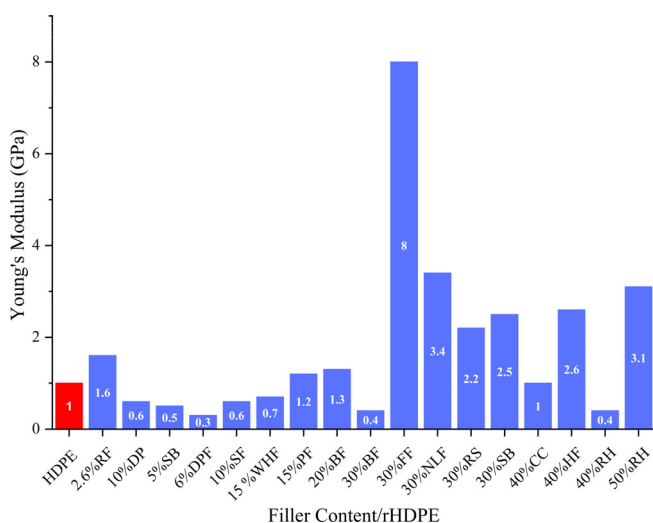


FIGURE 10 Maximum values of the Young's modulus of previously reported natural-filler-reinforced rHDPE [Color figure can be viewed at wileyonlinelibrary.com]

strength by approximately 18% compared to the wood flour reinforcement alone.⁴⁵ The tensile strength of recycled carbon fiber/rPP is increased with the increasing MAPP content (53–78 MPa); however, it is reduced (28 MPa) upon hybridization with the kenaf fiber.⁴⁹ In addition, it was reported that low fiber content (10 wt%) results in enhanced tensile strength properties compared to high fiber content (50 wt%). Oil palm biomass fibers showed excellent characteristics and therefore are considered as ideal filler reinforcements in plastics.⁵⁴ The mechanical properties of the rPP-based eco-composites with treated sisal fiber in the presence of MAPP compatibilizer were better than those of the neat rPP.⁵³ However, the alkali treatment of the curaua fiber slightly

improved the fiber–matrix adhesion, while the eco-composites with 20 wt% of untreated curaua fibers exhibited enhanced mechanical properties.⁴⁴ Tensile tests were conducted on woven jute-fiber-reinforced rPP to optimize the processing parameters such as pressure and temperature. The pressure was found to be the most significant parameter for the tensile properties.⁴⁸ An improved strength of 60 wt% of pine-nutshell-filled rPP is achieved by using C=C bonds and N=C=O groups as compatibilizers, which can effectively enhance the interfacial compatibility and mechanical stability of hydroxyl-rich biomass residues when blended with plastic.⁴³ Similarly, another study reported a significant enhancement in the mechanical properties of snail shell/rPP composites after incorporating dioctyl phthalate (plasticizer) and zinc stearate (stabilizer) compared to the neat rPP matrix.⁶⁰ An experimental work revealed that in comparison to a neat rPP sample, the tensile strength of banana fiber/rPP composites improves at 30 wt% of banana fiber loading, while a maximum flexural strength is attained at 10 wt% of banana fiber loading.⁴² In general, it can be concluded that rPP is a valuable material for producing natural fiber composites with better mechanical properties. A fire-retardant nanocomposite fabricated from rPP, kenaf fiber, and nano-CaCO₃ was successfully processed in a melt state. The tensile strength of the developed nanocomposite was marginally decreased compared to the baseline composite without nanoparticles. However, in both cases, an improved mechanical performance compared to the neat rPP was noticed.⁵¹ Yu et al.⁶¹ studied the influence of nanofillers including wheat straw and inorganic fillers as reinforcing components to rPP. The flexural modulus and flexural strength of manufactured hybrid nanocomposites were both reduced when reinforced with three kinds of inorganic fillers; heavy calcium carbonate, silicon dioxide, and fly ash which was induced by poor interphase adhesion. Idumah et al.⁵² developed novel kenaf/graphene nanoplatelets/rPP nanocomposites in terms of mechanical characteristics. The outperformance as reflected by high strength was ascribed to the improved interfacial adhesion because of kenaf flour modification, efficient stress transfer between matrix, micro-fibrous fillers and graphene nanoplatelets, in addition to homogeneous and uniform dispersion of graphene nanoplatelets in matrix. The maximum values of the tensile strength and Young's modulus of previously reported natural-filler-reinforced rPP are shown in Figures 11 and 12, respectively.

Only a few studies on reinforcing rPET and UP-rPET with natural fillers have been reported. Some studies found that the eco-composites with alkali- and/or silane-treated fillers show better mechanical properties than those with untreated fillers. The most promising results were achieved when the fruit bunch fibers were

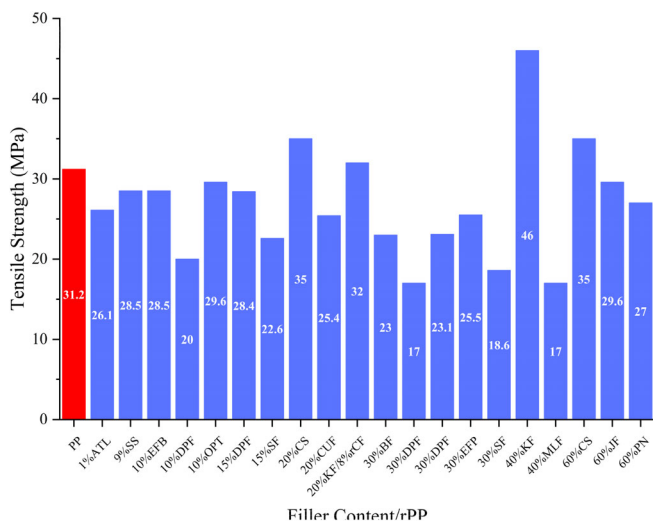


FIGURE 11 Maximum value of the tensile strength of previously reported natural-filler-reinforced rPP [Color figure can be viewed at [wileyonlinelibrary.com](#)]

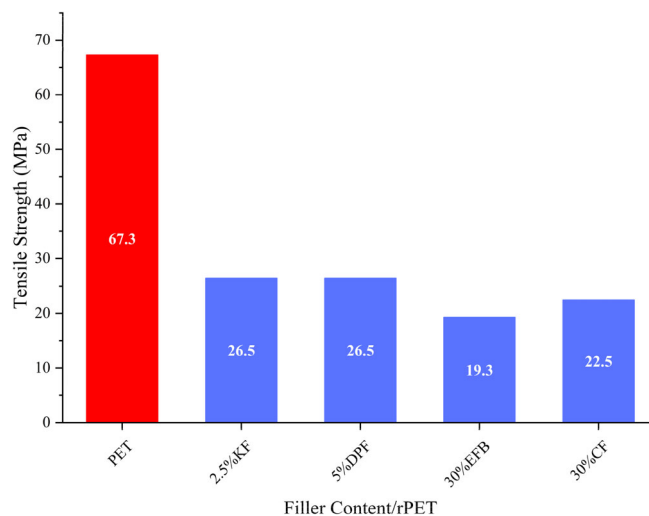


FIGURE 13 Maximum values of the tensile strength of previously reported natural-filler-reinforced rPET [Color figure can be viewed at [wileyonlinelibrary.com](#)]

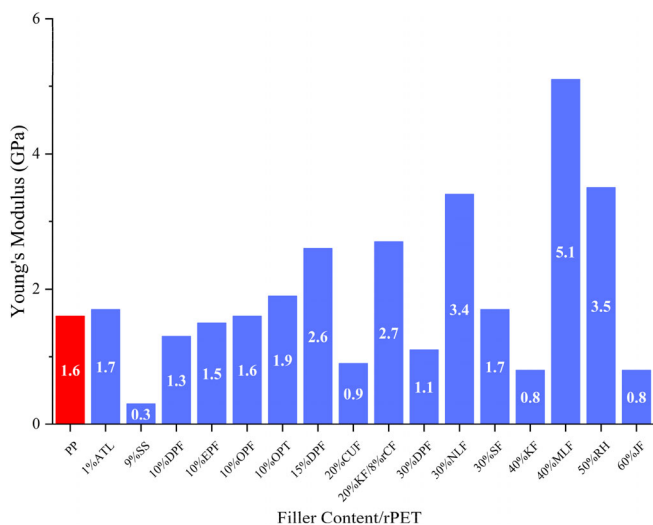


FIGURE 12 Maximum value of the Young's modulus of previously reported natural-filler-reinforced rPP [Color figure can be viewed at [wileyonlinelibrary.com](#)]

sequentially treated with silane MA, and NaOH, and their tensile strength increased by approximately 21%, 18%, and 13%, respectively.⁶³ Similarly, tensile and impact strengths of the coconut fiber/UP-rPET-based eco-composites were optimized in the case of treatment of 0.5% of silane over the case of treatment of 5% of alkali-treated coconut fiber/UP-rPET eco-composites.^{68,69} It was shown that alkali treatment enriches the interfacial adhesion through increased mechanical interlockings, resulting in enhanced resin–fiber interaction at the inter-phase region. Moreover, it was concluded that the alkali treatment enhanced the impact strength of the eco-composites containing treated kenaf fiber/UP-rPET.⁷⁰

The maximum values of the tensile strength and Young's modulus of previously reported natural-filler-reinforced rPET are shown in Figures 13 and 14, respectively.

Eco-composites with untreated and treated fibers showed significant difference in flexural strength compared to the unfilled polymer matrix. In the case of rHDPE resin, the corresponding eco-composites exhibited enhanced moduli and strengths compared to the eco-composites in the case of virgin HDPE, provided the modifiers are appropriately used during processing.³² The enhanced fiber adhesion to the rHDPE, particularly in the case of alkali-treated DPFs, was the primary factor for improving mechanical strength.²⁷ For example, the alkali-treated DPFs enhance the tensile properties of the developed bio-composites at optimized fiber loading.⁴⁷ It was experimentally found that the addition of the rHDPE to the virgin HDPE enhances the tensile strength by 12.9% and flexural strength by 47.5%.⁷⁴ It has been proven that all chemically treated RH form stronger adhesion with polymer matrix. Flexural strength of the bio-composites filled with ultraviolet-ozonolysis (UV/O₃)-treated RH demonstrated a similar trend compared to that of the bio-composites filled with alkali-treated RH. Therefore, UV/O₃ treatment can be considered as a substitute method for modifying the RH surface to enhance the adhesion between the hydrophilic RH fiber and hydrophobic rHDPE matrix.²⁹ In contrast, the flexural and tensile strengths of hemp fiber composite decrease with increasing the hemp content from 10 to 30 wt%.⁸⁹ Moreover, hybridization of such composites by replacing the recycled carbon fiber with kenaf fiber, was reported to reduce the bending strength of the composites from 274.1 to 114.3 MPa.⁴⁹ The incorporation of small-size RH particles (<0.5 mm) enhances the composite properties such as viscosity, flexural strength,

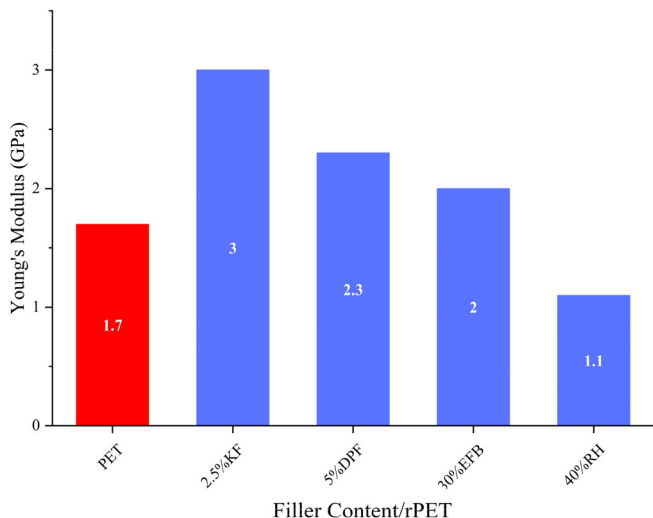


FIGURE 14 Maximum values of the Young's modulus of previously reported natural-filler-reinforced rPET [Color figure can be viewed at wileyonlinelibrary.com]

moduli, water absorption, and thermal stability of rHDPE composites.³¹ Moreover, sisal/rHDPE composites show 22% increase in their Izod impact strength compared to the pure HDPE.³⁷ It was also found that the sisal fibers were not effective in enhancing the flexural strength; however, it enhanced the Izod impact strength. The ductility of the material is reduced with increasing fiber loading. The incorporation of nanoparticles reduced the Izod impact strength and increased the hardness.¹⁴ However, a notable improvement in the impact strength of the composites was observed upon the incorporation of 20% of kenaf fiber.⁶⁷ Overall, the flexural properties of the rPP eco-composites were comparable to those of the composite with virgin PP. Figures 15–17 summarize the flexural strength, flexural modulus, and impact strength, respectively, of various reported eco-composites. The flexural strength in most cases has been improved compared to the neat polymer, particularly at the fiber loading of 5–20 wt%. Beyond this limit, a degradation in flexural strength is observed. The maximum strength was found in the case of PET where the strength was enhanced by five folds. In general, the improvement in the flexural strength was more pronounced than that in the tensile strength. Flexural modulus demonstrated a similar trend for several types of eco-composites.

3 | FRAMEWORK OF HIGH-PERFORMANCE ECO-COMPOSITES PRODUCTION

In recent years, eco-composites have emerged as competitive materials in many industries owing to their tailored

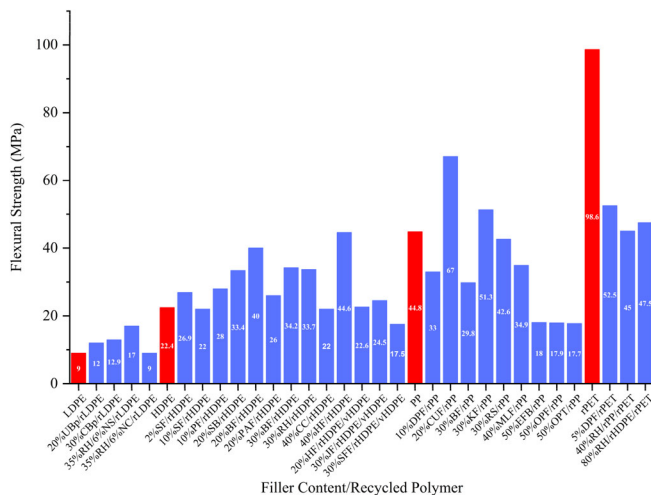


FIGURE 15 Maximum values of the flexural strength of previously reported natural-filler-recycled polymers [Color figure can be viewed at wileyonlinelibrary.com]

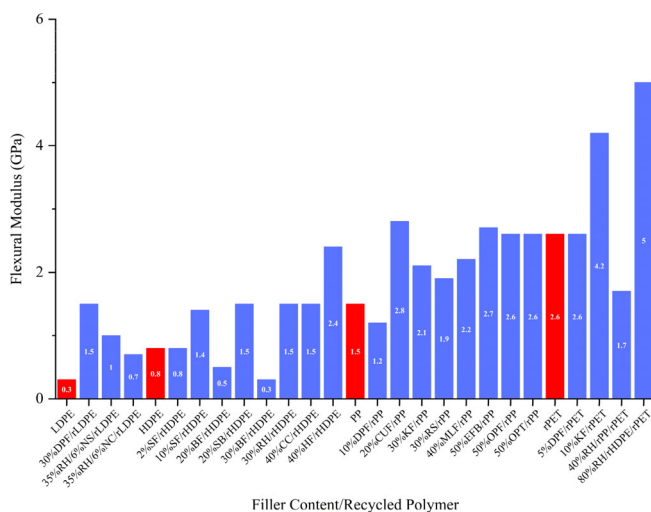


FIGURE 16 Maximum values of the flexural modulus of previously reported natural-filler-recycled polymers [Color figure can be viewed at wileyonlinelibrary.com]

properties and cost competitiveness. Quality and performance are rigorously evaluated to widen their applications. At present, the applications of eco-composites are limited to non-structural products and components. This is mainly due to the low values of strength of such materials regarding design requirements. Therefore, further comprehensive investigation and research are required to enhance their performance up to the mark of structural requirements. In this regard, two major issues should be highly considered: filler/polymer compatibility and production method. These two issues are addressed in the following subsections.

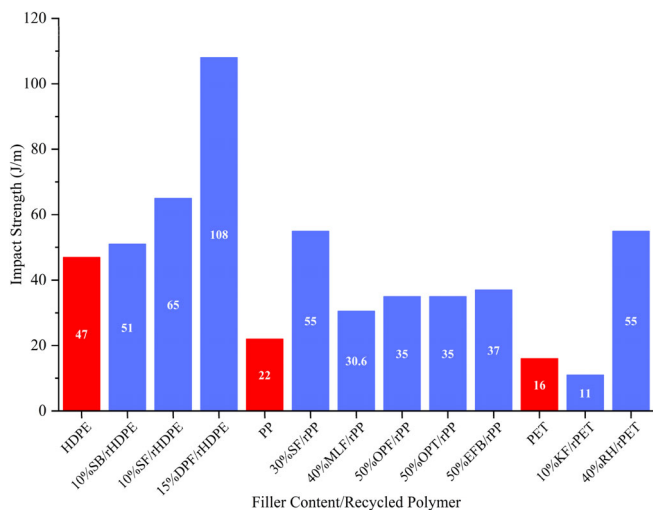


FIGURE 17 Maximum values of the Izod impact strength of previously reported natural-filler-recycled polymers [Color figure can be viewed at wileyonlinelibrary.com]

3.1 | Natural fiber composition and treatment

Natural fillers are produced from natural sources and are classified mainly in two categories: fibers and particles. It is well recognized that the natural fillers, in the form of flour, fibers, pulp, lignin, hemicellulose, pectin or cellulose, are suitable as reinforcements for polyolefins and other types of polymers.^{90–94} Table 3 summarizes the average chemical composition and mechanical properties of the natural fibers that are currently used in the production of eco-composites.

Better control of the microstructural features and biochemical character of the natural fibers are essential in selecting the appropriate manufacturing process for eco-composites.¹¹⁴ The main components of the plant fibers are cellulose, hemicellulose, lignin, pectin, and waxes. However, cellulose is considered as a natural polymer and its crystallinity is a vital factor that influences the bonding among microfibrils, which provide structural integrity to the natural fiber. Hemicelluloses, another essential component of the plant cell wall, are mainly responsible for providing secondary strength and moisture absorption. Lignin, which is a complex hydrophobic polymer, provides a backbone rigidity to the plants and prevents water penetration inside the fiber.¹¹⁵ Furthermore, it is also responsible for the resistance against a microbial attack and degradation, as it assists in binding cellulose and hemicellulose together.¹¹⁶ Pectin are known to provide flexibility to the plants. Waxes are the mixture of various alcohols and oils, which cover the fiber surface for protection.^{117,118} Figure 18 illustrates the chemical structure of the main components of plant fiber biomass.

A natural fiber is characterized as a group of cells with relatively small diameter compared to the length.¹¹⁹ The inclusion of the natural fibers to strengthen the recycled polymers is mainly dependent on the intrinsic strength of the fiber. In addition, the geometry, defects, inconsistency, crystallinity, and structure of the fiber are other critical factors responsible for the mechanical behavior of the fiber. It is well-established that the morphology, mechanical properties, and chemical composition of the fibers can be significantly controlled and improved through chemical treatment.¹²⁰ However, natural fibers have their own limitations. For example, the pendant polar groups in natural fillers stimulate water absorption, which contributes to poor interaction between the fiber and polymer, thereby limiting its mechanical strength. Substitution of hydrophilic functionalities by hydrophobic groups overcome the moisture absorption problem and facilitate the polar group in forming strong chemical bonds with the polymer.¹²¹ The inconsistency in quality and performance of the natural fibers as compared to the synthetic fibers hinders its applicability and commercialization. This inconsistency is due to the variability in the chemical structure of a plant, processing methods, and equipment used for the production of fibers.¹²²

The main aspects affecting the interfacial bonding between the fiber and matrix are the molecular attractive forces, chemical bonds, and mechanical interlocking. Basically, the hydrophilic nature of the plant fibers is not intrinsically compatible with the hydrophobic polymers. Furthermore, the pectin and wax constituents that form a layer around the fiber turn to be a hinder to mechanical interlocking with nonpolar polymer matrix. The existence of a large amount of hydroxyl groups delays its reaction with the matrix.¹²³ Consequently, to obtain an optimal mechanical interlocking and bonding, the natural filler and polymer matrix should be both chemically modified. As a result, the reinforcing fillers become capable of functioning as load-carrying elements, which provide strength and rigidity to the produced eco-composites. The polymer matrix maintains the fibers alignment (position and orientation) and protect them against any harsh environmental conditions.¹²⁴ In addition, the fiber treatment enhances the filler processability by cleaning its surface from impurities.

There are several methods for preparing natural fillers. The pre-treatment processes can be classified into three main categories: (a) abiotic (chemical, physical, and mechanical processes), (b) biotic (enzymatic and microbiological processes), and (c) multi-coupled (thermochemical and reactive extrusion processes).¹²⁵ New methods such as steam explosion⁴¹ and plasma treatment^{26,126} have been developed to effectively expand the

TABLE 3 Chemical composition and mechanical properties of the natural fibers used in the production of eco-composites

Natural fiber	Chemical composition (%)			Mechanical property			
	Cellulose	Hemicellulose	Lignin	Density (g/cm ³)	Tensile strength (MPa)	Elastic modulus (GPa)	Elongation at break (%)
Almond Leaves ^{95,96}	38.47	28.82	29.54	1.07	126.6	4.4	2.2
Banana ^{18,97}	64	19	5	1.3	355	33	5.3
Chestnut Cupula ⁹⁶	21.47	16.28	36.58	–	–	–	–
Cocoa ⁹⁸	30.79	21.09	25.55	–	–	–	–
Coconut/Coir ^{99,100}	30.58	26.7	33.3	1.2	175	4.0–6.02	30
Corn Husk ^{101,102}	31–39	34–41	2–14	1.254	160.49	4.57	21.08
Corn Straw ⁴³	39.38	21.76	20.20	–	–	–	–
Curaua ^{103,104}	70.7	21.1	11.1	1.4	543	63.7	1
Date Palm ¹⁰⁵	46	18	20	0.92	170–275	5–12	5–10
Durian Peel ¹⁰⁶	35.6	18.6	10.7	–	–	–	–
Flax ^{107,108}	64.1–71.9	64.1–71.9	2.0–2.2	1.5	500–1500	27.6	2.7–3.2
Hemp ^{107,108}	70.2–74.4	17.9–22.4	3.7–5.7	1.47	690	70	2.0–4.0
Jute ^{97,107}	45–53	18–21	21–26	1.3	393–773	26.5	1.5–1.8
Kapok ¹⁰⁹	35	22	21.5	–	–	–	–
Kenaf ^{107,108}	45–57	8–13	21.5	1.45	930	53	1.6
Newbouldia Laevis ¹¹⁰	23.88	11.06	15.93	–	–	–	–
Oil Palm ^{107,108}	65	17.5	10.12	0.7	248	3.2	2.5
Piassava ¹⁰⁸	28.6	25.8	45	1.4	134–143	1.07–4.59	7.8–21.9
Pine Nutshell ^{43,111}	45.73	18.8	23.72	–	–	–	–
Pineapple ¹¹²	68.5	18.8	6.04	1.52	413–1627	34.5–82.5	1.6–3
Quill Feather ¹¹³	–	–	–	0.9	100–203	3–10	6.9
Reed ²⁸	43	20	30.5	–	–	–	–
Rice Husk ^{97,108}	29.7–35.6	11.9–29.3	15.4–20	0.5	–	–	–
Rice Straw ⁹⁷	29.2–34.7	23–25.9	17–19	–	–	–	–
Sisal ^{107,108}	78	10	8	1.5	511–635	9.4–22	2.0–2.5

interfacial adhesion between the natural fiber and hosting matrix. Ultraviolet treatment (UV) is an efficient method that can increase the mechanical strength of the natural fiber composites. Chemical procedures can eliminate major part of lignin and hemicellulose from the fiber, thus resulting in stronger hydrogen bonds and better mechanical characteristics.^{126,127} Filler treatment should ensure an optimal removal of the outer layers and impurities, including the wax residing at the internal layer of filler surface. In fact, most of the reported studies have focused on modifying the surface topography of the fillers through partial removal of the impurities and waxes at the outer layer. Consequently, coupling agents and compatibilizers applied on the polymer surface will not be highly effective in forming chemical crosslinking with the functional group on the filler surface. This is

because the other functional groups residing in the internal layer of the plant cell are still covered by the cell wall (wax) and impurities. For achieving better results, microwave-assisted chemical treatment has been used to effectively accelerate the purification of the natural fillers and modify the cell-wall structure.^{128–130} A newly invented multi-step method based on an accelerated microwave-assisted heating process has been proven to successfully remove the non-cellulosic content, enhance the surface dewaxing and degumming, increase crystallinity, reduce moisture sensitivity, and avoid fiber degradation with reasonably high tenacity and elongation at break, as reported in a recently published patent.⁹⁴ Other advantages of the method include time, cost-effectiveness, and clean modification process. Figure 19 shows the effects of various treatment conditions on the natural filler surface extracted from date palm

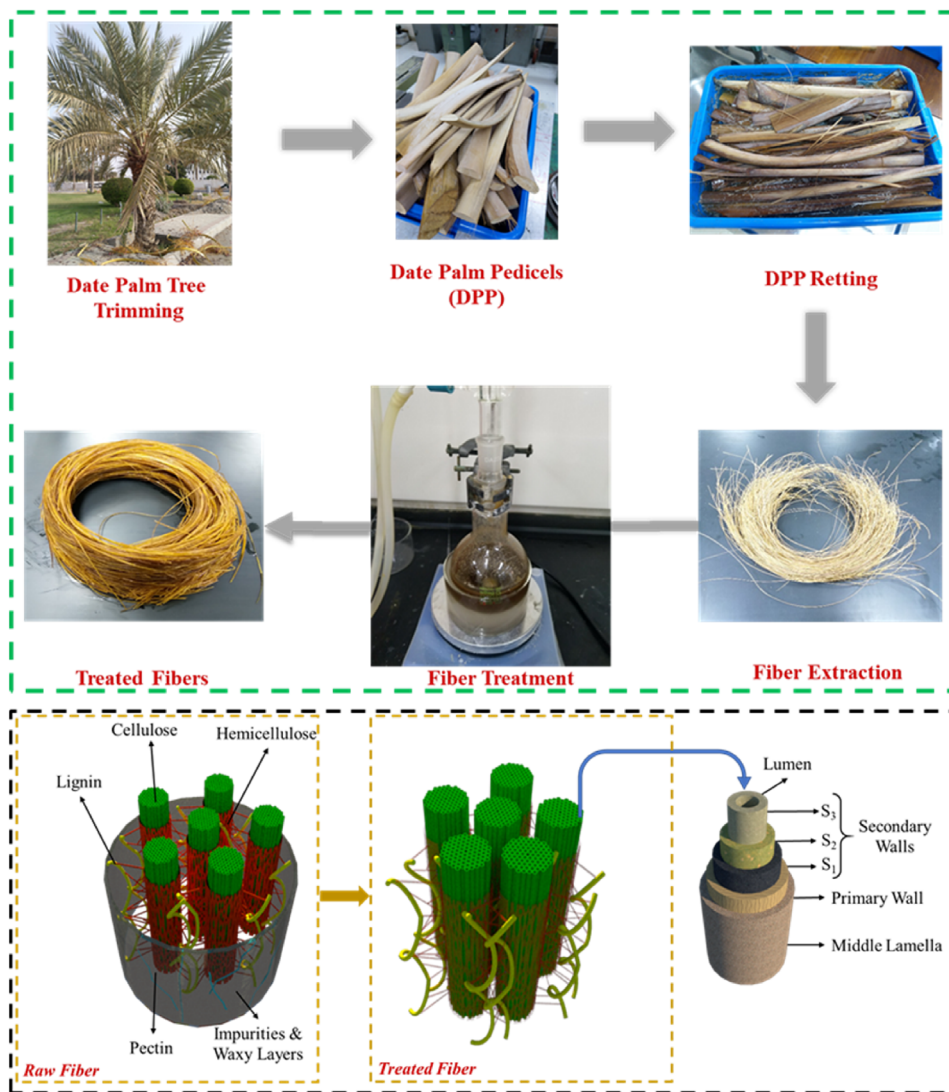


FIGURE 18 Structure of lignocellulosic biomass with cellulose, hemicellulose, and lignin [Color figure can be viewed at wileyonlinelibrary.com]

pedicels. Figure 19(a) shows the waxy layer on the untreated filler surface, which is not completely removed by alkali-treated filler using conventional heating, as indicated by the surface morphology in Figure 19(b). However, Figure 19(c), (d) confirm the removal of the waxy layer using the microwave-assisted heating method. The microwave-assisted dewaxing was validated by the disappearing of the peaks corresponding to wax and pectin in the untreated spectra at $\sim 1726 \text{ cm}^{-1}$, as shown in Figure 20. These findings suggest that the microwave-assisted treatment effectively increases the exposure of the functional group ($-\text{OH}$) on the filler surface, which enhances the filler/polymer chemical interaction and interlocking because the polymer can spread and occupy the internal voids at the fiber surface, as depicted in Figure 19(d).

The characterization of natural fibers can also be performed using solid-state nuclear magnetic resonance (ssNMR) for the assignment of chemical group signals, X-ray photoelectron spectroscopy (XPS) for

chemical group binding energy, SEM for surface morphology, and atomic force microscopy (AFM) for surface topography. The ssNMR of raw flax natural fibers as reported by Bourmaud et al.¹³¹ revealed common functional groups at 105, 88, 83, 74, 73, 65, 56 ppm for cellulose and 21, 33, 153 ppm for hemicellulose and lignin. Martins et al.¹³² investigated the effect of different treatment methods based on the NMR spectra (Figure 21). The signals at 21 and 174 ppm due to acetyl groups of hemicellulose and the signals in the lignin aromatic region of 110–159 ppm disappeared after the mercerization treatment, indicating full removal of hemicellulose and partial removal of lignin. However, the signals at 21 and at 174 ppm remained after the acetylation treatment. These two signals can be attributed to the carbon of the methyl group and the carbon of the carboxylic group from the acetyl group, respectively, which provide evidence of the productive acetylation treatment.

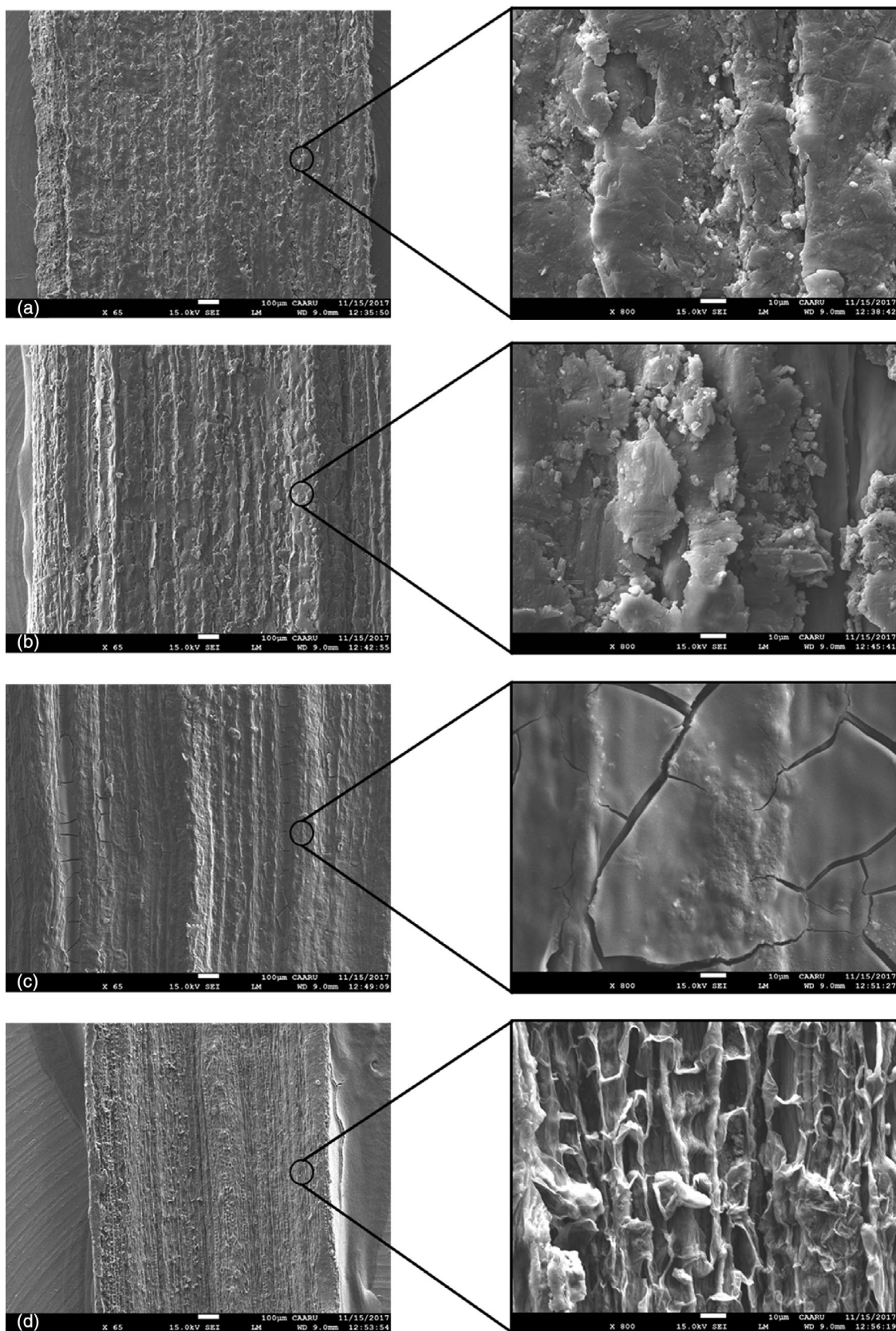


FIGURE 19 SEM micrographs of date palm leaf surface under different treatment conditions: (a) untreated fibers, (b) conventional heating, NaOH (5 wt%), (c) microwave-assisted heating, NaOH (5 wt% NaOH, 900 W, 5 min), and (d) microwave-assisted heating, NaOH (5 wt% NaOH, 900 W, 15 min)

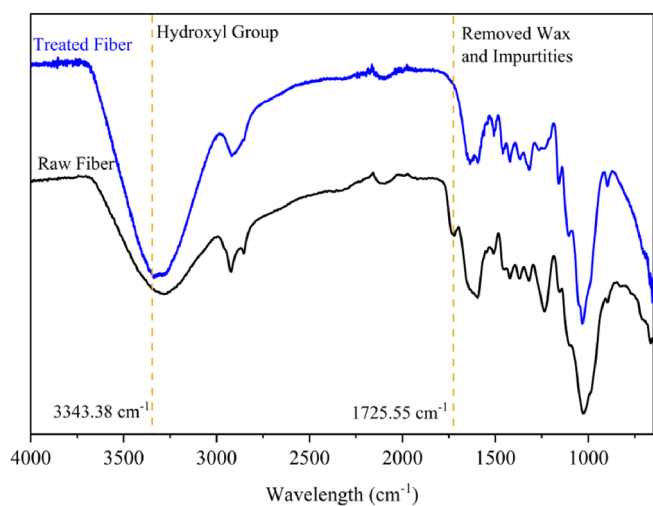


FIGURE 20 FTIR spectra of raw and treated date palm fibers [Color figure can be viewed at wileyonlinelibrary.com]

XPS analysis was performed on DPF which was functionalized following efficient methods that use azide and alkyne functional groups,¹³³ as shown in Figure 22. Generally, cellulose contains four different types of carbon bonds: C1, which represents carbons attached to other carbons or hydrogen; C2, which involves carbon bound to one oxygen atom, C3, which includes carbons attached to two oxygen atoms; and C4, which is carbon from the carboxyl group.

SEM and AFM images of a single flax fiber depicted in Figure 23 suggest that the morphology of the fiber is altered after the chemical treatment.¹³⁴ Before the mercerization treatment, the surface of the raw fiber appears to be covered by an envelope (thin film) of an amorphous polymer. These particles disappeared entirely upon treatment; the fibrils can then be observed by SEM and AFM.

Fiber treatment is an effective technique to modify the surface topography, and it is a crucial strategy to enhance the fiber properties. Table 4 summarizes the effect of the mercerization treatment on the tensile properties of natural fibers due to the rearrangement of the cellulosic microfibrils and partial removal of amorphous biomass such as pectin and hemicellulose. In some cases, the fiber's properties are degraded after the treatment, which is a result of fiber structure fibrillation and damage. Hence, careful control of the treatment parameters is crucial to avoid such damage.

Different modification processes can realize effective utilization of cellulosic ingredients that are derived from natural agro-residue and used as a bio-filler in recycled

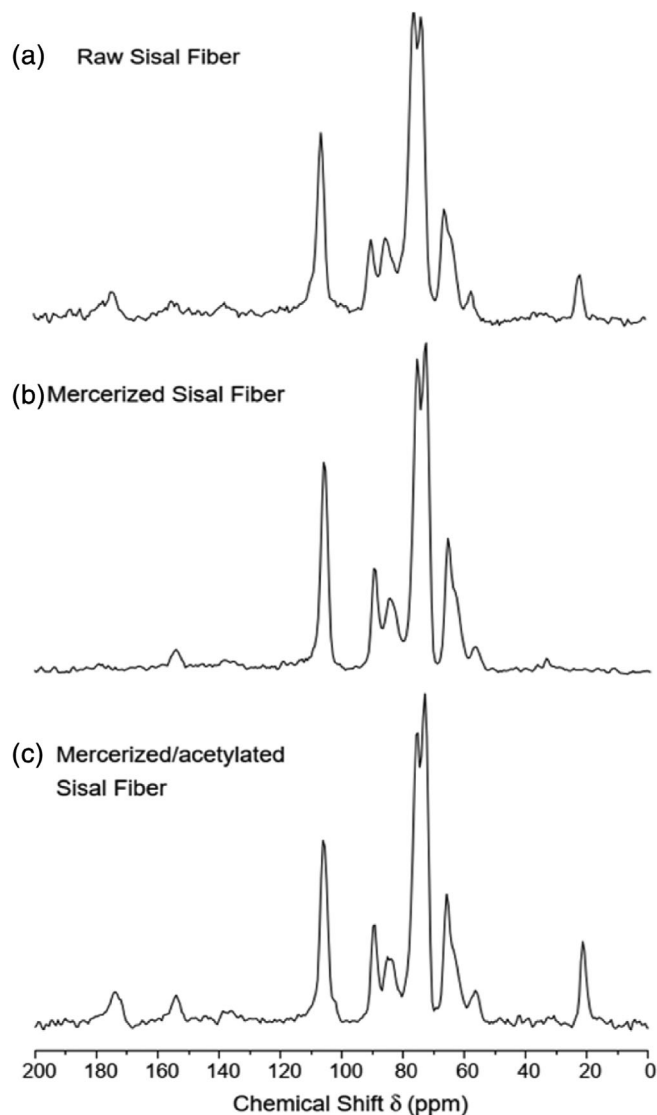


FIGURE 21 ¹³C CP-MAS/VACP solid-state NMR spectra of (a) raw sisal fiber, (b) mercerized sisal fiber, and (c) mercerized and acetylated sisal fibers.¹³²

polymers. The following provides further insight to these processes.

1. *Surface treatment*: to enhance the surface characteristics of fiber and extract the wax, impurities and the non-cellulosic biomass. If the non-cellulosic biomass is fully extracted, the fiber structure will be damaged, and the final product will be in the form of pulps or particles. Hence, the extraction process should be carefully controlled and optimized such that the integrity, structure, and characteristics of the modified fibers are maintained.
2. *Delignification*: to produce pure cellulose from cellulose-rich fillers. This can be the best solution for

cellulosic waste utilization, including waste from peels, leaves, husk, and other food waste. In fact, the regenerated cellulose was used as reinforcement, which exhibited an improvement in the mechanical properties of the cellulose-based eco-composites.¹⁴⁴

3. *Hydrolysis*: to produce nano-crystalline cellulose and cellulose whiskers, which serve as potential fillers for nanocomposites.

3.2 | Functionalization of polymers and natural fillers

Several works have focused on the enhancement of filler/polymer adhesion by modifying molecules through chemical or physical techniques.^{145–147} Mechanical interlocking can be established through proper filler surface modification. A bio-filler is polar in nature due to the presence of the hydroxyl group on the biomass surface, particularly on the cellulose molecules. This hydroxyl group can be utilized to produce more reactive chemical groups on the natural fiber surface. To develop high-performance eco-composites with adequate compatibility among gradients, functionalization of polymer is performed using different methods, including coupling agents and compatibilizers. Findings indicate that combining the chemical modification of the fiber and irradiation of the polymer produce eco-composites with enhanced compatibility than that obtained through only chemical modification. The irradiated eco-composites containing treated natural fibers exhibited

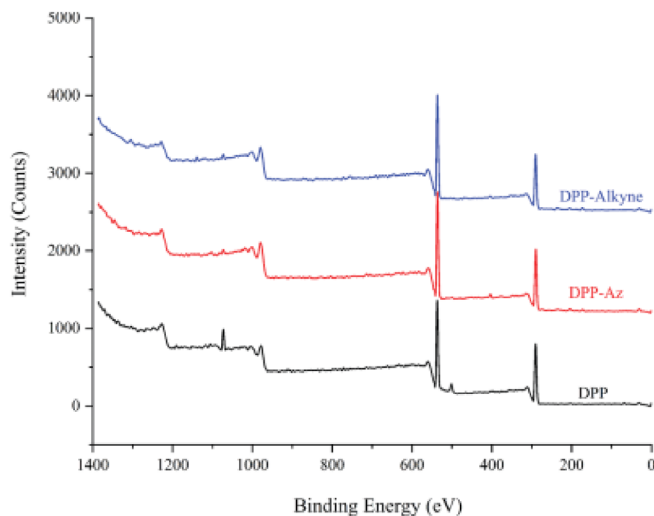


FIGURE 22 XPS spectra of treated natural fiber (date palm) [Color figure can be viewed at wileyonlinelibrary.com]

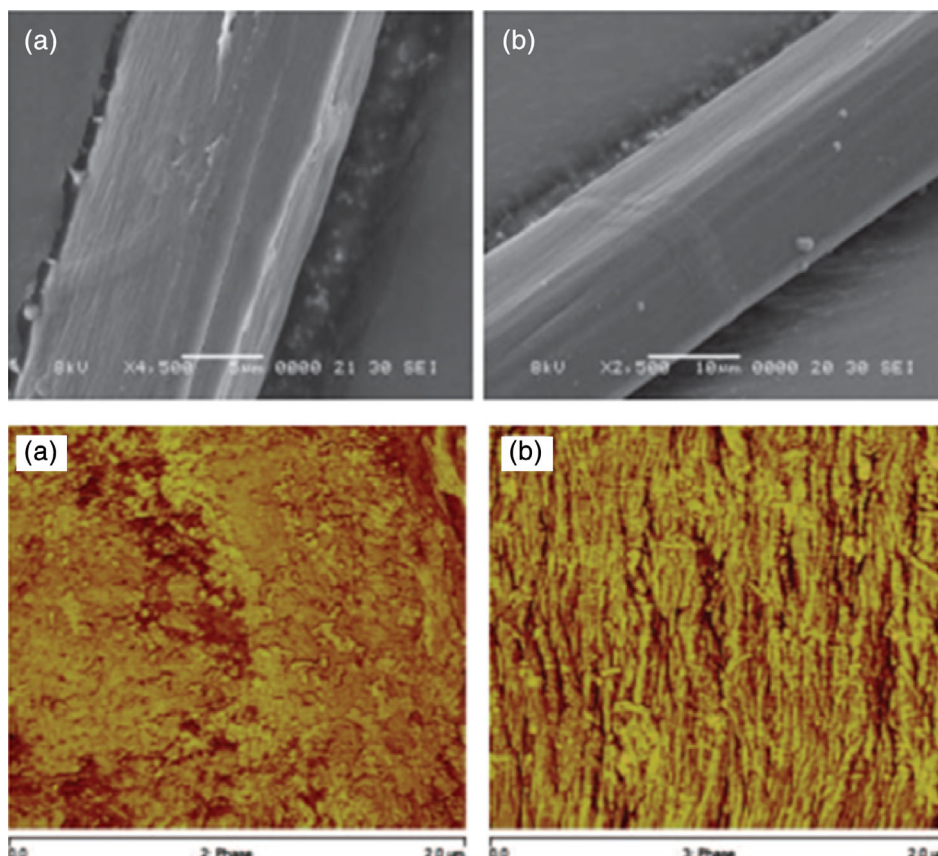


FIGURE 23 SEM and AFM phase images of an elementary (a) raw flax fiber and (b) 10% NaOH treated flax fiber¹³⁴ [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 4 Effect of various treatment methods on the tensile strength of natural fibers

Fiber type	Treatment procedure	S _{UT} (MPa)	
		Untreated	Treated
Date Palm Pedicles ⁹⁴	Ethanol/Water; Acidified Sodium Chloride; NaOH	80	444
Date Palm Rachis ¹³⁵	2% NaOH	174.2	242.6
Date Palm Mesh ¹³⁶	1% NaOH	176	310
Abaca ¹³⁷	5% NaOH	755	847
Banana ¹³⁸	5% NaOH	780.3	536.2
Coconut ¹³⁹	20% NaOH	186.4	280.9
Flax ¹⁴⁰	5% NaOH	630	627
Husk ¹⁴¹	5% NaOH	108.8	135.2
Husk ¹⁴¹	Alkali-KH570	108.8	154.9
Sisal ¹⁴²	2% NaOH	283.5	381.5
Sugarcane ¹⁴³	5% NaOH	169.51	204.5

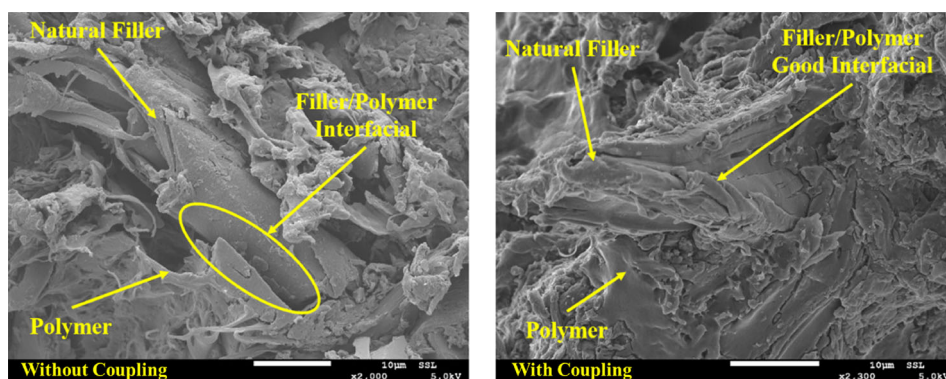


FIGURE 24 SEM micrograph of date palm flour reinforced polypropylene with and without coupling agent [Color figure can be viewed at wileyonlinelibrary.com]

enhanced mechanical properties, lower water up-hold, and higher thermal stability.^{38,148}

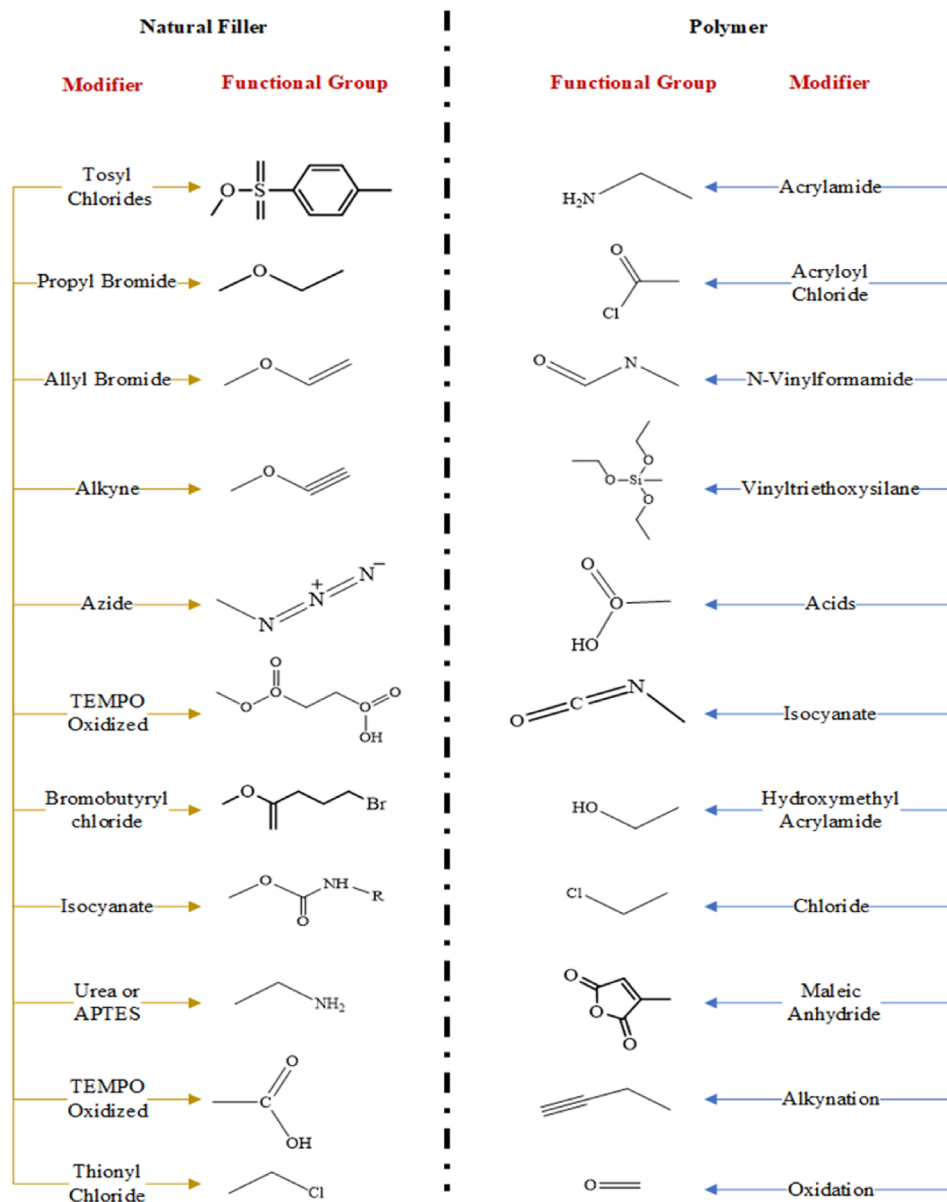
Most of the common polymer resins have low surface free energy and lack polar functional groups, thereby resulting in inherently inadequate adhesion properties. Thus, adhesion agents are included to develop better adhesion of polymers with mating components such as fillers or other polymers (Figure 24). Furthermore, repelling forces while mixing pose a challenge toward achieving a homogeneous distribution and well-blended ingredients. Similarly, coupling agents, either reactive or non-reactive, promote compatibility and reduce the interfacial tension among polymer and natural fillers.

Polymer surface functionalization can be performed either in a solid phase or melt phase in the presence of the functional chemical groups and an initiator (free radical reaction catalyst). The possible methods for polymer surface modification are summarized in Figure 25 (right column). For natural fillers, the most common forms of

surface modifications, such as surface purification and cleaning, enhance the surface topography and expose the hydroxyl groups at the filler surface.¹⁶⁵ Moreover, substitution of the hydroxyl groups with different reactive chemical group helps in the filler reactivity with the polymer as well as enhancing the filler hydrophobicity, particularly when superhydrophobic groups are utilized.¹⁶⁶ Normally, the chemical functionalization of the filler is performed prior to mixing. The possible methods for natural filler chemical reactivity enhancement are summarized in Figure 25 (left column).

If the chemical reaction occurs among the composite's ingredients, the intermolecular crosslinking and bonding between the fiber and polymer will be initiated. Therefore, the overall properties of the developed eco-composites are expected to be enhanced depending on the filler surface and chemical bonding type. By selecting a proper combination of the functional groups from the listed methods of surface modification of the natural filler

FIGURE 25 Schematic of the chemical methods for natural filler and polymer functionalization.^{133,149–164} [Color figure can be viewed at wileyonlinelibrary.com]



and polymer, as shown in Figure 25, various types of chemical bonding can be developed and used in the production of high-performance eco-composites. Strong filler-polymer crosslinking activates load transfer and enhances the mechanical and water repellent properties.^{166,167} Figure 26 presents two examples of developed natural filler/polymer chemical crosslinking. In the first example (Figure 26(a)), only the polymer surface is functionalized with a reactive group that can react with the hydroxyl group on the filler surface. In this case, it is recommended to treat the natural filler surface to expose the hydroxyl groups, which are covered by the waxy layer on the filler cells. The most common chemical groups for this approach are maleic anhydride (MAH) and acids such as acrylic acid that react with the filler hydroxyl group during melt mixing to form “ester bonds.”^{149,170–174}

Another similar and novel method that is not widely used is the functionalization of the polymer surface using an isocyanate group. This group is highly reactive with the hydroxyl group and capable of forming urethane bonds between the bio-filler and the mating polymer, as shown in Figure 26(a).^{168,175} The second example, shown in Figure 26(b), is based on a study by Yeo et al.,¹⁶⁹ which aimed to modify both the bio-filler and the polymer by amine bonding and MAH, respectively, to produce polypropylene-graft-MAH anchored lignin via the amidation reaction. This type of bonding strengthens the interfacial adhesion between the filler particles and counterpart matrix and significantly enhances the particle distribution in the matrix, which in turn results in a significant tensile strength enhancement.

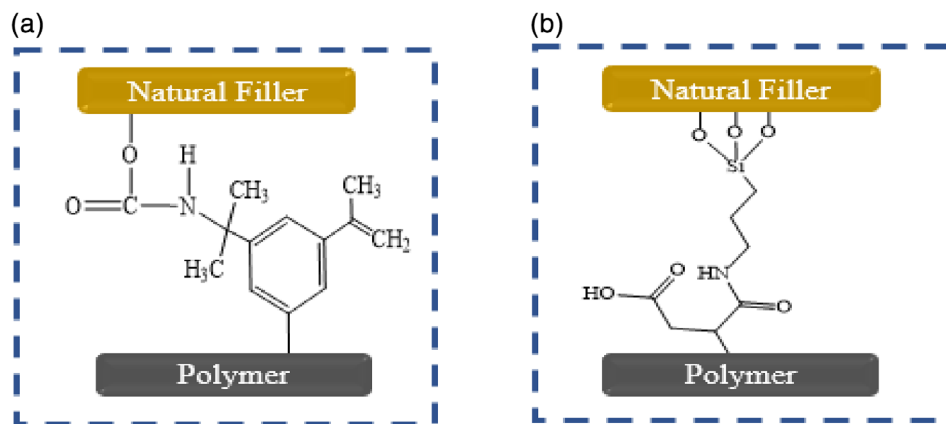


FIGURE 26 Natural filler-polymer chemical crosslinking: (a) filler and functionalized polymer,¹⁶⁸ (b) functionalized filler and functionalized polymer.¹⁶⁹ [Color figure can be viewed at wileyonlinelibrary.com]

3.3 | Production framework of eco-composites

Critical issues related to eco-composite processing should be carefully considered and addressed to ensure successful production. The organic fillers should be dried properly before mixing with plastics; otherwise, the two will not bond effectively. A uniform filler dispersion promotes better performance of the eco-composites through minimization of voids and maximization of bonding with the hosting matrix.¹¹³ Proper heating reduces the moisture content, which negatively affects the internal structure of the eco-composite. Furthermore, recycled plastics should be melted and processed below the degradation temperature of the added cellulosic biomass during the manufacturing of the eco-composites.¹² Moreover, advanced recycling technologies must be carefully selected to produce materials that are cost-competitive over virgin polymers. For mechanical recycling, new compatibilizers or compatibilization strategies should be explored that can encompass melt processing of blended polymers to deliver high-performance products.¹⁷⁶

Polymer functionalization is a crucial step toward creating better polymer/fiber bonding and producing high-performance eco-composites. Polymer functionalization is usually performed in the extruder during the melting phase, combined with the application of chemical functional groups for a specified time duration. Key parameters affecting functionalization include initiators such as peroxide, coupling agent contents such as MAH, polymer geometry such as size,¹⁷⁷ state of functionalization (melt state or solid state), temperature, and time. Efficiency indicators for optimal functionalization are yield, MFI, carboxylic index, and stability indicator. An appropriate manufacturing process considering the ingredients and target product should be carefully selected. The final performance of an eco-composite depends not only on the choice of the matrix, fiber, and their treatment, but also on the manufacturing process. The presence of imperfections during

manufacturing must be carefully addressed. Such imperfections can degrade the manufactured material, which can eventually result in accelerated damage due to:

- Micro-crakes in the matrix.
- Filler discontinuity, compaction, delamination, and de-bonding.
- Filler agglomeration.
- Shear stress concentration in matrix at fiber ends.
- Micro-voids in the fibers.
- Poor fiber/polymer interaction due to high moisture content before mixing.
- Sub-surface voids and porosities.

Based on a thorough review of related research works listed in Table 1, the manufacturing processes that are best fits for different types of recycled polymers and produce high-performance eco-composites are ranked as shown in Figure 27. Typical processes such as extrusion, compression molding, injection molding, or a combination of any two of these can be used to fabricate the eco-composites. For example, rLDPE- and rPET-based eco-composites can be manufactured by the hand lay-up method. Moreover, extrusion followed by compression molding appears to be the best technique for eco-composite fabrication, according to the results of the relevant published studies. This is due to the ability to obtain an optimal homogenization by melt mixing using extrusion, enhancing the mechanical interlocking, and reducing the void content through compression molding. Several studies reported that rHDPE and rPP are the most preferred plastics to develop matrices in eco-composites, as reflected by their reasonable strength and desirable performance. This is supported with the fact that HDPE and PP are the most used thermoplastics to manufacture bio-composites.

Polymer nanocomposites have gained a commercial footing, due in large part to their characteristics and convenient methods of production. The preparation procedures include sol gel process, in-situ polymerization,

solution casting, melt mixing and in-situ intercalative polymerization.¹⁷⁸ Solution casting is one of the oldest and most convenient methods to fabricate nanocomposite in thin films and small-sized sheets. In the solution casting of polymer nanocomposites, the polymer phase is dissolved in a non-aqueous solvent and mixed with nanosized particles in the same solution prior to casting on substrate. The solvent phase is taken out by evaporation and thereafter the dried film is separated from the substrate.¹⁷⁹ In this process, polymer and prepolymer are equally merged and made soluble in the mixture.¹⁸⁰ Combining additives into the casting solution is considered an effective way to adjust the phase separation process and as a result modifying the nanocomposite characteristics.¹⁸¹

4 | CHALLENGES AND FUTURE PERSPECTIVES

For a successful application of eco-composites, further development and research activities should address critical issues such as generation of new modified polymers, blending, reinforcement, additives, crosslinking, among others. Eco-composite production is still in its infancy phase. Several difficulties hinder its full commercialization, including material processing, product manufacturing, markets, and regulations. The following subsections highlight few challenging matters that require further research and better understanding for eco-composites to become competitive and find more applications in global markets.

4.1 | Recyclability

Thus far, a small percentage of plastic wastes have been collected and mechanically recycled. This is mainly due to the high prices of collection, cleaning, sorting, and treatments. Therefore, introducing efficient processes to

enhance the characteristics of recycled plastics is crucial to upsurge the willingness for recycling plastic wastes, phase-out their landfilling, and thus alleviate their harmful effect on the environment. Through joint efforts from design, manufacturing, and end-users, new processing techniques for recycling should be further developed in the future. Furthermore, the natural fiber composite market is mainly driven by the growing demand for lightweight and green products in several industries. Similarly, increasing the demand for recyclable products has promoted the bio-composite market worldwide. Hence, supply chain management of eco-composites should be thoroughly evaluated to obtain sufficient resources that sustainably meet the market demand.

4.2 | Filler modification

Although several modification processes for natural fillers exist, these methods still need improvements in terms of maximum utilization of agro-residue, filler hydrophobicity and compatibility with polymers, mechanical properties, as well as treatment time and cost. This can be achieved by employing new advanced technologies for natural filler treatments, including plasma, ultrasonic-, and microwave-assisted chemical and biological treatments. Careful adjustment of the process parameters and optimization methods will help in achieving better filler properties compared to the traditional treatment methods. In the future, new methods, including biological and nanoparticle, treatment are needed to enhance the sustainability of the adopted manufacturing processes.

4.3 | Polymer functionalization

Surface functionalization of polymers and natural fillers is expected to attract the attention of researchers over the

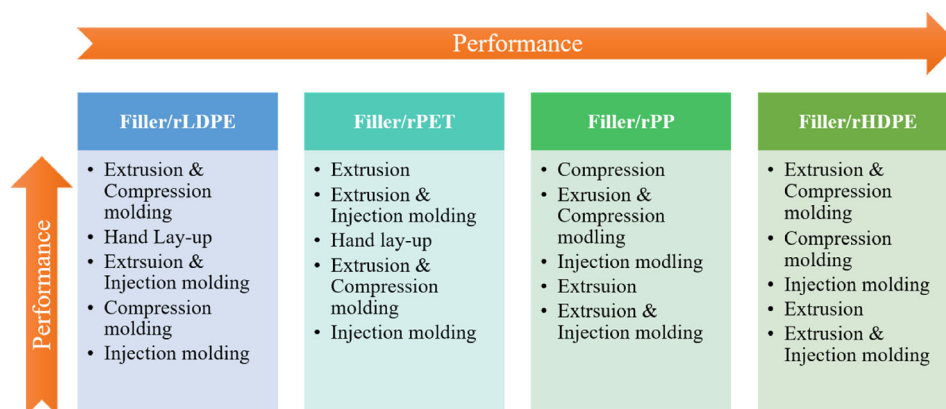


FIGURE 27 Graphical presentation for ranking the most preferred manufacturing processes along with the most preferred types of eco-composites [Color figure can be viewed at wileyonlinelibrary.com]

coming years. Further studies are required to explore new modification methods to achieve the desired interfacial bonding between the matrices and fillers. In this regard, functionalization of the polymer surface by plasma, microwave, and ultrasonic has proved to be effective and eco-friendly in obtaining hydrophilic surfaces of polymers. For the production, the eco-composites can be improved by adding proper stabilizers before processing, such as anti-ultraviolet and anti-oxidation additives. Similarly, enchaining the polymer flow and processing conditions can reduce the shape memory effect during the fabrication.

4.4 | Additive manufacturing and applications

Adaptive manufacturing, including 3D printing of eco-composites for emerging applications considering geometric flexibility is of significant interest. Recent advances in manufacturing complex structures can provide new horizons for eco-composites to overcome major limitations in load-bearing applications. The 3D printing technology extends enormous potential in the biomedical products, such as anatomical prototypes for surgery training/planning, rehabilitation, dentistry, tailored implants, drug delivery equipment, and organ printing.¹⁸² However, the low strength of 3D printed bio-composite materials is still a challenge, which requires further research to overcome the experienced limitations of the technology.¹⁸³ Eco-composites have high potential to fulfill the emerging needs for high strength-to-weight-ratio contemporary materials as demanded by the automotive and aerospace industries. Polypropylene based eco-composites incorporating different types of inorganic and organic low-density fillers for strength reinforcement have been successfully developed. Scientists and researchers worldwide are pursuing persistent efforts to further develop industrially useful eco-composites.

4.5 | Hybrid and nano eco-composites

In recent years, numerous nanoscale additives have been exploited in the reinforcement of virgin polymeric materials. However, a limited number of hybrid eco-composites incorporating nanoparticles and recycled postconsumer plastics have been processed. Mixing nano-clay or nanofillers with a bio-polymer matrix has the potential to improve the overall properties of the produced eco-composites. Nanoparticles such as TiO₂, rGO, SiO₂, ZnO, and CNT could dictate research and application areas in the near future¹⁸⁴ with the potential to be functionalized,

achieving higher level of performance, including waterproofing, fire retardancy, antibacterial resistance, UV-protection capability, insulation, self-cleaning, and others. Also, different forms of carbon-based elements including nanotubes, fibers, expanded graphite, flexible graphite, graphene, and reduced graphene oxide (RGO) have been widely used to develop hybrid nanocomposites due to their cost-effectiveness and easy synthesis and processing.^{185,186} Expansion of the surface areas of such particles helps in improving the bonding with the grains of the hosting matrix. Several studies have concluded that among the most important factors to achieve better homogenous mixture is to use the powder form of the matrix. However, nanocomposites are still limited to nano-cellulose fillers. Therefore, further studies should be conducted to explore the agro-residue nano-powder as it is less costly than cellulose and other derivatives.

4.6 | Eco-composite polymer blends

Significant research efforts are underway to produce polymer-blend composites that provide enhanced alternatives over pure polymer-blend counterparts.¹⁸⁷ Successful composite blends will be used to design a broad range of products with novel applications to enter unexplored markets. However, any newly developed eco-composite needs to exhibit high-quality performance, serviceability, durability, and reliability standards. Future areas of interest should include developing coupling agents and compatibilizers to enhance the polymer/polymer compatibility and the polymer blend/natural filler compatibility. Significant research is required to develop novel methods for chemical modification and investigate the real mechanisms of interactions occurring at the interfaces.

4.7 | Technical plastic-based eco-composites

The most commonly used technical plastics in aerospace and automotive industries are acrylonitrile butadiene styrene, polyamides, polycarbonate, and polybutylene terephthalate. Recent studies have concluded that technical plastics exhibit excellent properties such as high strength, durability, and thermal stability over common plastics after recycling. These attributes can be extended further by reinforcing technical plastics with natural fibers to meet the required industry standards. Accordingly, by adopting recycled technical plastics, manufacturers can effectively contribute toward meeting environmental goals and gain potential financial benefit from reclaiming a high-value waste stream.

4.8 | Bioplastics and green polymers

The expansion of bioplastics to replace conventional petroleum-based plastics has received significant attention from the research community due to the recognized issues of plastic pollution. Polyethylene generated from sugarcane ethanol is known as green polyethylene as it is developed from renewable sources. It is regarded as an innovative polymer that captures carbon dioxide from the atmosphere through its production and does not discharge it back to the environment during its life cycle.¹⁸⁸ The production of bioplastic films has been considered throughout many years with the integration of renewable sources such as starch, cellulose, plasticizer, and several other additives to acquire a high-quality material.¹⁸⁹ The bio-based polymers can be used in large-scale applications because they are chemically similar to their petrochemical counterparts. Meanwhile, a crucial factor for the growth of bioplastics is driven by its manufacturing costs. Bio-polypropylene is still in its pilot stage due to technical hurdles that include conversion and downstream processing technologies. With further research and development activities, it is projected that bioplastics will move into commercialization in the coming years.¹⁹⁰

5 | CONCLUDING REMARKS

Plastic wastes are considered as a real environmental concern that requires collective action to reduce the amount of MSW and diminution of natural resources. Thus, to improve the sustainability of eco-composites, promoting the use of recycled plastic solid waste to develop new classes of eco-composites by the addition of natural fillers is important. These eco-composites will sooner or later be more efficient than neat polymers or reinforced polymers using synthetic fibers. This critical review highlights the recent progression and main findings on eco-composite research, aiming to help researchers to plan, select, and develop various forms of eco-composite materials. The key attribute is to mobilize the use of recycled polymers in developing useful products. Improved manufacturing processes that can develop eco-composite materials with good quality for use in diverse applications are essential. In particular, the current review article highlights the following points.

- Eco-composites have emerged as a major class of fiber-reinforced composite materials, which are characterized by easy processing, good dimensional stability, and reasonable mechanical behavior.
- Performance of eco-composites primarily hinges on the nature, type, and strength of developed bonds among constituents. Hence, natural fibers should be treated prior to use in eco-composites.
- Recycled HDPE is considered the most compatible polymer with natural fillers, followed by recycled PP. In contrast, recycled PET and LDPE do not show much improvement when mixed with natural fillers. More research is required to obtain a better overview of the factors affecting the properties of recycled polymers.
- Productive utilization of cellulosic ingredients extracted from natural agro-residues in recycled polymers as bio-fillers can be realized through various modification methods to produce fillers at nanoscale.
- Mechanical interlocking can be accomplished (chemical or physical methods) via careful filler surface modification. However, because of the growing demand for high-performance composites for structural applications, conducting only filler surface treatment is not enough to produce effective eco-composites. Therefore, efforts are currently focused on strengthening the adhesion of the filler/polymer through chemical or physical bonding and crosslinking.
- Most reported research works focused on modifying the filler surfaces without proper removal of outer layer of impurities. This is insufficient to effectively enhance the filler/polymer compatibility. Hence, coupling agents and compatibilizers will not be highly effective because the functional groups on the filler surface are still covered by wax and impurities.
- Chemical modification of the polymer and/or natural fillers enhances the compatibility among the eco-composite ingredients. However, the chemical reaction by-products should be carefully separated while manufacturing. These products or gases might affect the worker's health and the eco-composite quality. Furthermore, the formation of internal voids degrades the mechanical properties of produced eco-composites.
- The functionalization of polymers is a critical step in developing improved polymer/fiber bonding and developing eco-composites of high efficiency. During the melting step, polymer functionalization is generally performed in the extruder in conjunction with the application of chemical functional groups for a specified period. Initiators, coupling agent, polymer geometry such as size, state of functionalization (melt state or solid state), temperature, and time are the main parameters influencing functionalization.
- The existing methods for natural fiber treatment and modifications can be thoroughly enhanced by using accelerated methods such as plasma and microwave-assisted processes.
- To successfully extract non-cellulosic biomass, a newly invented accelerated microwave-assisted process has

exhibited boosted surface dewaxing and degumming, increased crystallinity and decreased moisture sensitivity, and fiber deterioration resistance with relatively high tenacity and elongation at break.

- The highest tensile properties, enhanced impact energy, and better toughness were achieved at 10 wt% filler content. However, as the fiber content increased, several microstructure-related problems were encountered that caused property degradation.
- As the fiber content increased beyond 20% in the eco-composites, low homogeneity and compatibility between the natural fillers and recycled polymers were observed. In some cases, natural particle agglomeration occurred when the concentration exceeded 40 wt%.
- Compared to virgin PP-based bio-composites, the rPP-based eco-composites showed similar mechanical properties. As manifested by good dispersion and fiber-matrix bonding, natural fillers exhibited remarkable compatibility with the polypropylene matrix.
- It can be inferred that rPP is a valuable material with decent mechanical properties for manufacturing natural fiber polymer composites.
- Compared to neat polymers, the flexural strength of most recycled polymers has improved, particularly at fiber loads in the range of 5–20 wt%; degradation is noticed beyond this limit. In general, as compared to the tensile strength, the enhancement in the flexural strength was more pronounced. Flexural modulus demonstrated a similar trend for several types of eco-composites.
- Several attributes have resulted in the enhancement in the mechanical properties of the rHDPE-based eco-composites. First, the rHDPE density is marginally higher than the HDPE density, which demonstrates that rHDPE has low branching and therefore provides strong intermolecular forces and tensile strength. Second, the presence of additives in the recycling process could make the natural filler more compatible with the rHDPE matrix, which in turn will enhance the mechanical properties of the eco-composites.
- Viscosity at the melt state impacts the processability of the eco-composites that should be optimized and maintained. Recycling has been reported to increase the complex viscosity of the LDPE and HDPE blend that exhibits miscible activity in the melt state. However, the use of compatibilizers increases the viscosity of the eco-composites.
- Water uptake (moisture absorption) adversely affects the performance of eco-composites. It limits the applicability of the eco-composites under various operational conditions.
- Water absorption of the eco-composites occurs through fine voids, gaps between the polymer and the filler,

and polymeric matrix microcracks. The interfacial bonding of the filler/polymer thus strongly affects the water absorption of the eco-composites.

- The density of eco-composites is lower than that of other types of composites. The density is substantially dependent on the strength of the internal bonds among the ingredients.
- With the addition of a natural filler to the recycled matrix, the thermal stability of the developed eco-composites has been found to be significantly improved.
- High thermal stability, low thermal conductivity, and comparable strength were exhibited by blending recycled polyolefin and date palm leaves. They are therefore considered an attractive material for use in thermal insulation.
- The natural filler acts as a nucleation agent for recycled polymers. Therefore, it improves their crystallization on outer surface, and increase the crystallization temperature.
- It is worthwhile to develop a framework to optimize the thermal properties of target eco-composites. The thermal characteristics include thermal stability, coefficient of thermal expansion, thermal degradation, decomposition temperature, transition temperature, enthalpy, and crystallinity degree. Generally, both the thermal stability of cellulose and conservation of the internal structure of the microfibrils contribute to the enhancement of the mechanical properties.
- Understanding the microstructural features and biochemical characteristics of natural fibers is crucial to select a proper manufacturing process for the eco-composites.
- In general, extrusion followed by compression molding seems to be the best choice for eco-composite fabrication. This is because optimal homogenization can be obtained by melt mixing using extrusion and enhancing the mechanical interlocking while reducing the void content through compression molding.

In the future, further research and development activities are required to successfully develop eco-composites that functionally and economically fit the design of commercial products. In this regard, challenges include achieving better mechanical properties by producing more controlled microstructures with homogeneous filler dispersion and stronger bonding with the surrounding polymer. Effective and efficient methods of functionalization and modification of ingredients will contribute to enhanced properties of the newly developed eco-composites. Further research on developing new and different types of manufacturing methods and processes for the eco-composites will aid in boosting the

economic value of industrial products. Additionally, the development and assessment of nano-eco-composites is still an open field for research and exploration. The goal is to produce fully biodegradable, high-performance bioplastics at an affordable price.

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CONFLICT OF INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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