

## Review Article

# Rice Husk Filled Polymer Composites

Reza Arjmandi,<sup>1</sup> Azman Hassan,<sup>1</sup> Khaliq Majeed,<sup>2</sup> and Zainoha Zakaria<sup>3</sup>

<sup>1</sup>Department of Polymer Engineering, Faculty of Chemical Engineering, Universiti Teknologi Malaysia (UTM), 81310 Skudai, Johor, Malaysia

<sup>2</sup>Department of Chemical Engineering, COMSATS Institute of Information Technology, 54000 Lahore, Pakistan

<sup>3</sup>Faculty of Science, Universiti Teknologi Malaysia (UTM), 81310 Skudai, Johor, Malaysia

Correspondence should be addressed to Azman Hassan; [azmanh@cheme.utm.my](mailto:azmanh@cheme.utm.my) and Zainoha Zakaria; [zainoha@kimia.fs.utm.my](mailto:zainoha@kimia.fs.utm.my)

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Natural fibers from agricultural wastes are finding their importance in the polymer industry due to the many advantages such as their light weight, low cost and being environmentally friendly. Rice husk (RH) is a natural sheath that forms around rice grains during their growth. As a type of natural fiber obtained from agroindustrial waste, RH can be used as filler in composites materials in various polymer matrices. This review paper is aimed at highlighting previous works of RH filled polymer composites to provide information for applications and further research in this area. Based on the information gathered, application of RH filled composites as alternative materials in building and construction is highly plausible with both light weight and low cost being their main driving forces. However, further investigations on physical and chemical treatment to further improve the interfacial adhesion with polymeric matrix are needed as fiber-polymer interaction is crucial in determining the final composite properties. Better understanding on how the used polymer blends as the matrix and secondary fillers may affect the properties would provide interesting areas to be explored.

## 1. Introduction

The increasing demand for eco-friendly materials, increasing depletion rate, and soaring prices of petroleum based plastics and pressing environmental regulations have all triggered a growing interest towards the field of composites [1, 2]. Composites materials are formed by combining two or more materials to improve properties of their original components. When one or more of the materials used are derived from biological origins, they are then defined as biocomposites (ASTM D7075-04) [3].

In general, polymer composites consist of a polymer resin as the matrix and one or more fillers are added to serve specific objectives or requirements. For example, composites for aerospace and sports applications require high mechanical and thermal properties. Traditionally synthetic fibers such as carbon or glass fibers have been used to reinforce composites and are able to produce such properties. However, with the growing global environmental concerns, their slow biodegradability is a disadvantage. Therefore researchers are

finding other viable approaches to enhance or accelerate the biodegradability of polymeric composites. For this reason natural fibers provide good prospective as reinforcements fillers in thermosets, thermoplastics, and elastomers. Some main advantages of using natural fibers in composites are low cost, sustainability, light weight, and being nonabrasive and nonhazardous and more importantly they can accelerate biodegradability of the polymeric composites [4, 5].

Rice (*Oryza sativa* L.) is a primary source of food for billions of people and one of the major crops in the world. It covers around 1% of the earth's surface [6]. Statistics show that, during the period of 2010–2013, the average annual global production of rice was 725 million metric tons with Asia region alone producing over 90% of the total global rice production [7]. Rice husk (RH) is an inexpensive byproduct of rice processing and is separated from rice grain during the rice milling process. It is reported that, for every ton of rice produced, about 0.23 tons of RH is formed [8]. Rice milling is one of the most important industries in countries such as China, India, Indonesia, Malaysia, and Bangladesh [9].

TABLE 1: (a) Components and (b) physical properties of RH [21].

Property	RH
(a) Components (%)	
Cellulose	25–35
Hemicellulose	18–21
Lignin	26–31
SiO <sub>2</sub> (silica)	15–17
Solubles	2–5
Moisture content	5–10
(b) Physical properties	
Particle size ( $\mu\text{m}$ )	26.64
Surface area ( $\text{m}^2/\text{g}$ )	0.92
Density ( $\text{g}/\text{cm}^3$ )	1.00

The incineration of RH is discouraged, which in turn produces ash, fumes, and toxic gases, leading to serious air pollution. Moreover, silica is intrinsic to RH and has been successfully used to enhance the mechanical properties of composites [10]. In the paddy plants of Malaysia, with a land area of approximately 680,000 hectares, a total of 840,000 tons of RH is produced every year [11, 12].

RH is a cellulose-based fibrous material with a wide range of aspect ratios [13]. The major components and physical properties of RH are tabulated in Table 1 [14–21]. Due to its high availability, low bulk density ( $90\text{--}150\text{ kg m}^{-3}$ ), toughness, abrasiveness in nature, resistance to weathering, and unique composition, a variety of applications have been proposed in the literature. RH has the potential to be utilized as an insulating material, in the production of organic chemicals [22], panel boards and activated carbon [23], and supplementary cementing material [9]. Potential of RH as source of power generation and its financial viabilities have also been studied and have produced encouraging results [24, 25]. Even though some of this husk is converted into the above-mentioned end products, however, like many other agricultural byproducts, the industrial applications of this biomass are still limited with little economic value. Therefore, it is very important to find pathways to fully utilize the RH and an intense research scrutiny is currently undertaken worldwide to identify potential applications and to develop economically feasible processes for these applications on a commercial scale.

Various researchers have reported on a variety of applications that involve RH. However, it has also been reported that, in the manufacture of composites panels with the use of RH, there is a poor interaction between the RH and matrix materials, which results in weak particle-matrix adhesion [26]. Although previous studies reported that RH particleboard could be used in the manufacture of furniture and interior fitments, the physical and mechanical properties of the particleboard were lower than those of the particleboards made from wood particles [27–29]. The main reasons for lower physical and mechanical properties of the RH particleboards are low aspect ratio and waxy/silica layer of the RH particles [18]. The incorporation of RH into polymer matrices provides advantageous characteristics, such as biodegradability, light

TABLE 2: (a) Chemical and (b) physical properties of typical RHA fillers [34].

Property	WRHA	BRHA
(a) Chemical composition (%)		
CaO	0.360	0.120
MgO	0.160	0.078
Fe <sub>2</sub> O <sub>3</sub>	0.041	0.022
K <sub>2</sub> O	0.690	0.950
Na <sub>2</sub> O	0.034	0.018
Al <sub>2</sub> O <sub>3</sub>	0.025	0.023
P <sub>2</sub> O <sub>5</sub>	0.570	0.270
SiO <sub>2</sub> (silica)	92.200	53.880
Loss of ignition (LOI)	1.620	44.480
(b) Physical properties		
Particle size ( $\mu\text{m}$ )	6.60	19.50
Surface area ( $\text{m}^2/\text{g}$ )	1.40	26.80
Density ( $\text{g}/\text{cm}^3$ )	2.20	1.80

weight, toughness, and resistance to weathering, and also makes the final products more economically competitive [30, 31]. Compared with wood-based composites, the RH filled polymer composites have higher resistance to termite and biological attack and also better dimensional stability upon exposure to moisture. Thus, these composites are increasingly being used in building construction, such as frames for windows and doors, slidings, decks, and interior panels, and in the automotive industry for interior parts like door panels and trims [32].

RH, when burnt in open air outside the rice mill, yields two types of ash that can serve as fillers in plastics materials, namely, white rice husk ash (WRHA) and black rice husk ash (BRHA). The upper layer of the RHA mound is subjected to open burning in air and yields BRHA in the form of a carbonized layer. On the other hand, the inner layer of the mound being subjected to a higher temperature profile results in the oxidation of the carbonized ash to yield WRHA that consists predominantly of silica [33]. Table 2 shows the chemical and physical properties of typical RHA fillers [34]. As can be seen in Table 2, WRHA has been analyzed and found to have approximately 95% silica content, while BRHA has typically only approximately 54% silica content and a substantial carbon content that is about 44%.

Compression molding, injection molding, and extrusion are the most commonly used processing techniques for natural fiber reinforced thermoplastic composites. Enhancement of processability of natural fiber reinforced thermoplastics is an important issue, as the addition of interphase modifiers reduces the flow properties of the composites considerably by increasing the interaction between the fiber and the matrix [19, 20]. On the other hand, incorporation of processing aid improves processability by reducing the viscosity of the molten composites. Processability enhancement, while retaining and/or improving the performance properties, is more economical and can be an added advantage for the natural fiber composites industry [20]. RH has been

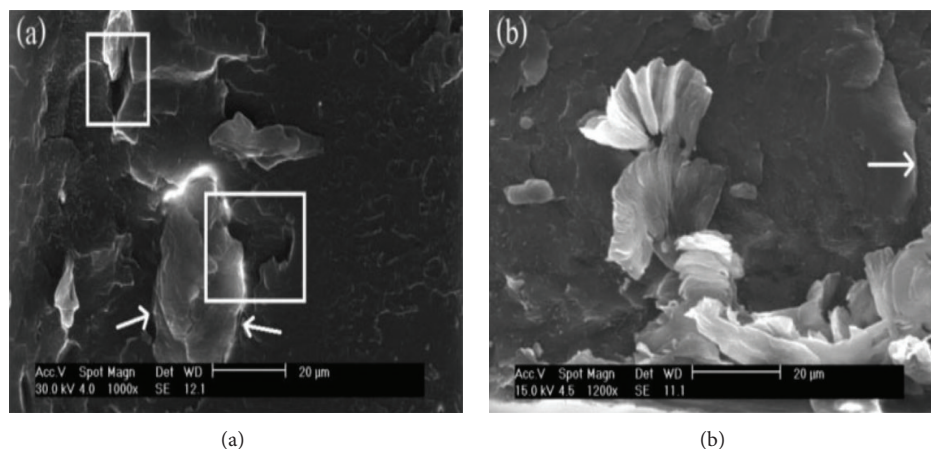


FIGURE 1: SEM micrographs of the fractured surface of composite films with varying MMT content: (a) 0 and (b) 6 wt% of MMT content [42].

incorporated into a wide variety of materials such as low-density polyethylene (LDPE) [18], high-density polyethylene (HDPE) [35], polylactic acid (PLA) [36], polypropylene (PP) [37], and polyvinyl chloride (PVC) [38]. In general, the resulting composites exhibit improved mechanical, thermal, chemical, physical, and other properties with the addition of RH. However, RH was shown to have varying degree of reinforcing effect on the different polymer matrices. In this respect, the aim of the present study is to describe the possibility for utilization of RH as fillers in the different polymer composites.

## 2. Rice Husk Composites

Many studies on the physical, mechanical, and thermal properties of RH filled polymeric composites and how these incorporation affects various thermosets, thermoplastics, and elastomers have been attempted [18, 35–38]. The composite properties are largely dependent on matrix properties and hence selection of the matrix is based on desired end properties of the composites. The literature review reveals that polyethylene (PE), PP, PVC, and PLA are among popular choices for RH filled polymer composites. The subsequent sections begin with some general characteristics of the polymers along with their particular applications and then report the influence of incorporating RH on the properties. In addition, properties of RH filled polymer composites are also summarized to gain a deep intuitive understanding on the subject of developing multifunctional material.

**2.1. Rice Husk/Polyethylene Composites.** PE is a synthetic thermoplastic polymer which is made from the monomer ethylene. It is an important family member of the polyolefin resins. It is the most widely used polymer in the world and predominates in plastics owing to a number of reasons such as high elongation at break, good chemical resistance, excellent water barrier and good oxygen barrier properties, light weight, and lower cost. PE has contributed greatly to the development of high performance materials and their

production and usage have been enormously increased [39]. PE is being made into products ranging from packaging films to bottles and automobile fuel tanks. PE is classified into different categories based mostly on its density and branching. It is worth mentioning that its mechanical and barrier properties are greatly influenced by the extent and type of branching. The long, chainlike molecules, in which hydrogen atoms are connected to a carbon backbone, can be produced in linear or branched forms. Branched versions are known as low-density polyethylene or linear low-density polyethylene; linear versions are known as high-density polyethylene and ultrahigh-molecular-weight polyethylene. Researchers have extensively studied the mechanical properties of RH filled PE and properties of some RH based composites are given in Table 3.

In our recent study [42], rice husk/maleic anhydride-modified polyethylene/montmorillonite (RH/MAPE/MMT) filled LDPE nanocomposites films (containing 0 to 6 wt% of MMT content) were prepared by extrusion blown film. The films were characterized by morphological, mechanical, oxygen ( $O_2$ ) barrier, and thermal properties. The delamination of MMT layers evidenced from X-ray diffraction (XRD) results revealed the formation of intercalated nanocomposites, which was confirmed by the increase in the interlayer spacing and substantial reduction in their peak intensity of MMT. Scanning electron microscopy (SEM) confirmed that adding MMT did not alter the adhesion features between RH and LDPE (Figure 1). Furthermore, addition of RH/MAPE/MMT to the LDPE has improved the mechanical and  $O_2$  barrier properties. Tensile and tear properties were improved with the addition of MMT and the maximum improvement of 8% in tensile strength and 5% in tear strength was observed at 3 wt% of MMT content. Concurrently, the modulus continues to increase while elongation at break decreases with increasing the MMT content. However, the most significant finding of this research was the large increase in  $O_2$  barrier, in which the maximum barrier was observed at 4 wt% of MMT content (more than twofold). Additionally, thermogravimetric analysis (TGA) and differential scanning

TABLE 3: Mechanical properties of RH filled PE composites.

Matrix	RH (wt%)	Secondary filler (wt%)	Coupling agent or compatibilizer (wt%)	Tensile strength (MPa) (% incr.)	Tensile modulus (GPa) (% incr.)	Flexural strength (MPa) (% incr.)	Flexural modulus (GPa) (% incr.)	Impact strength (% incr.)	Reference
HDPE	65	—	—	13.5	2.39	33.5	2.90	26	[19]
HDPE	65	—	E-GMA (2.5)	22.5	2.37	49.06	3.32	28	[19]
HDPE	65	—	ethylene-acrylic ester-maleic anhydride (2.5)	20.9 (55)	2.42 (1.30)	40.2 (13.5)	2.81 (-3.1)	80 (III)	[20]
HDPE	30	—	Ultra-Plast TP10 (1)	—	—	—	—	75 (-32)	[40]
HDPE	50	—	MAPE (2)	25.64	2.5	—	—	18.39	[41]
HDPE	50	MMT* (2)	MAPE (2)	29.83	3.2	—	—	17.82	[41]
rHDPE	45	Sawdust (17.1)	MAPP (7.9)	7.5	0.6	—	—	15.20	[35]
rHDPE	44.5	Sawdust (17.1)	MAPP (7.9)	11.6	1.02	—	—	33.20	[35]
rHDPE/rPET (75/25)	60	—	MAPE* (3)/E-GMA (5)	23 (15)	1.55 (89)	35 (17)	3.5 (192)	25 (-69)	[12]

% incr. = percentage increase; not. = notched; unnot. = unnotched.

\* phr.

TABLE 4: DSC results of the nanocomposites film [42].

Samples	$T_{\text{onset}}$	$T_{\text{pm}}$	$T_m$	$T_c$	$X_c$
LDC-0	511	551	107	95	19.1
LDC-2	519	553	107	96	19.5
LDC-3	524	559	108	95	19.8
LDC-4	525	559	108	95	20.7
LDC-5	520	557	107	96	20.3
LDC-6	514	552	108	95	19.6

LDC: LDPE/RH/MAPE (90:4:6 wt%) composites film reinforced with different MMT content (0–6 wt%);  $T_{\text{onset}}$ : onset temperature of degradation;  $T_{\text{pm}}$ : degradation temperature at peak maximum;  $T_m$ : melting temperature;  $T_c$ : crystalline temperature; and  $X_c$ : degree of crystallinity.

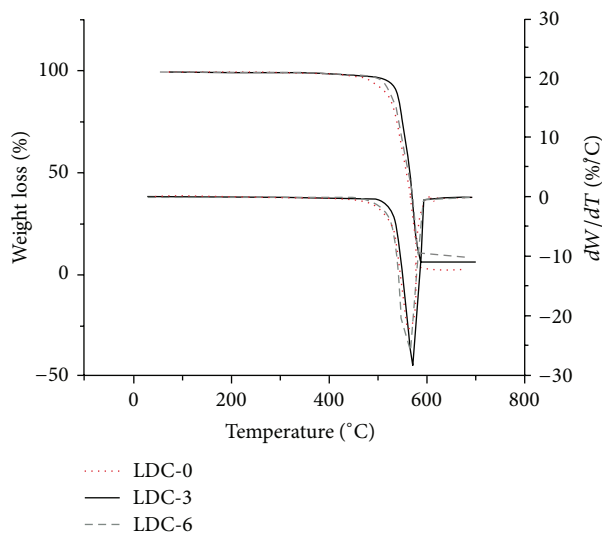


FIGURE 2: TGA and DTG thermograms of the nanocomposites film. LDC: LDPE/RH/MAPE (90:4:6 wt%) composites film reinforced with different MMT content (0, 3, and 6 wt%) [42].

calorimetry (DSC) results are shown in Figure 2 and Table 4, respectively. As can be seen, the degree of crystallinity ( $X_c$ ) and thermal stability of LDPE/RH/MAPE/MMT nanocomposites are better than those of LDPE/RH/MAPE composites and the maximum improvement was at 4 wt% of MMT content. The unique combination of  $O_2$  barrier, biodegradation, and mechanical properties for the LDPE/RH/MAPE/MMT nanocomposites films showed that these nanocomposites films provide potential candidates for a variety of food packaging.

The adhesion between the reinforcing fiber and the matrix plays an important role in the final mechanical properties of the materials. In another study by Majeed et al. [43], RH flour (having particle size of less than 75  $\mu\text{m}$ ) and nanoclay filled LDPE composites were prepared by extrusion blown film. In this study, MAPE was used as compatibilizer in various concentrations ranging from 0 to 8 parts per hundred (PPH), and the effect of MAPE compatibilizer on the morphological, tensile, and oxygen barrier properties was examined. The XRD results showed the addition of 2, 4, 6, and 8 PPH of MAPE increased the interlayer spacing of MMT

by 20, 33, 36, and 38% compared to the uncompatibilized composites, respectively. From morphological observations, it can be concluded that the compatibilizer helps polymer molecules to penetrate and delaminate the clay platelets to realize large filler aspect ratio. In addition, it was found that adding compatibilizer enhances natural fiber dispersion in the continuous LDPE phase, and the RH aggregates started disappearing with increased compatibilizer concentration, which in turn improves homogeneity, as confirmed by SEM (Figure 3). Furthermore, the compatibilizer improved the tensile properties. It was observed that adding only 2 PPH of MAPE to the composites system improved the film tensile strength by approximately 22% compared to uncompatibilized films. The tensile properties of the composites films continued to increase as the MAPE concentration increased. This improvement in tensile properties is due to improved interfacial interactions between filler and matrix, owing to the MAPE. Therefore, composites films with high compatibilizer content showed better tensile strength. Moisture absorption usually deteriorates the mechanical properties of composites due to debonding of reinforcements from the polymer matrices. However, in this particular study, moisture absorption increased the tensile strength of uncompatibilized composites, and the authors attributed this increase in tensile strength to the fiber swelling, which could fill the gaps between fiber and polymer matrix and eventually could lead to an increased strength. It is notable that the increase in tensile strength is not significant for compatibilized composite samples with no significant change observed between the properties of dry and water aged samples. On the other hand, moisture absorption decreased the tensile properties for the compatibilized composites. Indeed, the most significant finding of this research was the large increase in oxygen barrier. Oxygen barrier property of the composite films improved remarkably by the presence of MAPE compatibilizer (more than twofold by 4 PPH MAPE). The decrease in permeability also confirms that the impermeable clay platelets are well dispersed as the extent of dispersion of clay platelets plays a major role in creating a tortuous path for the permeating molecules.

Recently, Kwon et al. [18] reported the effect of adhesive type and content on the dimensional stability and mechanical properties of three-layer particleboards. The composition layers were made by the mixture of wood particles (face layer, 30 wt%) and RH particles (core layer, 70 wt%) under laboratory conditions (Figure 4). Two types of thermosetting adhesives, liquid urea-formaldehyde (UF) and phenol-formaldehyde (PF), and thermoplastic adhesive (LDPE) powder were used as binder in the experiments. They found that the dimensional stability and mechanical properties of particleboards with RH core were significantly improved by the incorporation of LDPE into the core layer. It was studied that at the amounts of UF and PF adhesives decreased from 8 to 4 wt% at the same LDPE content (10 wt%); the thickness swelling (TS) and water absorption (WA) of the particleboards increased but were significantly lower than those of the control specimens. The reason was that a hydrophilic material (RH) was replaced by a hydrophobic material (LDPE). They explained that although the internal bond (IB) strength of particleboards containing 10 wt% LDPE

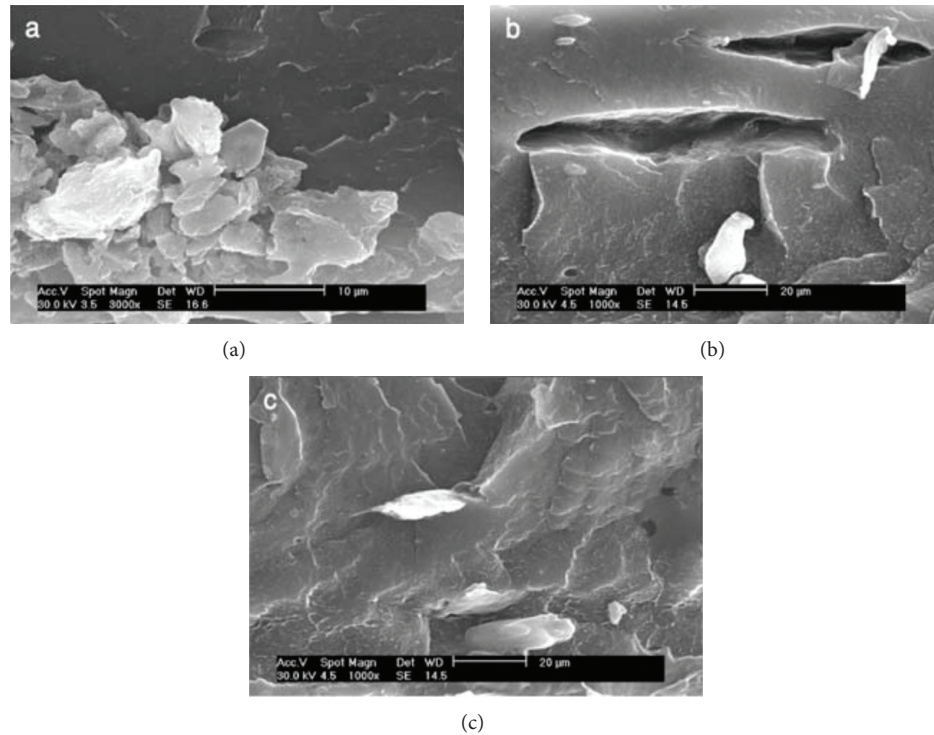


FIGURE 3: SEM micrographs of the fractured surface of composites films with varying compatibilizer concentrations: (a) 0, (b) 2, and (c) 6 PPH [42].

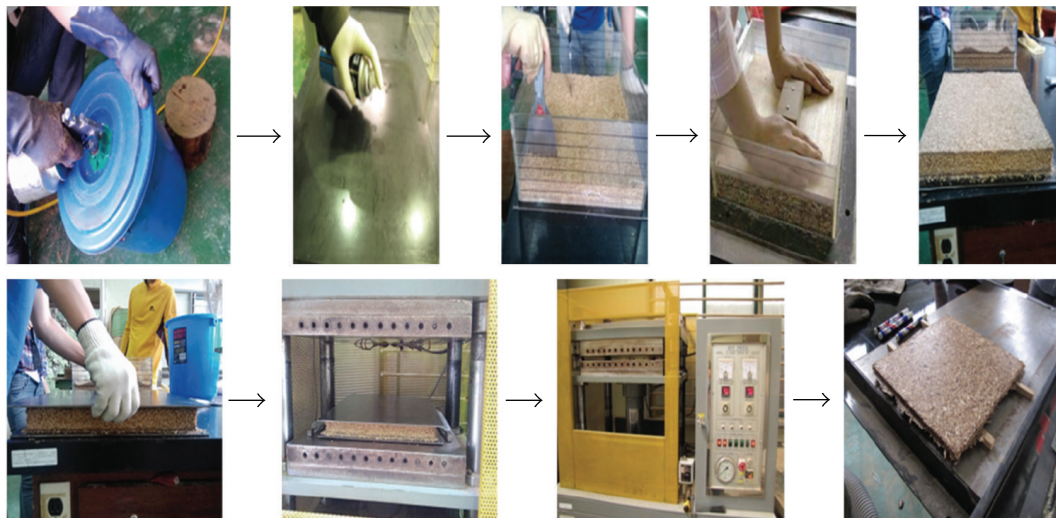


FIGURE 4: Laboratory-scale production of three-layer particleboards [18].

decreased with decreasing UF adhesive content in the core layer, it was higher than that of the control particleboards. Different fracture modes were observed during the IB tests. The fracture modes of particleboard samples with and without LDPE were presented in Figure 5 [18]. This study also demonstrated that the bending strength (MOR) and modulus of elasticity (MOE) of the particleboards containing 10 wt% LDPE decreased with decreasing the amounts of UF and PF adhesives. However, there were no statistically significant

differences in the MOR and MOE values of the particleboard types as the adhesive content decreased from 8 to 4 wt% in the core layer. Based on the findings obtained from Kwon et al.'s [18] study, it can be suggested that the RH particleboards containing LDPE (in particular above 20 wt% of LDPE) are suitable for use in damp places, such as bathrooms, toilets, kitchens, and laundries.

Chuayjuljit et al. [44] successfully obtained silica ( $\text{SiO}_2$ ) from RHA. They explored the possibility of using silica

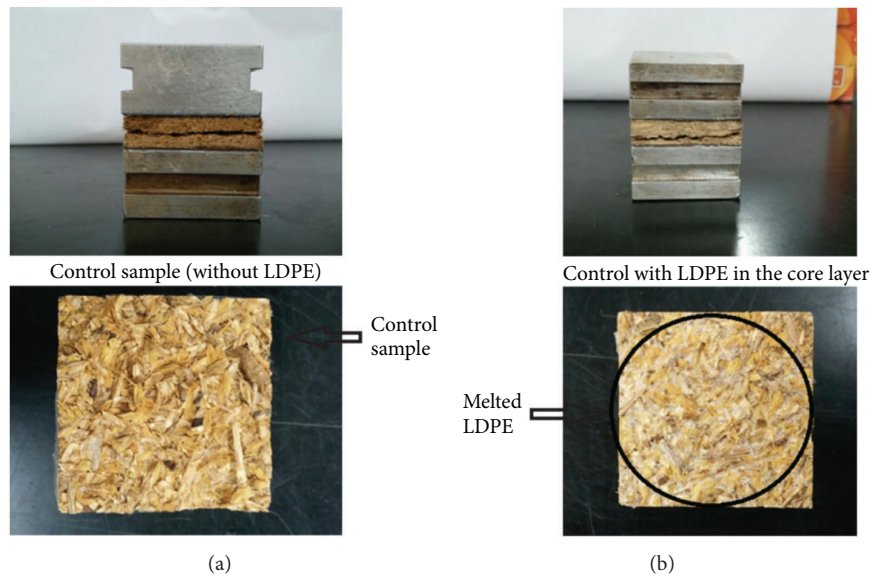


FIGURE 5: Fracture modes and fracture surface of the IB test samples without LDPE (a) and with LDPE (b) [18].

TABLE 5: Properties of RHA silica and commercial silica [44].

Properties	RHA silica	Commercial silica
Average particle size (micron)	4.27	6.61
Specific surface area ( $\text{m}^2 \text{g}^{-1}$ )	$131.35 \pm 2.34$	$511.97 \pm 4.70$
Bulk density ( $\text{g cm}^{-3}$ )	0.677	0.319
pH	5.7	6–8
Oil absorption ( $\text{g g}^{-1}$ )	1.92	2
Silica content (% $\text{SiO}_2$ )	99.6	99.0

from RHA (RHA silica) as antiblocking agent in LDPE film. Properties of RHA silica were compared with commercial silica, as can be seen in Table 5. In addition, the appropriate amount of silica to be used as an antiblocking agent in LDPE film was also investigated. The results indicated that silica prepared from RHA, although composed of finer particles, showed less porosity than that of commercial silica, resulting in a lower specific surface area. Nevertheless, it is possible to use the obtained RHA silica as an antiblocking agent for LDPE film. They found that the addition of RHA silica to LDPE film modifies the film blocking behavior by inducing film roughness. However, the blocking is reduced not as much as in the case of commercial silica. It is investigated that, to the same antiblocking capability, 2000–3000 ppm of RHA silica is needed as opposed to 500–1500 ppm of commercial silica normally used in the plastic film industry. It was found that, as the amount of silica increased, certain mechanical properties (tensile strength, elongation at break, and tear strength) were decreased markedly. This is probably because silica has a lower bulk density and larger average particle size.

Panthapulakkal et al. [19] explored the potential of extruded RH filled HDPE composites profiles for structural applications. In their study, four different coupling agents

based on ethylene-(acrylic ester)-(maleic anhydride) terpolymers and ethylene-(acrylic ester)-(glycidyl methacrylate) terpolymers were used to enhance the performance properties of the composites. The chemical composition and molecular weight of these coupling agents, and also tensile and flexural strength of HDPE/RH (32.5 : 65 wt%) composites are given in Table 6. They studied the effects of various coupling agents (2.5 wt%) on various performance properties of RH filled HDPE composites. The results indicated that these coupling agents significantly enhanced the tensile and flexural strength of the HDPE/RH composites, and the extent of the coupling effect depends on the nature of the interface formed. It is notable that the tensile and flexural strength of HDPE/RH composites without present of any coupling agent are 13.5 and 33.5 MPa, respectively. Their results showed that RH filled HDPE composites gain enough strength and rigidity by incorporating coupling agents and can be used for making structural materials. They found that incorporation of coupling agents enhanced the resistance to thermal deformation and the water absorption properties of the composites, whereas it reduced the extrusion rate significantly. Among the four coupling agents used, the coupling agent with a glycidyl methacrylate functional group and without any methyl acrylate pendant group on the polymer backbone (EGMA1) was found to be the best coupling agent for the RH filled HDPE composites. They concluded that the extrusion rate of these composites is reduced by the incorporation of the coupling agents and has to be enhanced by proper use of process aids.

In another study by Panthapulakkal et al. [20], the effect of coupling agent and processing aid on the performance properties of RH filled HDPE composites was studied. In their study, HDPE composites filled with 65% RH were extruded using a single screw extruder with die dimensions of  $3 \times 8$  mm. They investigated the processability and performance properties of the HDPE composites, which were

TABLE 6: Interrelation between chemical compositions of coupling agents and mechanical strength of HDPE/RH (32.5 : 65 wt%) composites [19].

Coupling agent (2.5 wt%)	Butyl acrylate (wt%)	Maleic anhydride (wt%)	Methyl acrylate (wt%)	Glycidyl methacrylate (wt%)	Molecular weight ( $\text{g mol}^{-1}$ )	Tensile strength MPa	Flexural strength MPa
EBMA1	6	3.1	—	—	120000	20.9	40.2
EBMA2	17.7	3.1	—	—	120000	19.3	42.7
EGMA1	—	—	—	8	100000	22.5	49.6
EGMA2	—	—	15	9	—	15.7	33.8
HDPE/RH*	—	—	—	—	—	13.5	33.5

\*Without coupling agent.

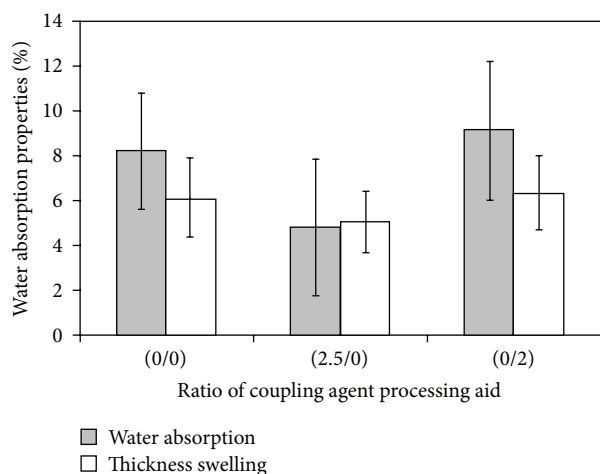


FIGURE 6: Tensile and flexural modulus of the composites with and without process additives [20].

highly dependent on the concentration of the coupling agent and processing aid in the composites formulation. In their study, attempt was made to optimize the composites formulation with respect to both coupling agent and processing aid to achieve optimum mechanical and water absorption properties with an optimum extrusion rate. It was observed that incorporation of a terpolymer (ethylene-acrylic ester-maleic anhydride) based coupling agent enhanced the tensile strength, flexural strength (Figure 6), and resistance to water absorption properties of the composites with a significant reduction in the processability. However, this study revealed that the addition of processing aid reduced the strength and stiffness of the composites and the effect was at a higher content of processing aid in the formulation. They found that the extrusion rate of the composites improved by the incorporation of processing aid and processing aid adversely affected water absorption resistance of the composites. They observed that the composites with a coupling agent to processing aid ratio of 0.73 : 0.59 by weight showed an optimum combination of performance properties and extrusion rate.

In an interesting study, the compounding of RH and HDPE was investigated using corotating twin-screw extruder [40]. Four sizes of RH were studied at various compositions. The size ranged from 500  $\mu\text{m}$  and below, while the content

of RH in the composites varied from 30 to 50 wt%. Also, a fixed amount of Ultra-Plast TP10 as a compatibilizer and Ultra-Plast TP 01 as lubricant was added to the composites compound. It was noticed that among the four RH sizes (A, 250–500; B, 125–250; C, 63–125, and D, <63  $\mu\text{m}$ ), RH with the size of 250–500  $\mu\text{m}$  which has the largest particle was considered to be the most appropriate size as the composites filler based on thermal stability test. The melt flow rate of RH/HDPE decreased with the increasing in RH filler and apparent viscosity also increased with composition for all fillers sizes. They found that, when RH was incorporated into the HDPE matrix, the melt flow rate of RH size A (250–500  $\mu\text{m}$ ) composites performed well above the control limit set. Although composites with RH size C and D had better melt flow at 30 wt%, the fillers tended to degrade at high processing temperature, such as 190°C or when longer residence time was used. Melt flow rate above 4 g 10 min<sup>-1</sup> was found to be the lower limit for injection molding process. It is observed that RH size A has the highest impact resistance among the other sizes. The large particles of RH have low agglomeration and dispersed better in the composites matrix. Filler incorporated at 30% of RH size A was found to be suitable for processing and to have optimum rheological properties with respect to impact strength compared to 40 and 50 wt% of RH.

Kord (2011) fabricated nanocomposites based on HDPE and rice husk flour (RHF) blend (ratio of 50 : 50 wt%) with different content of nanoclay using an internal mixer [41]. In his study, the influence of nanofiller at four levels, 0, 2, 4, and 6 parts per hundred compounds (phc), on the thermal and rheological behavior was studied. The morphology of nanoclay was determined by XRD, and the effect of morphology on the thermal and dynamic mechanical properties was also considered. It was indicated that the crystallization temperature, crystallization enthalpy, and crystallinity level increased with increasing of nanoclay up to 2 phc and then decreased. Additionally, the dynamic mechanical behavior of nanocomposites was improved by the addition of nanofiller and XRD patterns revealed that the nanocomposites formed were intercalated. It was also reported that the sample containing 2 phc of nanoclay had higher order of intercalation and better dispersion, as confirmed by XRD. It was found that with increasing of coupling agent (MAPE) a fully exfoliation morphology could be obtained. Therefore, he concluded



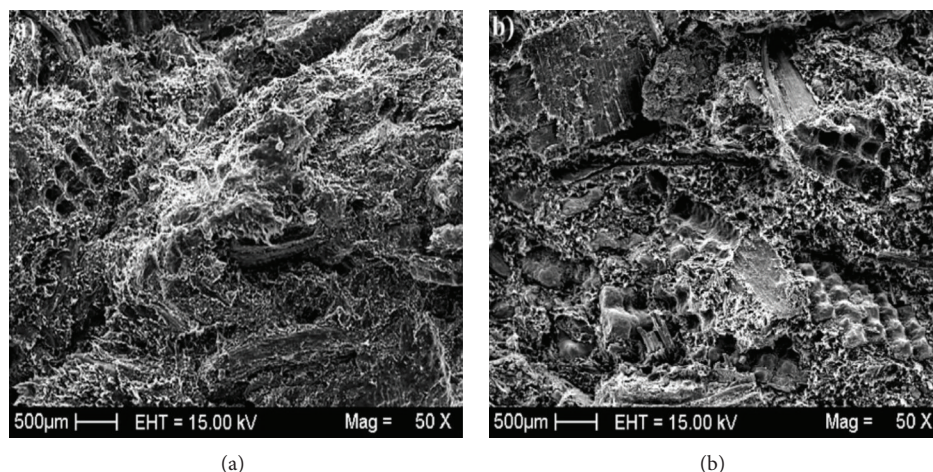


FIGURE 7: SEM micrographs of fracture surfaces of sample from composites at 50x: (a) 0.5% AOs and (b) 20% FRs [35].

that thermal and rheological properties of the HDPE/RHF nanocomposites were improved by increasing of MAPE content.

Hamid and coworkers [35] studied the physical and mechanical and thermal properties of recycled high-density polyethylene (rHDPE) reinforced with a high content of hybrid RH and sawdust. They also studied the effect of antioxidants (AOs) and fire retardants (FRs) as additives. The rHDPE and natural fibers were compounded into pellets by a compounder; then the pellets were extruded using a corotating twin-screw extruder and the test specimens were prepared using hot and cold press processes. Also, maleic anhydride-grafted polypropylene (MAPP) was used as a coupling agent to increase the adhesion of the fiber-matrix interface. Results showed that the samples with 0.5 wt% of AOs and 20 wt% of FRs produce the most reasonable strength and elasticity of composites. Furthermore, the effect of the presence of AOs and FRs on water uptake was minimal and microstructure analysis of the fractured surfaces of composites confirmed improved interfacial bonding. As can be seen in Figure 7, composites with (a) 0.5% AOs and (b) 20% FRs content show the good dispersion of hybrid fibers in the matrix. Therefore, good dispersion of fibers in composites leads to high performance on physical and mechanical properties. There is no clear gap between hybrid fibers and matrix (Figure 7), indicating the good interface bonding. The fibers were perfectly attached to the matrix and it is also evident that the fibers were strongly imbedded in the matrix, indicating the efficiency of the mixing, which was attributed to a good interfacial interaction between hybrid fibers and matrix. It was concluded from the results that the addition of AOs and FRs as mineral fillers improves not only the aging and fire resistance aspects but also the durability performance of rHDPE reinforced with a hybrid of RH and sawdust. As a result, various parameters influence the mechanical properties of fiber-reinforced composites: fiber aspect ratio, fiber-matrix adhesion, stress transfer at the interface, mixing temperatures, and the addition of a coupling agent.

More recently, Chen et al. [12] fabricated RHF composites based on uncompatibilized (UPB) and compatibilized (CPB) recycled high-density polyethylene/recycled polyethylene terephthalate (rHDPE/rPET) with ethyleneglycidyl methacrylate (E-GMA) copolymer by a two-step extrusion and hot pressing with fiber content of 40, 60, and 80 wt%. They noted that the tensile and flexural properties of the composites were significantly increased with increasing RHF content, especially the tensile and flexural modulus. In contrast, as expected, they observed that the addition of RHF to the polymer blends (PB) matrices decreased the elongation to break and impact strength of the composites. However, the results indicated that the compatibilization of PB matrices by E-GMA compatibilizer could further increase the overall mechanical properties of CPB-based composites. In their research, WA test was examined in distilled and seawater. In long-term water immersion test, Chen et al. [12] found that the WA and TS increase with RHF content and immersion time for all types of composites irrespective of the types of polymer matrix (UPB and CPB) and immersion conditions (distilled and seawater). They also observed that the diffusion coefficient, thermodynamic solubility, and permeability obtained from the Fickian model and the swelling rate parameter obtained from swelling model increased with the RHF content. However, all these parameters decreased with the compatibilization of rHDPE/rPET by E-GMA, especially at lower content matrix. SEM analysis of the compatibilized matrix composites confirmed the improved interfacial bonding of matrix-matrix and filler-matrix phases (Figure 8). As can be seen in Figures 8(a) and 8(a'), the UPB displayed an incompatibility morphology of an obvious phase segregation structure between rHDPE and rPET, whereas the CPB exhibited a finer dispersion of rPET component inside the HDPE matrix. Figures 8(b) and 8(c) show a coarse morphology in the polymer matrices. From Figure 8(b), it can be clearly observed that the larger particle size shows no evidence of interfacial interaction and adhesion within the matrix phase, which confirmed the incompatibility of the two matrix individual components. The black arrows in the

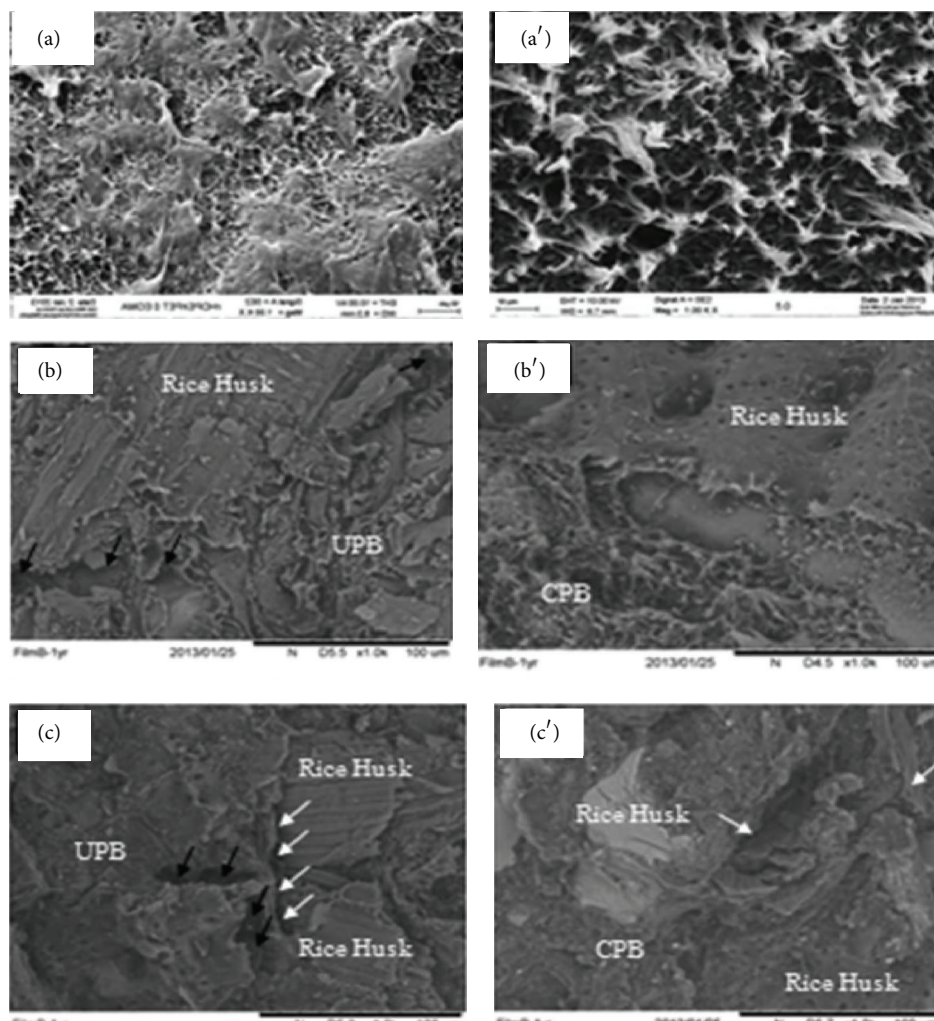


FIGURE 8: SEM micrographs of (a) 0 wt% UPB, (a') 0 wt% CPB, (b) 40 wt% RHF-UPB composite, (b') 40 wt% RHF-CPB composite, (c) 80 wt% RHF-UPB composite, and (c') 80 wt% RHF-CPB composite (magnification, 1000x) [12].

Figures 8(b) and 8(c) show that the available spaces or sites for the water molecules diffuse and form hydrogen bonds with the polar rPET by breaking the existing bonds between the hydroxyl groups of the rPET chain. However, RHF-reinforced composites with CPB matrices exhibited a finer surface morphology (Figures 8(b') and 8(c')) with smaller size matrix domains, which indicates the good dispersion or rPET particles inside the rHDPE matrix and the better interfacial adhesion within the matrix. By comparing Figures 8(b/b') and (c/c'), it can be observed that the RHF fillers at the concentration of 40 wt% were well embedded in the PB matrix, which indicates the relatively good interface adhesion between the fibers and matrix. Meanwhile, the white arrows in the latter figure show a clear gap between the polymer matrix and the RHF fillers, which enhances the water diffusion. This indicated the poor filler-matrix interfacial bonding, as a result of insufficient amount of coupling agent at high RHF content (80 wt%). Based on the results and findings in this work [12], they proposed that

the agrofiller/recycled plastics composites are suitable for outdoor applications in terms of low WA and TS.

**2.2. Rice Husk/Poly(lactic Acid) Composites.** PLA is a synthetic thermoplastic polymer and one of the most promising candidates for biodegradable polymers with excellent mechanical properties [45]. PLA is produced on a large scale from fermentation of cornstarch to lactic acid and subsequent chemical polymerization. As PLA is obtained from renewable resources, therefore, it represents a good candidate to produce disposable packaging in addition to its application in the textile and automotive industries and clinical uses [46]. Pure PLA can degrade to carbon dioxide, water, and methane in the environment over a period of several months to two years, compared to other petroleum plastics needing a very long period. Although PLA is an eco-friendly plastic with good biocompatibility, poor hardness, slow degradation, hydrophobicity, and lack of reactive side-chain groups limit

TABLE 7: Mechanical properties of RH filled PLA composites.

Matrix	RH (wt%)	Coupling agent or compatibilizer (wt%)	Tensile strength (MPa) (% incr.)	Tensile modulus (GPa) (% incr.)	Flexural strength (MPa) (% incr.)	Impact strength ( $Jm^{-1}$ ) (% incr.) Unnot.	Reference
PLA	AT RHF (25)	—	7.35 (-82)	—	13.9 (-84)	—	[48]
PLA	WT RHF (25)	—	39.47 (-21)	—	52.4 (-39)	—	[48]
PLLA/starch (42/42)	5	TC/MDI (10/1)	8.58	0.062	—	35.23	[49]
PLLA/starch (37/37)	15	TC/MDI (10/1)	14.58	0.225	—	22.31	[49]

AT RHF = alkaline-treated rice husk fiber; WT RHF = water-treated rice husk fiber; TC = triethyl citrate; MDI = diphenyl-methane-4,4'-diisocyanate.

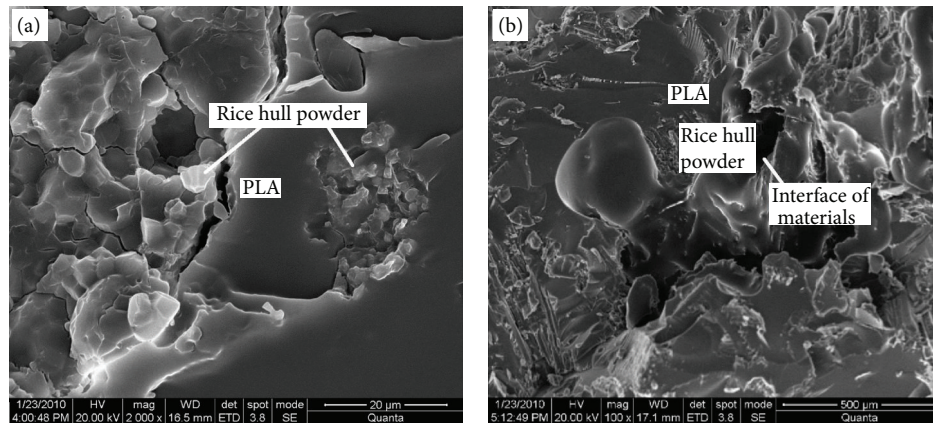


FIGURE 9: SEM micrographs of composites: (a) 20 g of RHP and (b) 20 g of RHP + 2 g of MAPP [50].

its application [47]. The mechanical properties of RH filled PLA composites are illustrated in Table 7.

In research by Hua et al. [50], the rice husk powder (RHP) was modified by PLA using melt-blending technique, and MAPP was used as coupling agent. The mechanical properties, water absorbability of RHP/PLA composites, and the relations between these properties and microscopic characteristics were investigated. Their result showed that, with the increasing content of RHP, the maximum tolerable tension and tensile strength increased. The water absorbability of materials increased with the increasing of RHP content (after 24 hours). They also found that coupling agent could significantly reduce the water absorbability of composite materials. They observed that the hygroscopicity was slow in the beginning and it became strong after a certain period of time because the characteristics will change remarkably after the RHP soaking in the water for a long time. It is also reported that, with the increasing of RHP, the hardness of composite materials increased the tensile strength enhancement, and the maximum bearable tension increased too. As shown in Figure 9(b), there is no obvious change in the content density and the porosity of the RHP in the composites with MAPP compared to composites without MAPP (Figure 9(a)). But it shows that the pore becomes obviously small and smoother. In addition, when the adhesion between PLA and RHP became closer, the agglutination performance of materials enhanced, the composite materials had lower

water absorptivity, and the tensile strength and the most high tension all obtained enhancement obviously when the MAPP was added to RH composites. Therefore, the usage of coupling agent is one of the effective ways to strengthen the performance of composite materials.

Zawawi et al. [51] did a comparison study on microwave properties of natural fiber reinforced PLA composites in order to recognize their potential as alternatives to common printed circuit board (PCB) for electronic communication industries. Thus, they reported on measured dielectric properties for two composites (kenaf/PLA and RH/PLA) and their results were compared. The samples were prepared in equal wt% (50:50 wt%), and the complex dielectric permittivity ( $\epsilon = \epsilon' - j\epsilon''$ ) and loss tangent ( $\tan\delta$ ) of both composites have been studied in the frequency range of 500 MHz to 10 GHz. It was observed that, from 500 MHz to 3.32 GHz, real permittivity ( $\epsilon'$ ) values are consistent throughout the wide frequency range, at approximately 3.3. However, the permittivity seems to decrease at higher frequencies starting from 3.35 GHz for both composites, down to 2.5 at 10 GHz. From their observation, real permittivity ( $\epsilon'$ ) parameter has shown significant variation with frequency range. It is also observed that kenaf/PLA composites with the same weight content have higher permittivity ( $\epsilon'$ ) value compared to RH/PLA composites. Meanwhile, they noticed that loss tangent ( $\tan\delta$ ) is low and remains similar for both composites. The results showed that both kenaf/PLA and RH/PLA composites can

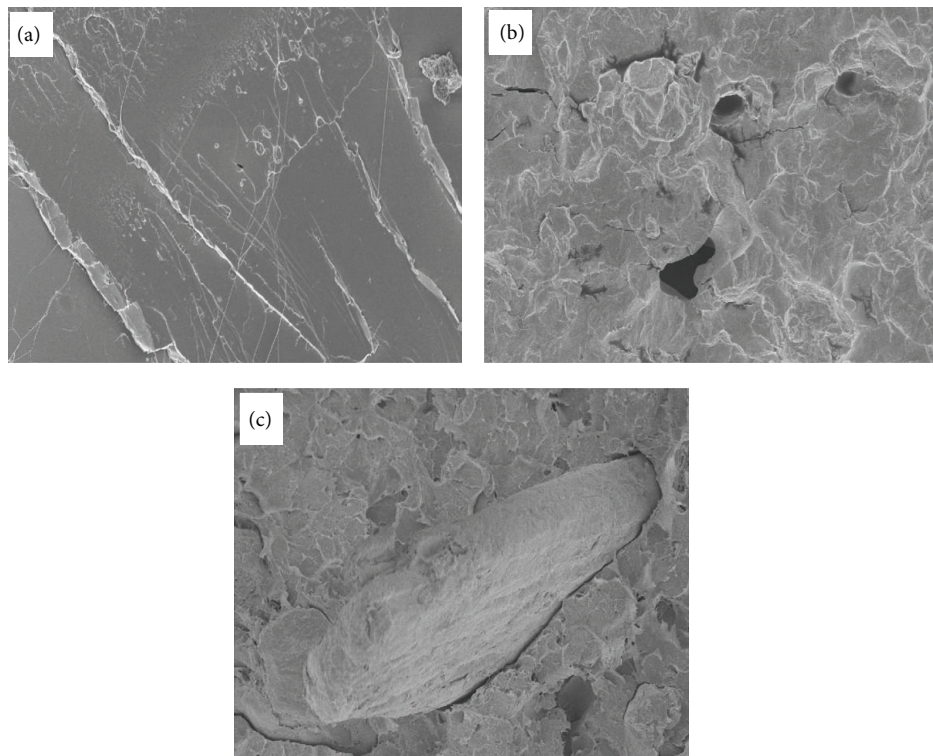


FIGURE 10: FESEM micrographs of flexural fractured surfaces of (a) pure PLA, (b) PLA/AT RHF, and (c) PLA/WT RHF composites [48].

be used for microwave applications due to their permittivity range and low loss tangent comparable to existing commercial substrates, such as epoxy-based dielectrics.

Most recently, a research group in (2014) focuses on the influence of surface treatment and fiber sizes on mechanical behavior, physical properties, and morphology of RH fiber reinforced PLA [48]. Modified RH fiber was prepared by using 6 wt% sodium hydroxide (NaOH) and distilled water. PLA composites reinforced with modified RH fiber (25 wt%) was mixed using the internal mixer and fabricated by the mini injection molding. Farah Dina et al. (2014) found that the tensile and flexural strength of PLA composites with 100, 200, and 500  $\mu\text{m}$  particles sizes of water treated fiber are much higher than those of alkaline treated ones. DSC and TGA analyses were performed and indicated that the water and alkaline treatments reduced the thermal stability of the PLA composites (Table 8). The authors claimed that surface treatment by NaOH on RH fiber could not successfully improve the interfacial adhesion between fibers and PLA matrix. Field emission scanning electron microscopy (FESEM) micrographs of flexural fractured surfaces of composites are shown in Figure 10. Microcrack and pores are indicated by circle and arrow, respectively (Figure 10(b)). This behavior is due to the brittle fracture of PLA matrix adjacent to the alkaline treated RH fiber (AT RHF). The interface between the alkaline treated RH fiber and PLA is loose. It may be due to the loss of fiber-matrix interfacial adhesion and the failure of the fibers in the load distribution which are the evidence of poor fiber-matrix adhesion. From Figure 10(c), it is clear that the PLA/water treated RH fiber (WT RHF)

TABLE 8: DSC and TGA results of WT RHF and AT RHF filled PLA composites.

Samples	$T_g$ ( $^{\circ}\text{C}$ )	$T_m$ ( $^{\circ}\text{C}$ )	$\Delta H_m$ (J/g)	Decomposition temp. of the different weight loss (wt%)		
				5	50	75
PLA	64	171	99	364	395	407
PLA/WT RHF	66	169	77	347	382	400
PLA/AT RHF	57	160	52	296	339	—

fracture surface is rougher than the PLA/alkaline treated RH fiber (Figure 10(b)), which facilitates the mechanical interlocking. Additionally, two phases can be clearly seen and RH fibers were pulled out from the matrix in the fracture process, with large voids thereby being created, as indicated by arrow (Figure 8(c)).

Working in another direction, Battagazzore et al. [52] extracted silica powder from RH by a simple extraction procedure (Figure 11) for preparing PLA composites with differing filler contents (5, 10, 20, and 30 wt%) using melt compounding. The thermal, mechanical, and morphological properties have been investigated. Indeed, the presence of extracted silica has induced significant improvements of Young's modulus as well as a slight reduction of the oxygen permeability of PLA. The collected values at 10 and 30 wt% have been compared with prepared samples using a commercial silica and they found that the achieved mechanical properties turned out to be higher than those exhibited by the homologous samples prepared by using a commercial

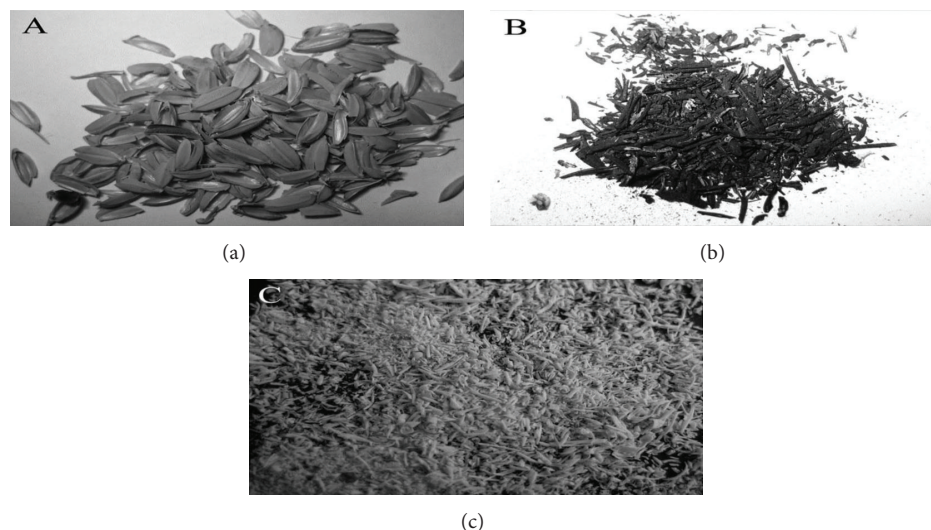


FIGURE 11: Typical images of (a) RH, (b) RHA, and (c) extracted silica [52].

silica, as can be seen in Figure 12. The economic analysis of the materials and whole process (silica extraction and composites preparation) highlighted that the production of a formulation containing 20 wt% silica can be considered economically sustainable, when the energy recovered from the RH burning is reused.

A group in France [53] investigated specific surface treatments, which aimed to improve the adhesion between PLA, RH, and Einkorn wheat husk. In the first part of their study, husks (RH and Einkorn wheat husk) were treated by alkaline solutions at three concentrations (2, 5, and 10%) at room temperature for different times between 6 and 48 hours. Results revealed that the alkaline treatment dissolved a fraction of waxes, lignin, and hemicellulose, so that the polarity of their surface was increased and therefore alkaline treated husks were more sensitive to moisture and had lower surface energies. Moreover, the alkaline treatments had more influence on wheat than on RH, as confirmed by chemical mechanism determination (Figure 13), FTIR (Figure 14), and SEM observations (Figure 15). In the second part of the Tran et al. (2014) study, different surface treatments including alkaline treatments with NaOH solutions at different concentrations and silane agents ( $\gamma$ -aminopropyltriethoxysilane (APS) and  $\gamma$ -glycidoxypropyltrimethoxysilane (GPS)) on the characteristics of RH and wheat husk were investigated. These silane treatments were applied either alone or on previously alkaline treated husks (NaOH 5% for 24 h). The results showed that silane treatments reduced the moisture sensitivity and raise the energy surface of the husks. In the last part of their study, treated husks were incorporated into PLA to produce composites. Nevertheless, alkaline treatments did not improve the mechanical properties of composites. However, an increase in the mechanical properties and the thermal stability of composites was obtained. Better results were obtained with APS agent compared to GPS agent because of a higher reactivity; the group amine of APS showed more effect on PLA than group epoxy of GPS. When

silane treatments were combined with an alkaline pretreatment, composites showed higher mechanical properties and thermal stability than composites with only silane treatment. Alkali treatment cleaned the surface of husk and activated the hydroxyl groups. They found that these active hydroxyl groups reacted with the silanes more easily than the hydroxyl group on the surface of untreated husks. It was also given in evidence that wheat husks (with higher cellulose content) lead to better performances when they are incorporated within PLA compared to RH. They proposed future works on the influence of the treatment surface on the aged resistance of composites.

In another work, Sareeladdanon and Potiyaraj [54] prepared polymer composites of PLA and linear low-density polyethylene (LLDPE) with the addition of different amount of rice husk silica (RHS), 0.5 and 1 wt%, using melt-mixing in a twin-screw extruder and processed into film by a chill roll cast extruder. The mechanical properties of PLA/RHS and LLDPE/RHS composites were investigated through tensile testing and tear testing. The results showed that the tensile modulus and tensile strength of PLA increased with the addition of 0.5 wt% RHS and dropped with further addition of RHS (1 wt%); this may arise from the agglomerates of silica particles in the PLA matrix. However, the elongation at break of PLA increased with the addition of silica at the higher content. On the other hand, the tensile modulus of LLDPE decreased with the addition of silica because of nonuniform dispersion of silica particles.

In a recent publication, Mohammadi-Rovshandeh et al. [49] investigated the use of RH and bleached rice husk (BRH) in the form of powder and the effect of lignin removal on mechanical and thermal properties of prepared composites with PLA and starch. They confirmed that the lignin has been removed from RH using FTIR technique, as indicated by disappearing of lignin aromatic C=O peak ( $1648\text{ cm}^{-1}$ ) in the FTIR spectra of BRH (Figure 16). The tensile, Charpy impact, and hardness tests were implemented to study the

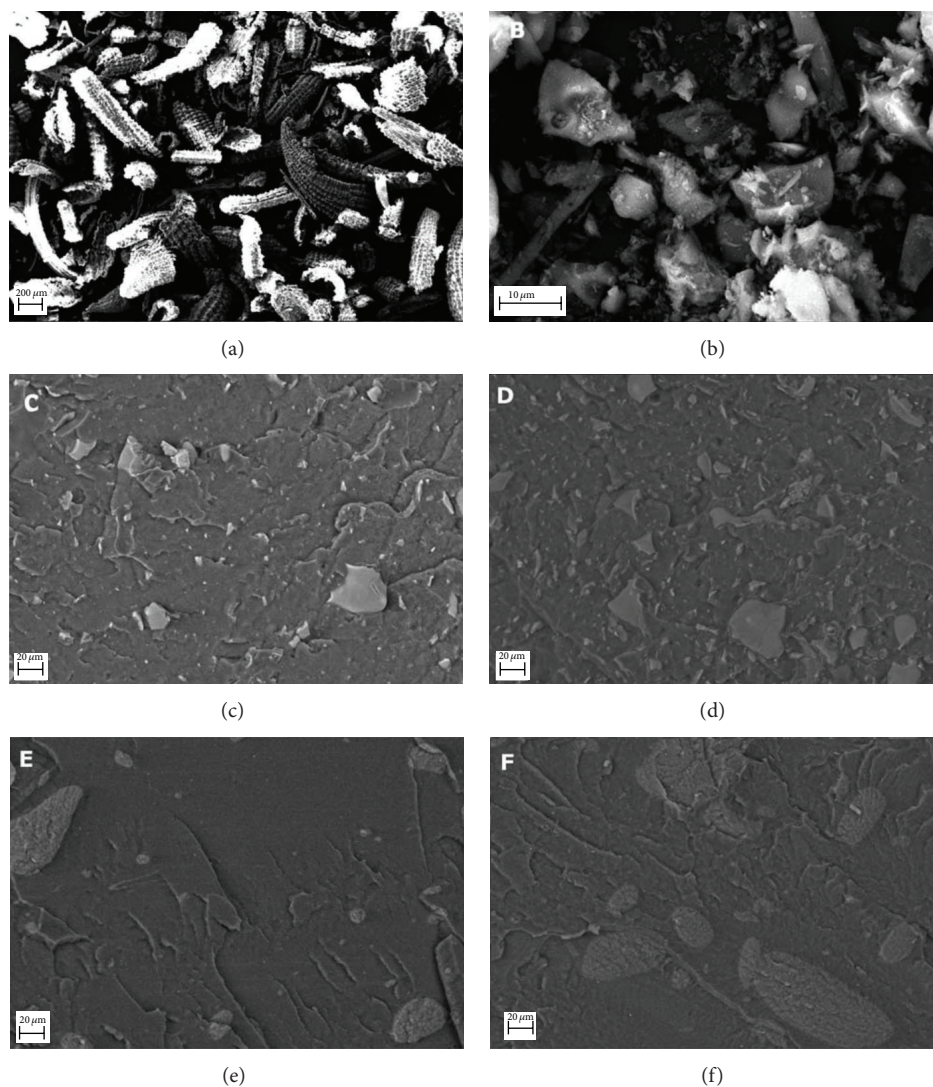


FIGURE 12: SEM micrographs of silica powder (a and b) and composites containing 10 and 30 wt% of extracted (c and d) and of commercial silica (e and f) [52].

mechanical properties. It was shown that the filler content in PLA/starch blends could improve the mechanical properties of composites. It was concluded that filler caused an increase in the toughness and Young's modulus of composites. It was also found that filler improved the tensile strength and hardness of PLA/starch blend. The mechanical properties revealed that 5–10% of BRH or RH may be used in composites which result in improvement of the tensile properties as well as hardness. It was clearly observed that the BRH samples show larger modulus, Charpy impact strength, and hardness compared to those composites filled with RH. Despite larger values for the elongations at break of RH composites, the mechanical properties showed a significant improvement (25–50%) on lignin removal. In addition, the differential scanning calorimetry thermograms revealed an improvement in crystallization behavior and reduction of  $T_g$  from 47 to 31°C in the case of BRH filled polymer composites. Finally, they reported that, due to the removal of lignin in the

samples and modification of mechanical properties, the BRH could have the ability of wide use in the bioplastics and food packaging industries.

In an interesting research in Italy, Battagazzore et al. [36] focused on the extraction of cellulose from RH through a multistep process (Figure 17) and used different extracted cellulose contents (5, 10, 20, and 30 wt%) to fill PLA composites by melt-blending. The crystalline structure and morphology of the extracted cellulose have been investigated by using XRD and SEM, as can be seen in Figure 18. Four main peaks have been found at 16°, 17°, 22°, and 34° (Figure 18(a)). Indeed, the extracted cellulose has a crystalline structure with respect to hemicellulose and lignin, which are amorphous in nature. More specifically, cellulose has a crystalline structure due to the hydrogen bond interactions and Van der Waals forces between adjacent macromolecules. The thermal, mechanical, and oxygen barrier properties of the resulted PLA composites have been investigated and correlated with the observed

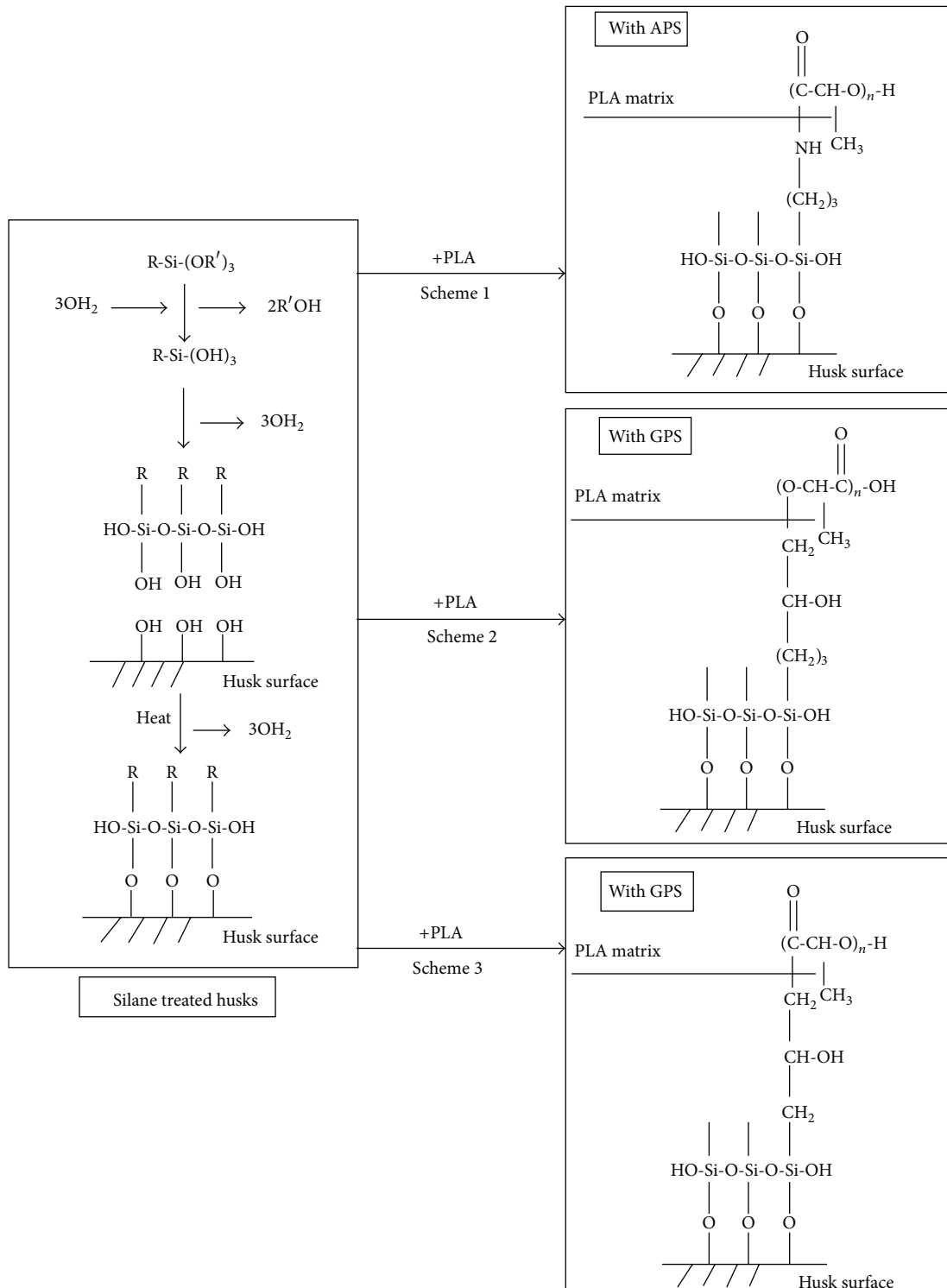


FIGURE 13: Chemical mechanisms at the interfacial area between silane (Scheme 1: APS and Schemes 2 and 3: GPS) treated natural fibers and PLA matrix [53].

morphologies (Figure 19). They reported that, in spite of a broad micrometric distribution of the cellulose particle size (as indicated by arrow in Figures 19(b) and 19(d)), the mechanical properties turned out to be strongly improved

and the oxygen permeability properties have proven to be slightly reduced as well. The most interesting aspect of this work was that similar mechanical and oxygen permeability properties have been achieved for both commercial cellulose

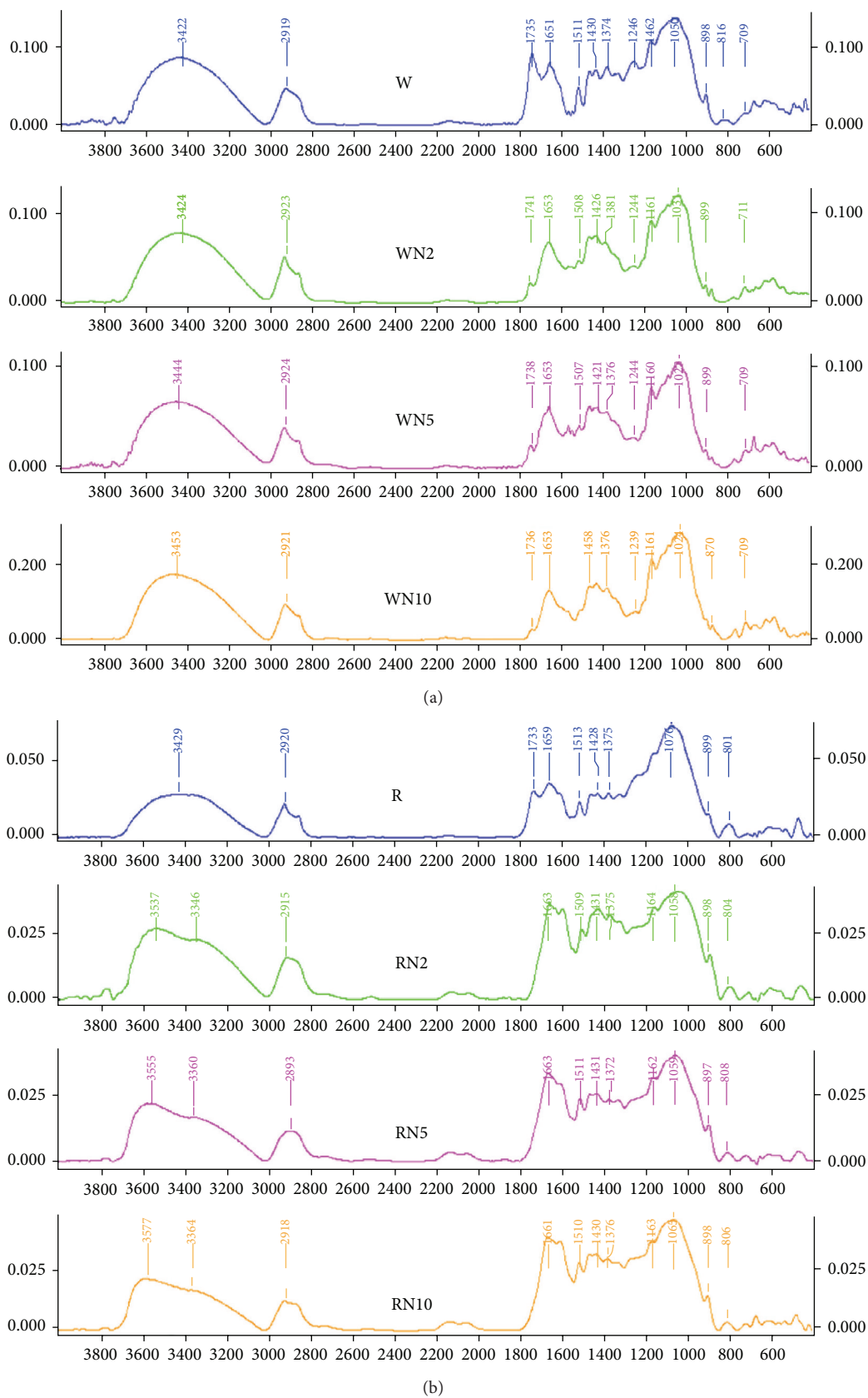


FIGURE 14: FTIR spectra of untreated and alkaline treated (a) wheat husk and (b) RH at different NaOH concentrations (2, 5, and 10%). Einkorn wheat husk (W), einkorn wheat husk + NaOH (WN), long-grain RH (R), and long-grain RH + NaOH (RN) [53].



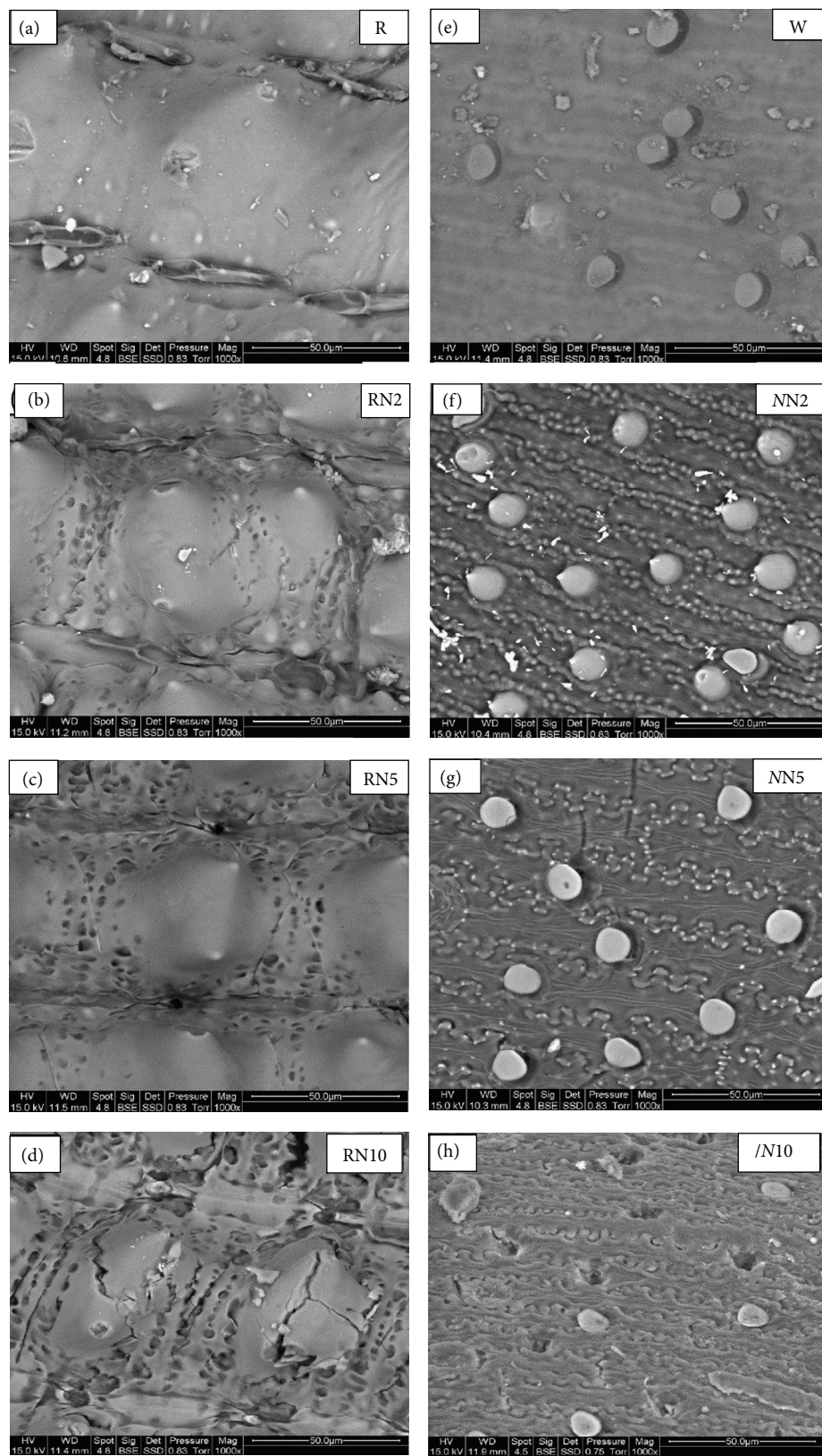


FIGURE 15: SEM micrographs of husks before and after treatment by NaOH: (a) untreated RH; (b-d) treated RH with NaOH concentration of 2%, 5%, and 10%, respectively; (e) untreated wheat husk; and (f-h) treated wheat husk with NaOH concentration of 2%, 5%, and 10%, respectively [53].

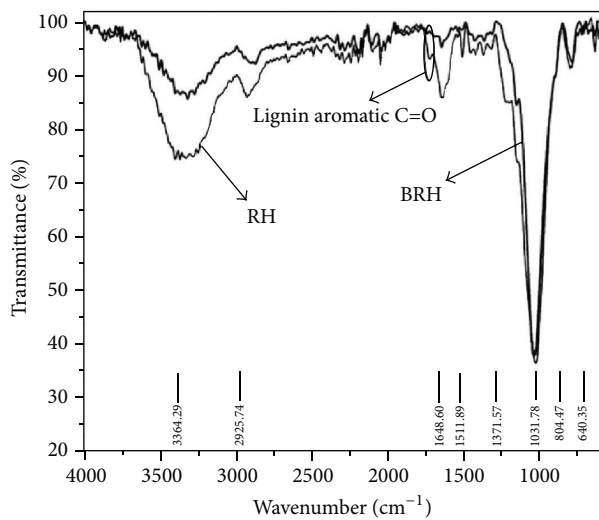


FIGURE 16: FTIR spectra of RH and BRH [49].

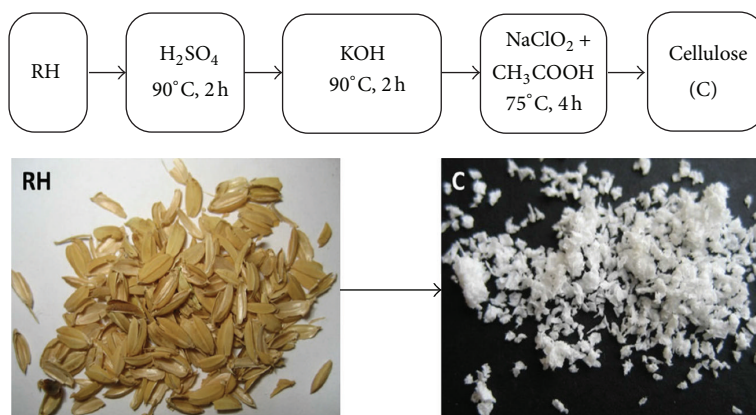


FIGURE 17: Schematic representation of the cellulose extraction from RH [36].

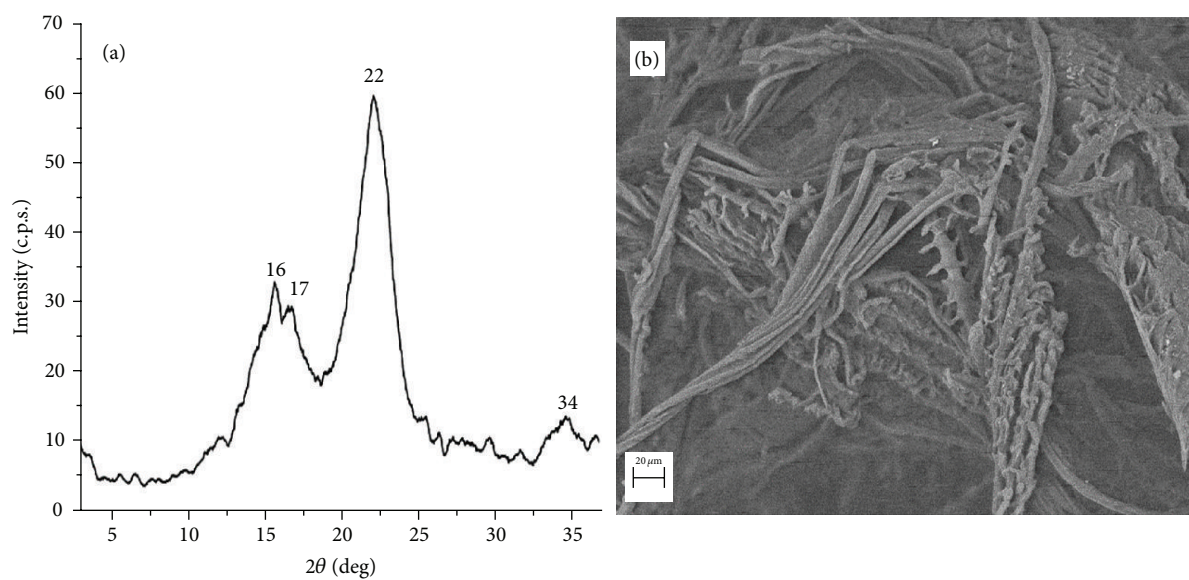


FIGURE 18: (a) XRD patterns and (b) SEM micrograph of the extracted cellulose [36].

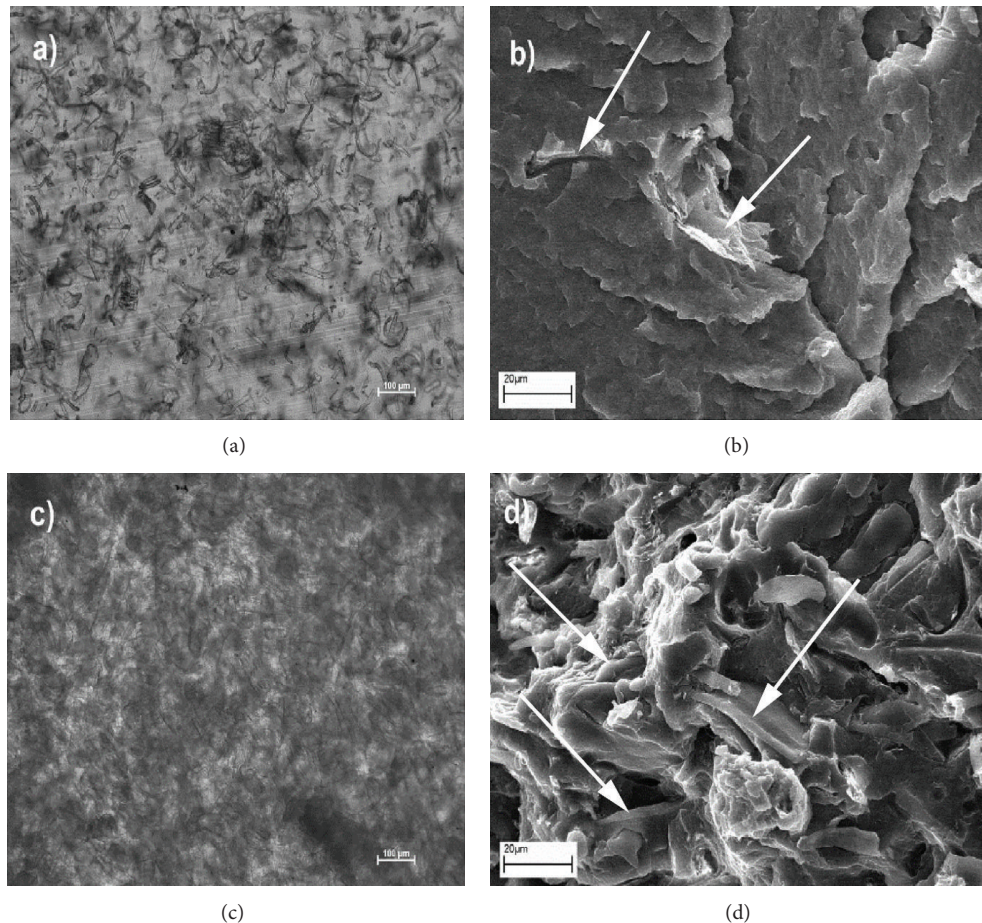


FIGURE 19: Optical and SEM macrographs of PLA filled with 5 wt% cellulose (a and b) and PLA filled with 30 wt% cellulose (c and d), respectively [36].

and cellulose derived from RH. Above all, Battagazzore et al. [36] demonstrated that an industrial waste as RH can be recycled for conferring enhanced final properties to PLA.

**2.3. Rice Husk/Polypropylene Composites.** PP is an addition polymer chemically synthesized from the monomer propylene. It is a thermoplastic polymer mainly used in packaging, laboratory equipment, automotive components, yogurt containers, and margarine tubs. Its transparency and resistance to heat and chemicals nature make it suitable for a variety of applications. Melt processing of PP can be achieved through extrusion and molding. However, the most common shaping technique used is injection molding. Table 9 shows experimental results from different researches in terms of mechanical properties of RH composites by employing PP as matrix material.

Many researchers have reported comparative studies on different fibers based composites. Premalal et al. [21] compared two types of fillers (RHP and talc) to prepare PP composites using a Brabender Plasti-Corder internal mixer at 180°C, which contained 0, 15, 30, 45, and 60 parts per hundred parts of polymer (pph) of fillers. The processability and mechanical properties of the composites with respect

to the filler type and filler content were investigated. It was found that talc composites are easier to process than RHP filled polymer composites. They reported that, in terms of mechanical properties, Young's modulus and flexural modulus increased, whereas yield strength and elongation at break decreased with the increasing in filler contents for both types of composites. They observed that the PP/RHP composites have lower yield strength, Young's modulus, flexural modulus, and higher elongation at break than PP/talc composites. SEM studies of tensile fracture surfaces of the composites are shown in Figure 20. The voids which can be seen in Figures 20(a) and 20(b) clearly indicated the poor interaction between RHP and matrix. As reported by Premalal et al. (2002), the polar nature of RHP and nonpolar nature of the matrix can be cited as one of the possible causes for this observation. By increasing the RHP content (Figure 20(b)), significant filler aggregation has also appeared, which contributed to the poor stress transfer from matrix to filler resulting in poor properties. On the other hand, the talc filled polymer composites (Figures 20(c) and 20(d)) show brittle-type failure. They do not exhibit a tendency towards formation of large agglomerates. Compared to the RHP composites, a stronger talc-matrix interface is

TABLE 9: Mechanical properties of RH filled PP composites.

Matrix	RH (wt%)	Coupling agent or compatibilizer (wt%)	Tensile strength (MPa) (% incr.)	Tensile modulus (GPa) (% incr.)	Flexural strength (MPa) (% incr.)	Flexural modulus (GPa) (% incr.)	Impact strength ( $\text{Jm}^{-1}$ )		Reference
							Not.	(% incr.)	
PP	30*	—	24 (-21)	2.1 (80)	—	1.9 (27)	—	13 (-64)	[21]
PP	WRHA* 40	—	23	1.18	—	2.48	—	—	[33]
PP	10	—	29 (-15)	—	—	—	15 (-21)	23 (-45)	[55]
PP	30	—	—	—	26	1.6	40	—	[56]
PP/NBRr (70/30)	10*	—	13 (-33)	1.1 (2)	—	—	—	—	[57]
PP	50	MAPP (4)	37.5 (5)	0.8 (78)	34 (143)	2.28 (61)	—	—	[37]

\* phr.

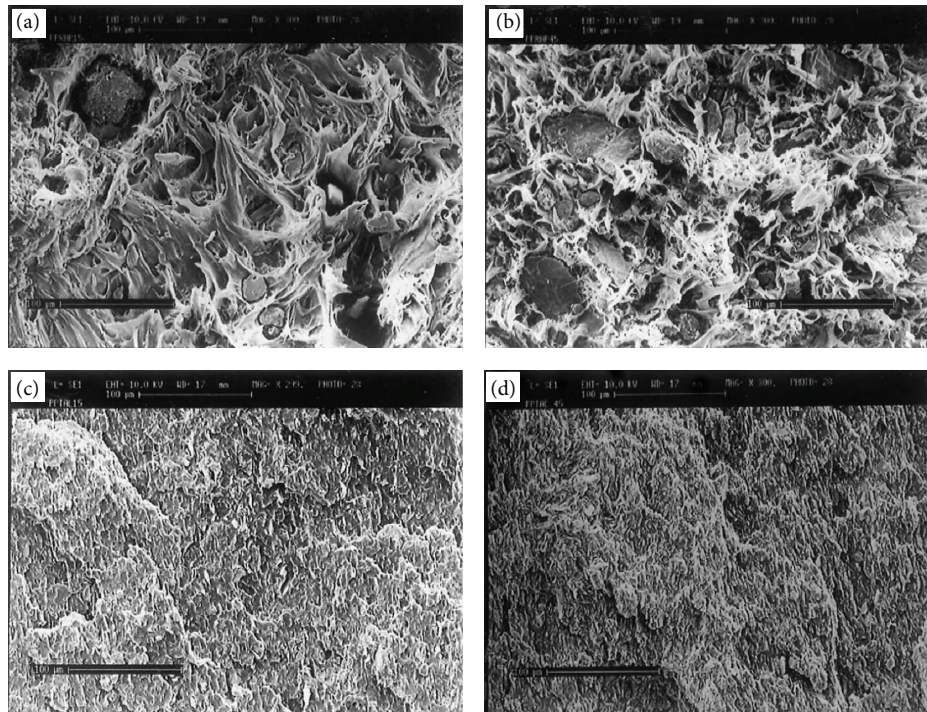


FIGURE 20: SEM micrographs of the tensile fracture surface of (a) and (b) PP/RHP composites and (c) and (d) PP/talc composite at 15 and 45 php of fillers content, respectively (mag.  $\times 300$ ) [21].

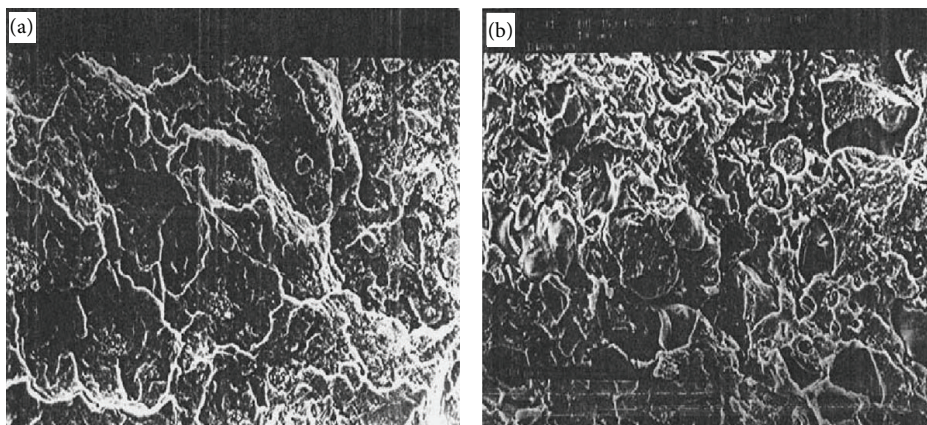


FIGURE 21: SEM micrographs of tensile fracture surface of (a) 40/0 and (b) 20/20 silica/WRHA (wt/wt) of PP/silica/WRHA hybrid composites, respectively (mag.  $\times 300$ ) [33].

evident from the SEM micrographs. Furthermore, it seems that better filler distribution is achieved in talc composites than RHP composites.

As reported by Ismail et al. [33], PP hybrid composites were prepared using silica and WRHA fillers as reinforcing agents. In their study, both fillers were used at the similar total filler content (40 wt%). Ismail et al. (2003) found that the flexural modulus and tensile modulus of the composites increased with increasing of WRHA content in silica/WRHA weight ratio of PP/silica/WRHA hybrid composites, which was attributed to the higher aspect ratio of WRHA compared to silica. However, they noted that the tensile strength,

elongation at break, stress at yield, and water absorption of the composites increased with increasing of silica content in silica/WRHA weight ratio of PP/silica/WRHA hybrid composites. It was reported that the smaller particle size of silica could affect on the interfacial adhesion between silica and PP matrix. Degradation study showed that percentage loss of above properties increases with increasing of WRHA content in silica/WRHA weight ratio of composites. SEM micrographs of tensile fracture surfaces of composites are shown in Figure 21. It can be observed from Figure 21(a) that the fracture surface is rough with many tear lines. The rougher surface structure indicates that failure occurred with

higher tensile strength. Silica fillers were well embedded in PP matrix and good adhesion occurred between silica and PP matrix. In contrast, Figure 21(b) shows the filler agglomerates pull-out, which is responsible for the poor tensile strength and poor dispersion of WRHA in the PP matrix. It can be concluded that the silica has better adhesion with PP matrix compared to WRHA.

Another study presented by Yang et al. [55] showed that RHF-reinforced PP composites were prepared using twin-screw extruder and physical, mechanical, and morphological properties of PP/RHF were investigated. In their study, four levels of filler contents (10, 20, 30, and 40 wt%), six levels of test temperature ( $-30$ , 0, 20, 50, 80, and  $110^{\circ}\text{C}$ ), and five levels of crosshead speed (2, 10, 100, 500, and  $1500\text{ mm min}^{-1}$ ) were designed. By increasing the RHF loading and crosshead speed of test samples, the composite became more brittle. The authors attributed this increase in brittleness with increasing filler loading to the decrease in fraction of thermoplastic polymer and increased interfacial area. Concerning increase in brittleness with increasing crosshead speed, the frictional resistance between filler and matrix polymer decreased at higher crosshead speeds, and this increased the brittleness. The tensile strengths according to the filler loadings presented the same tendency at each test temperature. At the lower test temperatures ( $-30$  and  $0^{\circ}\text{C}$ ), the composites exhibited strong and brittle properties like glass, but the tensile strength drastically decreased as the test temperature was increased from ( $0$  to  $20^{\circ}\text{C}$ ) due to the glass transition of the polymer matrix. The matrix polymer became more ductile and softened as the test temperature increased. Therefore, they suggested that this problem could be reduced by using a compatibilizing or coupling agent. In this experiment, Yang et al. described that by increasing of test temperature the thermoplastic polymer was softened and the composites showed plastic matrix deformation, which decreased the tensile strength and modulus. They reported that, by increasing of filler contents, morphological study revealed more filler particles and increased the numbers of holes where filler particles have pulled out traces (Figure 22).

In the past ten years (2005), our research group [56] reported on the effects of coupling agent and impact modifier on the mechanical properties of PP/RH composites. In our study, as suggested by Yang et al. [55], ethylene 1-octene copolymers (EOC) and MAPP were used as the impact modifier and coupling agent, respectively. These compounds were melt blended using twin-screw extruder and then injection-molded into standard test samples. An improvement of 35% in flexural strength with the addition of 4 wt% of MAPP into the composites with 30 wt% of RH was observed. However, the flexural modulus and impact strength were decreased. Significant enhancement was also found in the impact properties by the incorporation of 20 wt% of EOC into the composites, but the flexural modulus reduced.

In a research by Santiagoo et al. [57], the properties of RHP filled PP/recycled acrylonitrile butadiene rubber (NBRr) composites with and without coupling agent (APS) were investigated. The composites with different RHP filler content (0 to 30 phr) were prepared using a Haake internal mixer. They studied the mechanical properties,

swelling behavior, and water absorption of PP/NBRr/RHP composites. They demonstrated that the addition of RHP to PP/NBRr/RHP composites increased processing torque, tensile modulus, water absorption, and swelling in oil but decreased the tensile strength and elongation at break of the composites. It was also found that, due to the strong bonding between APS treated RHP filler and PP/NBRr matrices, the APS treated RHP composites have higher processing torque, tensile strength, tensile modulus, and lower elongation at break when compared to untreated RHP composites. Furthermore, the APS treatments on RHP improved the adhesion between hydrophilic RHP fiber and the hydrophobic PP/NBRr polymer matrices, which led to reduction of water and oil absorption into PP/NBRr/RHP composites.

Recently, Duy Tran et al. [37] focused on preparation of composites based on PP and Vietnamese RHF (size distribution,  $100\text{--}125\text{ }\mu\text{m}$ ) with the presence of MAPP as a compatibilizer. The researchers have investigated the effect of RHF and MAPP content on properties of PP composites. The results showed that the addition of MAPP to PLA/RHF composites not only did improve tensile and flexural strength but also improved toughness of composites, as confirmed by SEM micrographs (Figure 23). In the absence of MAPP, the composites displayed a rough morphology with appearance of the phase separation between the RHF particles and PP matrix (Figure 23(a)), which clearly showed the poor interaction between filler and matrix. However, in the presence of 4 wt% MAPP (Figure 23(b)), the morphological observation demonstrates that the compatibilizer reduces the separating positions and covered the free spaces between filler and the matrix, which indicated improve adhesion at the interfacial region between filler and matrix. Besides, they found that the thermal stability of composites was also enhanced in the presence of compatibilizer. Their results showed that MAPP acted as the effective factor to improve interfacial adhesion between RHF filler and PP matrix. Duy Tran et al. [37] claimed that Vietnamese RHF is a promising renewable resource to be applied for polymer composites.

**2.4. Rice Husk/Polyvinyl Chloride Composites.** PVC is a thermoplastic polymer. It is a vinyl polymer constructed of repeating vinyl groups having one of their hydrogens replaced with a chloride group. It is the third most widely produced plastic after polyolefin and is medium strong and transparent material, resistant to chemicals, grease, and oil. It is used for sewage pipe lines and other pipe line applications owing to its biologically and chemically resistant characteristics. It is also used for windows, door frames, and such other building materials by adding impact modifiers and stabilizers. It becomes flexible on addition of plasticizers and can be used in cabling applications as wire insulator. Mechanical properties of RH/PVC composites have been studied by various researchers to understand the effect of RH content on strength of the resulting composites and their ability to resist deformation and are shown in Table 10.

In one set of our experiments [62], RHA fillers were added to acrylonitrile butadiene styrene (ABS) impact modified unplasticised PVC (PVC-U) in order to reduce the cost and

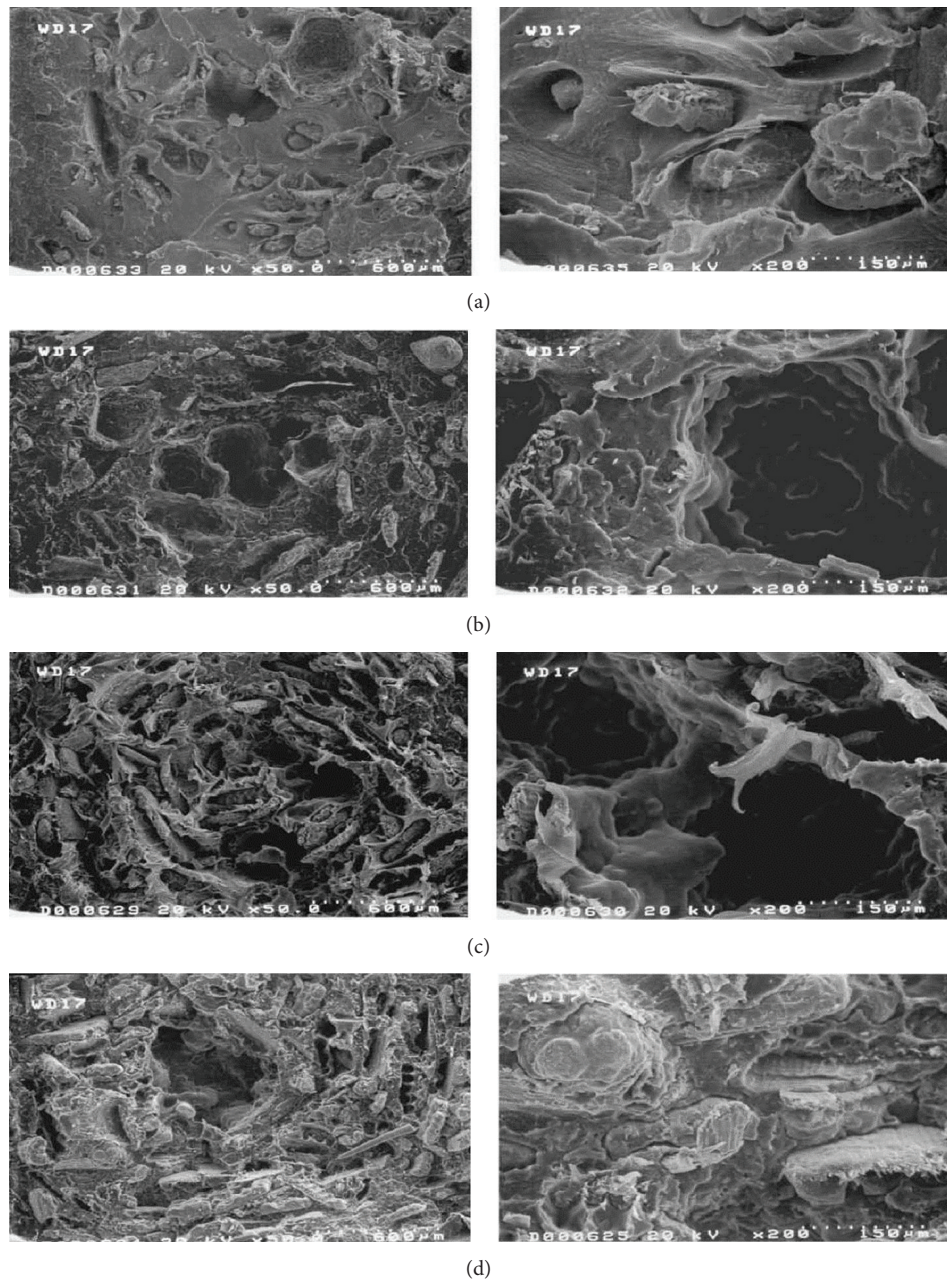


FIGURE 22: SEM micrographs of the tensile fracture surfaces of the PP/RHF composites with different filler contents: (a) 10, (b) 20, (c) 30, and (d) 40 wt% of RHF contents [55].

maintain or improve properties. A processability study was conducted to investigate the fusion behavior of filled impact modified PVC-U using Brabender Plasti-Corder with fitted mixing head. The processability study showed that as the RHA content increased the fusion time increases. In such cases, attempts to analyze and compare the effect of RHA content and various types of coupling agents on the fusion behavior of impact modified PVC-U were carried out. Our work was carried out on the preprepared RHA, with different content levels. It was found that 10 phr RHA filled impact

modified PVC-U samples has shorter fusion time and higher fusion torque compared to other levels of filler contents. Upon the addition of 20 phr RHA, the end torque value decreased compared to unfilled sample. It was also found that the zirconate-treated samples have longer fusion time and lower processing torque compared to other types of coupling agents. Our experimental results also showed that the low contents of the filler have the faster fusion during processing.

Unmodified and modified RHP filled PVC composites with different amounts of RHP content (0, 10, 20, 30,

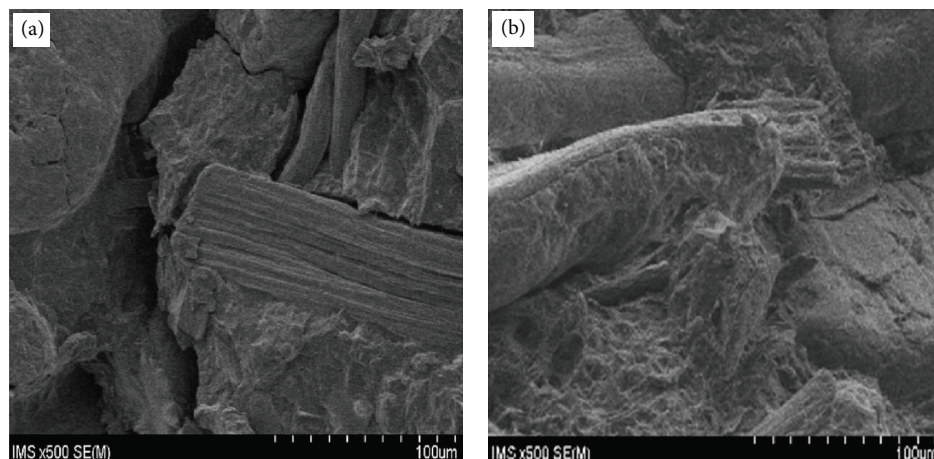


FIGURE 23: SEM micrographs of (a) PP/RHF and (b) PP/RHF with 4 wt% of MAPP [37].

and 40 wt%) were developed by Chand and Jhod [58]. Mechanical, thermal, and electrical properties of these composites were determined. Density of PVC/RHP composites decreased with increasing RHP content in the PVC composites. The tensile strength of PVC/RHP composites was found to decrease with increasing of RHP in the composites (from 33.9 to 19.5 MPa). However, adding of MAPP modified RHP improved the tensile strength of PVC/RHP composites. In addition, the researchers found that the flexural strength and flexural modulus of composites also increased on treatment of RHP due to the improved bonding between RHP and PVC matrix. They reported that Arc-resistance of PVC/RHP composites decreased with increasing of RHP content in the composites. Furthermore, they found that, due to the presence of impurities and water molecules, volume resistivity and surface resistivity decreased with increasing of RHP content in the composites. They confirmed that Vicat softening temperature of PVC/RHP composites increased in both treated and untreated RHP filled PVC composites.

In an intensive study, Xu et al. [59] fabricated PVC and natural fiber composites by melt compounding and compression molding. The influence of fiber type (i.e., bagasse, rice straw, RH, and pine fiber) and content level of styrene-ethylene-butylene-styrene (SEBS) block copolymer on composites properties were investigated. Mechanical analysis showed that storage modulus and tensile strength increased with fiber contents at the 30 wt% level for all composites. SEBS showed moderate effect on tensile strength of PVC/natural fiber composites, but no obvious contributions to impact strength of the composites were observed. However, among the four natural fibers used, rice straw offered better mechanical properties than bagasse, RH, and pine fiber. They reported that the addition of natural fibers decreases the thermal stability of neat PVC but also weakened the self-accelerating effect during degradation of PVC. The researchers also found that the SEBS had little effect on the thermal stability of the composites. After being immersed in water for four weeks, PVC/RH composites showed lower water absorption rate and better dimensional stability in water compared to the other three types of composites. The

results showed that the addition of SEBS increased the water absorption rate of PVC/natural fiber composites. They concluded that PVC composites filled with agricultural fibers had properties comparable with those of PVC/wood composites. They proposed future work with effective coupling agents for PVC (virgin and recycled) and natural fibers.

In the study by Crespo et al. at Polytechnic University of Valencia [60], new composites materials were fabricated, based on lignocellulosic fillers filled thermoplastic composites. In their study, carboxylate plasticizer-di-isononylcyclohexane-1,2-dicarboxylate were used as a plasticizer. Filler amount influenced the PVC/RH composites regardless of particle size (150, 500, or 1000 µm). Mechanical properties and SEM analysis of composites were investigated. They found that although the mechanical properties decreased, the resulted composites became more rigid with increasing filler content in all of the particle formulations. They reported that the behavior of the composites at different plasticizer concentrations was very similar to that of plasticized PVC without filler and also resistance values decreased as plasticizer concentration increased regardless of particle size or filler amount. They concluded that the particle distribution had a considerable influence on the properties of the PVC/RH composites, and it was seen that smaller particles equipped the material with improved mechanical properties due to the better dispersion and alignment of the particles within the PVC polymer matrix, as indicated by SEM micrographs (Figure 24).

In 2010, our research group [38] investigated the effects of RH and acrylic impact modifiers on the mechanical properties of PVC-U composites. The composites were prepared using a two-roll mill at 165°C before being hot-pressed at 185°C. In our study, it was found that the incorporation of RH fillers from 10 to 40 per hundred resins (phr) has increased the flexural and tensile modulus of the unmodified and modified (8 phr impact modifier) PVC-U composites. The flexural strength for both unmodified and modified PVC-U composite was observed to increase until 20 phr of RH content. However, the tensile and impact strength of PVC-U composite decreased with increasing RH content. The SEM



TABLE 10: Mechanical properties of RH filled PVC composites.

Matrix	RH (wt%)	Coupling agent or compatibilizer (wt%)	Tensile strength (MPa) (% incr.)	Tensile modulus (GPa) (% incr.)	Flexural strength (MPa) (% incr.)	Flexural modulus (GPa) (% incr.)	Impact strength (Jm <sup>-1</sup> ) (% incr.)	Reference
PVC	10	—	19.5 (-43)	—	95 (-12)	3.1 (-16)	—	[58]
PVC	30	SEBS (2.5)	33 (61)	—	—	—	15.5 (-38)	[59]
PVC	20	Carboxylate plasticizer* (40)	6.2 (-63)	0.035 (335)	—	—	—	[60]
PVC	20	Acrylic impact modifier* (4)	58 (-2)	1.8 (20)	83 (-8)	4.1 (2.5)	—	[38]
PVC	10	—	19.4 (-43)	—	92.4 (-13)	3.07 (-16)	116 (-30)	[13]
PVC	TRH (10)	—	27.6 (-19)	—	102.2 (-4)	3.52 (-3.5)	140 (-15)	[13]
rPVC	35	—	12.84 (-15)	1.55 (25)	—	—	—	[61]

THR = treated RH using maleic anhydride and benzylperoxide.

\* phr.

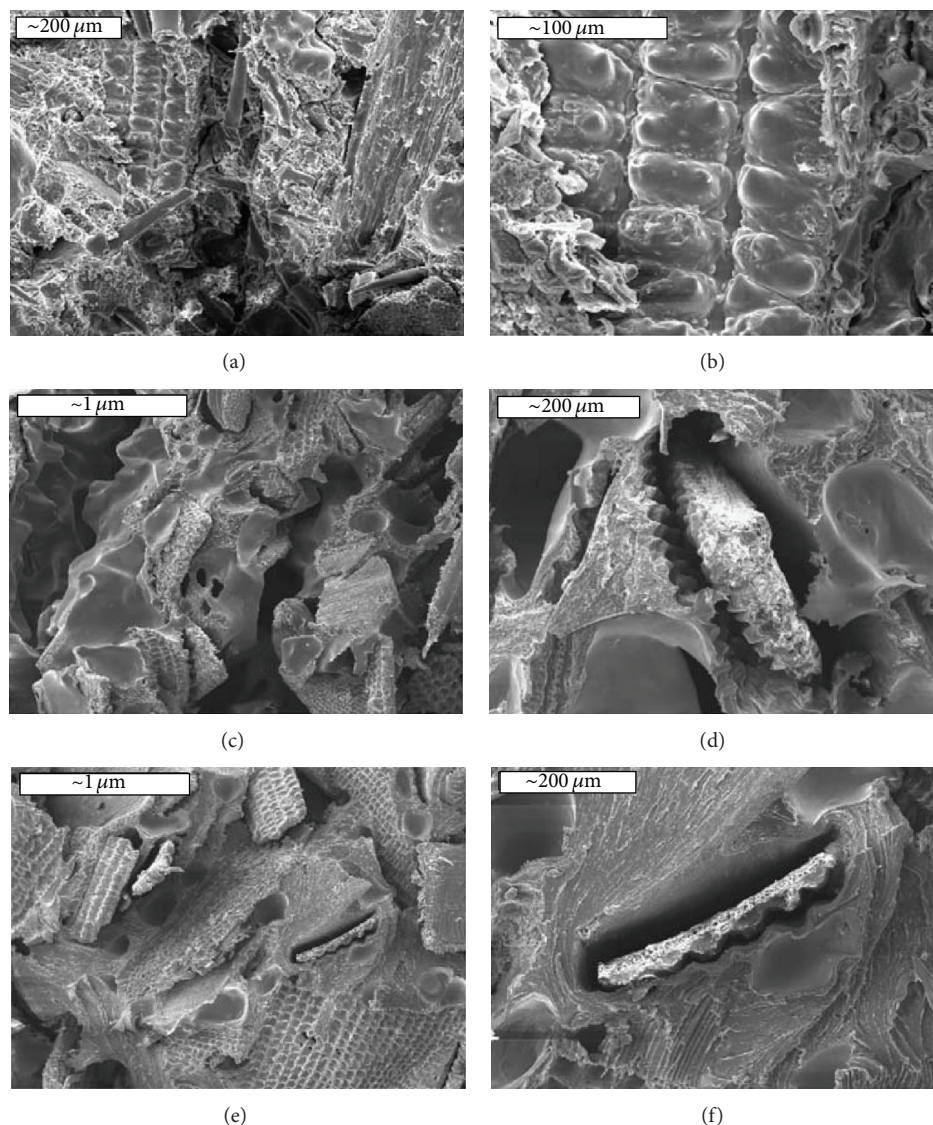


FIGURE 24: SEM micrographs of plasticized PVC containing 20 wt% RH filler with different plasticizer and particle size: (a) 40 phr, 150  $\mu\text{m}$ ,  $\times 150$ ; (b) 40 phr, 150  $\mu\text{m}$ ,  $\times 500$ ; (c) 60 phr, 500  $\mu\text{m}$ ,  $\times 50$ ; (d) 60 phr, 500  $\mu\text{m}$ ,  $\times 150$ ; (e) 60 phr, 1000  $\mu\text{m}$ ,  $\times 50$ ; and (f) 60 phr, 1000  $\mu\text{m}$ ,  $\times 150$  [60].

micrographs showed that the RH fillers agglomerated and unevenly distributed throughout the matrix, as indicated by circles (Figure 25). The result showed that the impact strength of the filled PVC-U composites (20 phr filler) increased, but the tensile and flexural properties decreased with increasing impact modifier content from 0 to 8 phr. It was also found that the formulation containing 8 phr of acrylic impact modifier and 20 phr of RH content has the best balance of stiffness and toughness properties.

In the study by Chand et al. [13], a good interfacial interaction between RH and PVC has been achieved by modifying the surface of the former using MAPP as a compatibilizing agent. In their study, the positive effect of RH reinforcement as well as surface treatment has been reflected in the improved tribological and mechanical properties of the resulted PVC composites. According to Chand et al.

[13], the filler concentration should be kept to a minimum if optimum improvement is desired. Therefore, low filler concentration of RH (10 wt%) was proved to be the optimum concentration to achieve maximum strength as well as tribological properties. However, they reported that the larger filler concentration results in larger filler-filler interaction, inhomogeneous dispersion, and inadequate wetting of filler by polymer resin. They noted that the wear resistance of RH filled PVC composites can be increased if the surface of RH is treated suitably with a coupling agent. They observed that, by increasing abrasive grit size, the wear rate increased. SEM micrographs were used to understand the wear resistance of MAPP modified RH filled PVC composites (Figure 26). It is also reported that, with increasing sliding distance, wear rate gradually decreased and attains an almost steady state in multipass condition. Size effect was also observed

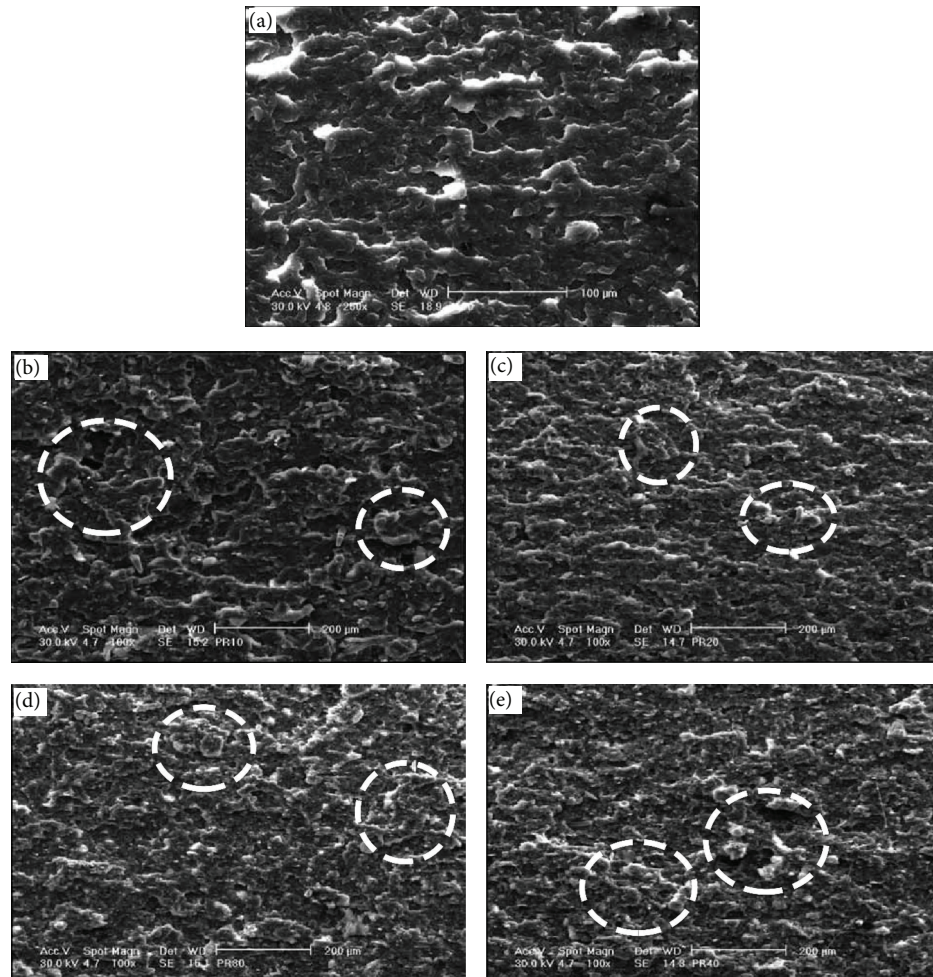


FIGURE 25: SEM micrographs of impact fracture surfaces of (a) unfilled unmodified PVC-U; (b) RH filled impact modified PVC-U composites at 10, (c) 20, (d) 30, and (e) 40 phr of RH content [38].

for composites having 40 wt% of modified filler. Lancaster-Ratner correlation was found to be more linear for MAPP-treated RH filled PVC composites compared to untreated RH/PVC composites. They concluded that probably this is due to increased filler-matrix adhesion upon treatment in the case of the former.

In another study by Chand et al. [63], the influence of a foaming agent on the mechanical and wear behavior of surface modified RH filled PVC composites has been studied. In their study, the surface of RH was modified using MAPP and benzylperoxide for better coupling with the PVC polymer. In Chand et al.'s [63] research, six composites having 10, 20, and 30 wt% of unmodified and surface modified RH having a small percent of a foaming agent have been investigated and compared with foamed PVC. It was observed that with an increase in RH content wear resistance as well as compressive strength of composites increased. They found that wear resistance of PVC foamed composites further improved with surface treatment of RH by MAPP, due to improvement in compatibility between RH and PVC compound, as confirmed by SEM analysis (Figure 27). They proposed a correlation

between compressive strength and wear resistance and also they reported that foaming agent reduces the density of composites.

In another work by Chand and coworkers [64], the effect of addition of RH in PVC composites on the dielectric properties at different frequency and temperature has been investigated. Measurements have been performed in the frequency range of 1–10 kHz and temperature range of 32–80°C. Chand et al. [64] proposed a new reasonable model based on existing equations which was experimentally verified. In their investigation, frequency and temperature dependent parameters were experimentally obtained and taken as contribution to the proposed model. The dielectric constant for composites with various fibers to matrix ratio was determined with this model. They reported that dielectric constant of the composite was highly influenced by the fiber composition. The experimental results showed that dielectric constant ( $\epsilon'$ ) increased with the addition of RH in PVC and decreased with increasing frequency, which indicated that the major contribution to the polarization came from orientation polarization. Additionally, they demonstrated that the

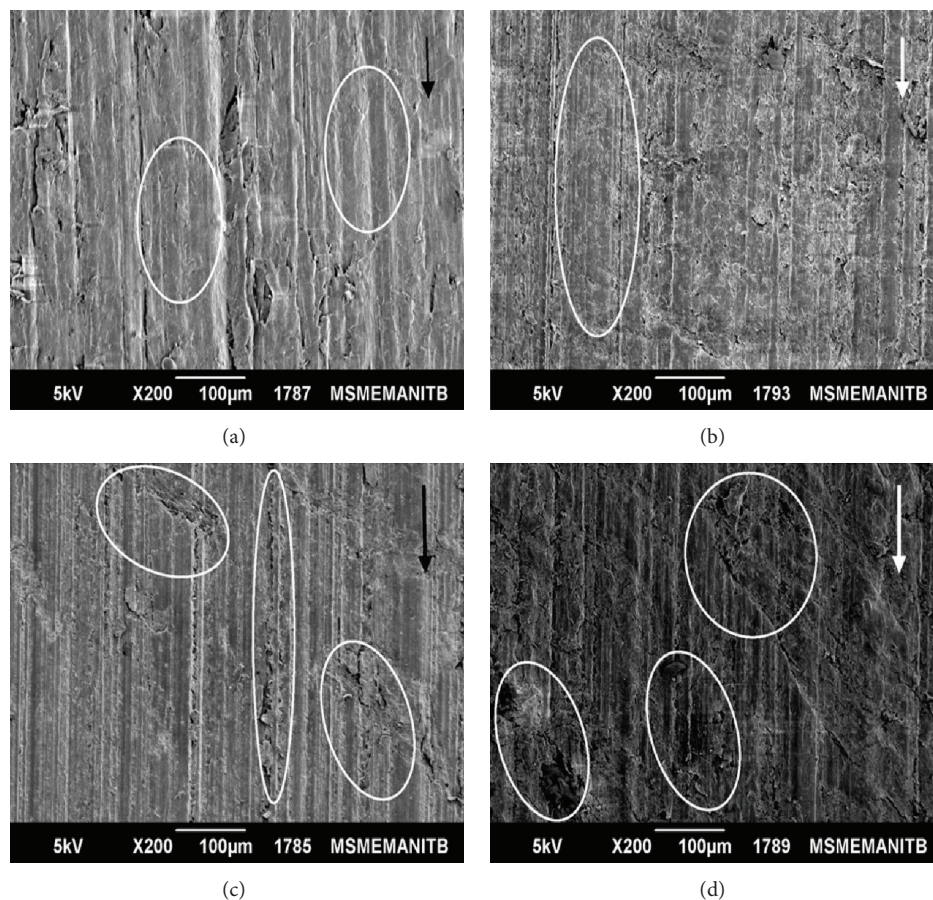


FIGURE 26: SEM micrographs of maleic anhydride modified RH filled PVC composites: (a) PTRH10 and (b) PTRH30; unmodified RH filled PVC composites: (c) PRH20 and (d) PRH40; with the sliding speed of 10 s, sliding distance of 6.28, and applied load of 10 N. (P = PVC; T = treated (modified) and downward arrow shows sliding direction) [13].

dielectric constant ( $\epsilon'$ ) increases with increasing RH content temperature due to greater freedom of movement of dipoles within PVC at higher temperatures. Researchers reported that the proposed frequency and temperature dependent model to calculate the dielectric constant ( $\epsilon'$ ) of the RH filled PVC composites matches the experimental results. They also found that  $\text{Tan } \delta$  peaks at different frequencies are dominant in case of 30 and 40 wt. % of RH composite.

A recent study by Ramle et al. [61] demonstrated good interfacial interactions between RH and recycled PVC (rPVC) by incorporation of aminosilane as coupling agent. The aminosilane-treated rPVC was mixed with RH and then fabricated using hot compression molding. Three different ratios of RH:rPVC (65:35, 55:45, and 45:55) were used to prepare RH/rPVC composites. The positive effect of RH reinforcement as well as surface treatment has been reflected in the improved tensile properties of the resulted rPVC composite. However, the tensile properties of RH/rPVC composites decreased with the increasing RH content. They claimed that this is due to the poor adhesion between RH and rPVC matrix. Ramle et al. (2013) reported that the flexural stress of the treated composite decreased with increasing

RH content. However, the incorporation of aminosilane as coupling agent caused to significantly increase the tensile stress of RH/rPVC composites with 45 wt% of RH content. SEM micrographs of RH/rPVC composites are shown in Figure 28. The incompatibility between RH and PVC can be clearly observed, as indicated by the circle (Figure 28(a)), resulting in low tensile strength. However, better adhesion and bonding between the RH and PVC were achieved after the addition of aminosilane, as indicated by the circle (Figure 28(b)). Based on these findings, it is beneficial to carry out further study in the development of RH filled rPVC waste composites.

### 3. Conclusion and Future Prospects

This review aims to highlight previous works on RH filled polymer composites and provides relevant data for several applications as well as further research areas. RH has been used in combination with various polymeric matrices including PE, PP, PVC, and PLA to form polymer composites. RH filled polymer composites were prepared by different

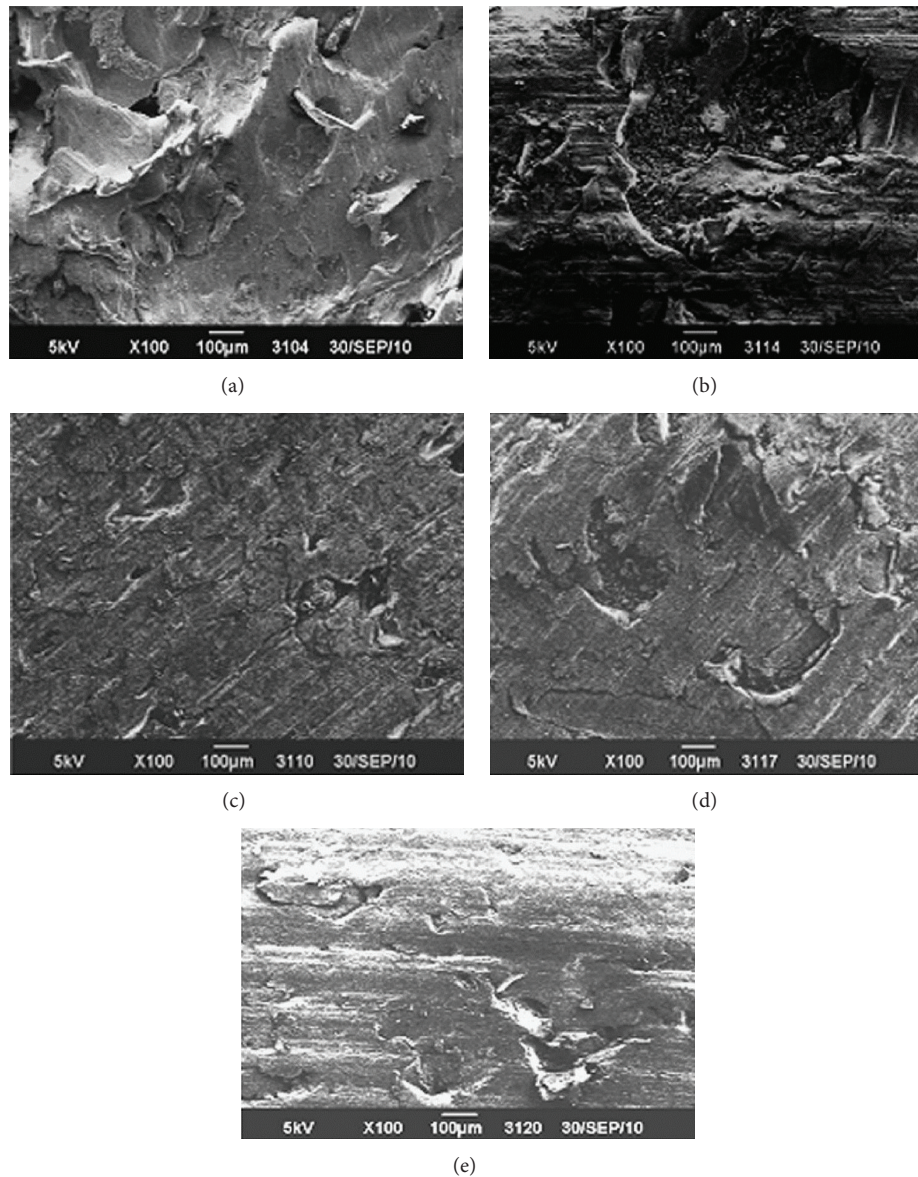


FIGURE 27: SEM micrographs of worn surfaces of foamed PVC without RH: (a) FPRH0; RH filled foamed PVC: (b) FPRH2 and (c) FPRH3; MAPP modified RH filled PVC composites: (d) FPTRH2 and (e) FPTRH3. (P = PVC; T = treated; F = foaming agent) [63].

methods, for example, single screw extruder, corotating twin-screw extruder, internal mixer, and compression molding. The different preparation methods have resulted in composites with different mechanical, physical, thermal, and morphological properties. The addition of RH resulted in reduced tensile strength of composites. Therefore, the use of various types and amount of coupling agent/compatibilizer (approximately  $\leq 5$  wt%) have been reported in order to improve the mechanical properties of composites. Furthermore secondary fillers such as MMT along with compatibilizers have been used to improve mechanical properties of composites. Interestingly, RH filled PE composites have been found to possess better overall mechanical properties compared to other composites such as PLA, PP, and PVC.

Based on this review, the application of RH filled polymer composites as alternative composites materials in building and construction is highly plausible with both lightweight and low cost as its main driving forces. For such applications, biodegradability and weathering properties of the composites need to be evaluated. Although many studies have reported on the use of coupling agent and compatibilizers, further investigations on physical and chemical treatment to further improve the interfacial adhesion with polymeric matrix are needed. The reason is that fiber-polymer interaction is important in determining the final composite properties. So far only single polymer has been used as the matrix in RH filled composites. Limited work has been reported on the use of secondary fillers. Therefore, better understanding of the

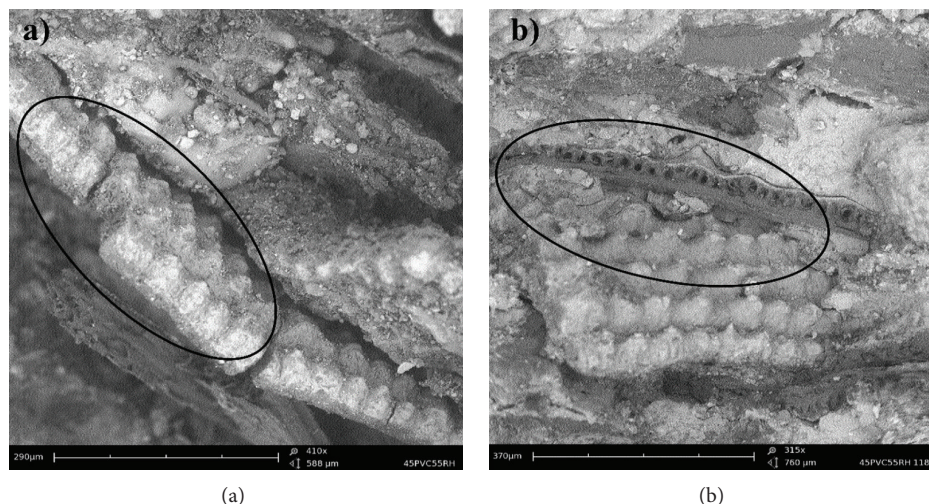


FIGURE 28: SEM micrographs of RH/rPVC composites (55% RH + 45% rPVC) (a) without the addition of aminosilane and (b) with addition of aminosilane [61].

use of polymer blends as the matrix and secondary fillers in controlling the properties of polymer composites would provide interesting areas to be investigated.

### Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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