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ENHANCING MECHANICAL AND FLAMMABILITY PROPERTIES OF RICE HUSK-REINFORCED RECYCLED HIGH-DENSITY POLYETHYLENE COMPOSITES THROUGH CHEMICAL TREATMENT

This study investigated chemical treatment's impact on mechanical and flammability properties of rice husk-reinforced recycled high-density polyethylene (r-HDPE) composites. Three treatments (maleate, alkali, acid) were applied to rice husk, and composites were tested for tensile strength, elongation at break, and modulus of elasticity. Results showed that tensile strength decreased with increasing rice husk content, except for composites treated with maleic anhydride grafted polyethylene (MAPE), which exhibited the highest strength at 10% filler content. Break elongation generally decreased with increased rice husk content, while the modulus of elasticity improved with higher rice husk content, except for acid-treated composites. Scanning electron microscopy revealed better interfacial bonding in composites with lower rice husk loading. Maleate treatment enhanced flame resistance. In conclusion, chemical treatment has the potential to enhance the properties of rice husk-reinforced r-HDPE composites for various applications.

Keywords: Recycled high-density polyethylene; rice husk; chemical treatment; flammability properties; composites; mechanical properties; sustainable materials

1. Introduction

Recently, natural organic fibres like rice husk, wood, wool, hemp, and banana leaf show the potential as alternative reinforcing materials for the production of glass fibre, carbon fibre, aramid fibre, and inorganic fillers. These fibres have advantages in terms of low cost, economic, lightweight, renewable, biodegradable, non-toxic and safe, increased specific strength, modifiable surface, wide applications, and tough properties [1-5]. Due to the high quantity of products, many studies have been conducted to fulfil the wide field of composite materials.

Rice husk is a natural waste product used as fertilisers and the material is important in concrete application, where rice husk consists of 74% organic content and 26% inorganic content by weight [6]. Previously, rice husk has also been applied for thermoplastic reinforcement. The work eventually produced plastics with distinct mechanical properties. However, incompatibility was observed with the hydrophilic lignocellulosic filler as the hydrophobic matrix. Thus, coupling agents such as maleic anhydride polyethylene are required to modify the interface [7].

Rice husk fibre is added to plastic waste to produce green composites and the advantages include low cost, non-hazardous, lightweight, and accelerate the biodegradability of polymeric composites [8]. Green composites are used to resolve environmental problems and produce the most effective properties for products [9]. This research investigates the composites prepared by using recycled high-density polyethylene (r-HDPE) and rice husk fibre. The reinforcing filler and thermoplastic materials as matrices are currently receiving much interest where the materials will eventually be developed into eco-friendly products with a wide range of different physical properties. These materials are also combined with polymer matrices to improve the properties. The natural reinforcing filler is utilised because the filler is cheap and environmental pollution can be prevented [10].

Lignocellulosic materials like rice husk are used as fillers, and r-HDPE serves as the matrix to produce composites for

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mechanical property testing. Poor interfacial bonding between these materials is mitigated by using maleic anhydride grafted polyethylene (MAPE) as a coupling agent, facilitating interfacial bonding with the hydrophilic filler and improving wetting properties of the hydrophobic polymer [11,12].

The low cost of plastics gives many advantages to people and the society; thus, people overuse plastics drastically every day. Plastics are also used in construction, electrical and electronic applications, automotive industry, as well as medical equipment. Chemicals like stabilisers or colorants that are used to produce plastics are harmful to living things. This will cause a negative impact on human health and the environment, as well as animals and plants. These unwanted plastic products also contribute to environmental pollution. Non-degradable plastics require a longer period if the plastics are dumped in landfills. In this research, the landfill space for plastic waste is the main concern in order to prevent problems in landfills. Researchers are taking initiatives to produce useful products from recycled plastics. Eventually, production costs and energy can be saved as plastic waste is reused, recycled, and reduced in order to produce a clean environment.

With the objective to determine the best way to improve the efficiency of rice husk-reinforced r-HDPE composites, this study investigates the effects of several treatments on the mechanical and flammability properties of the materials. In order to enhance the interfacial bonding between lignocellulosic fillers and the polymer matrix, coupling agents like MAPE will be investigated. The purpose of using these different methods is to investigate their effects on the compatibility of fillers with polymers. By reducing plastic waste and enhancing recycling efforts, this research aims to make green composites appropriate for a variety of applications while also promoting environmental sustainability.

2. Materials

2.1. Raw materials

Rice husk fibre (RH) that act as reinforcing fibre was supplied by Kilang Beras Sukaramai Sdn Bhd, Yan, Kedah with particle size of 100 μm and standardized wood thermoplastic composite (WTC) grade. The solid density of rice husk was 1.5 g/ml and the moisture content was 6.62%. The materials used for the matrix polymer were the recycled high-density polyethylene (r-HDPE) with 923 kg/m^3 density, 0.72 g/10 min of melt flow index at 190°C from BioComposite Extrusion Sdn. Bhd.

2.2. Chemicals

Maleic anhydride grafted polyethylene (MAPE) is a coupling agent for enhancing the interfacial adhesion between composites. MAPE has melt index of 5 g/10 min at 190°C, the density of 0.92 g/ml at 25°C, and melting temperature of 135°C. MAPE is insoluble in water and in pellet form. MAPE was obtained from Aldrich-Chemistry. Hydrochloric acid (HCl) was used in acid treatment of rice husk. HCl is a strong acid with odour and the solvent is completely soluble in water. Meanwhile, sodium hydroxide (NaOH) was used in alkali treatment of rice husk. NaOH is a strong alkali and very corrosive, which can cause eye and skin irritation. Furthermore, NaOH is soluble in water.

3. Procedures

3.1. Strong acid treatment

HCl was used to perform acid treatment on the filler. HCl solution was prepared using 1 M HCl. 100 g of rice husk was soaked in HCl solution for 1 h at 80°C. The soaked rice husk was then filtered and washed with distilled water to prevent excess acid trapped on the surface of rice husk upon reaching neutral pH. The acid-free rice husk was dispersed widely over an aluminium tray and placed in an oven at the temperature of 105°C.

3.2. Alkali treatment

100 g of clean and dried rice husk were treated by soaking the husk in 1 liter of NaOH solution and boiled at 100°C for 1 h with constant stirring. After that, the alkali-treated rice husk was left to cool and then filtered. Next, the filtered rice husk was washed by distilled water to inhibit further treatment by the alkaline residue. The treated rice husk was placed in an oven for 1 day at the temperature of 105°C for drying purpose. Finally, the treated rice husk was transferred into a sealed polybag.

3.3. Maleate treatment on rice husk

70 g of rice husk was mixed with 1.5 liter of water for 15 min at room temperature with constant stirring. After that, the solids were separated by using filter paper and dried in an oven for 2 days. MAPE was prepared for this experiment. The weight of rice husk, r-HDPE, and MAPE was determined and calculated in grams. r-HDPE was premixed with rice husk and MAPE before being added to a twin-screw extruder. A co-rotating twin-screw extruder was used to compound r-HDPE with rice husk. The process prior to compounding was conducted to confirm and maintain MAPE blend homogenously in the composites. The extruded strand was pelletised and stored in polybags.

3.4. Composite Compounding

r-HDPE and rice husk were compounded by using a twin-screw extruder with screw speed of 50 rpm. Different temperatures were set at each different zone. The first, second, third, and last zones were set to 180, 190, 200, and 190°C, respectively. The compounded composites were converted into long strands by the extruding machine. The long strands were then pelletised by using a pelletiser. After that, the pelletised compounds were placed in an oven for drying to remove moisture that can cause voiding and affect the properties of the compounds. After drying, the pelletised compounds were placed in a compression machine. The temperature of the machine was set at 190°C for 8 min, followed by cooling of the samples for 5 min. TABLE 1 presents the composite formulation based on the chemical treatments.

TABLE 1

Composite Formulation

Chemical Treatment	Sample Notation	r-HDPE (wt.%)	Rice Husk (wt.%)	MAPE (phr)
Without Treatment	W-10	90	10	0
	W-20	80	20	0
	W-30	70	30	0
Acid Treatment	AC-10	90	10	0
	AC-20	80	20	0
	AC-30	70	30	0
Alkali Treatment	AK-10	90	10	0
	AK-20	80	20	0
	AK-30	70	30	0
Maleate Treatment	M-10	90	10	3
	M-20	80	20	3
	M-30	70	30	3

4. Testing

4.1. Tensile test

Tensile testing, following ASTM D-638 standards, assessed mechanical properties, including tensile strength, elongation at break and modulus of elasticity. Five samples per treatment ratio were tested using an Instron 5569 Universal Testing Machine at a constant speed of 10 mm/min and room temperature. Dumbbell-shaped samples (156 mm length, 130 mm width, 3 mm thickness) were gripped and pulled until failure, with results analysed using the instrument software.

4.2. Fourier transform infrared microscopy

Fourier transform infrared spectroscopy (FTIR) is the analytical technique that provides the fingerprint of a sample based on IR spectra. The IR spectroscopy works by illuminating samples with wavelength from 4000 cm^{-1} to 600 cm^{-1} and the amount of light transmitted by the samples is measured. In this research, Perkin-Elmer 2000 spectrometer was used to obtain the results. Samples of rice husk subjected to different chemical treatments after tensile test were chosen for FTIR testing.

4.3. Scanning Electron Microscopy

A JEOL JSM-6460LA SEM was used to study fracture surfaces and phase separation of the filler in polymer matrix in this research. Besides, SEM was carried out to determine particle size, shape, and texture of the samples. Samples of rice husk subjected to different chemical treatments after tensile test were chosen for SEM testing.

4.4. Flame Test

Flame test is one of the important processes used to identify burning properties and resistance. In this study, specimen bars were pushed towards 125-mm flame according to ASTM 5048-90 to conduct the test.

5. Results and discussion

5.1. Tensile strength

TABLE 2 presents the tensile strength of different rice husk loadings (i.e., 10%, 20%, and 30%) with different chemical treatments and without treatment. The treatments involved are divided into maleate, HCl, and NaOH treatments, and also the untreated set. From the results, tensile strength is affected by both rice husk loading and also chemical treatment. The maleate treatment with the addition of 10% rice husk in the matrix of r-HDPE composites exhibited the highest tensile strength compared to other chemical treatments and rice husk contents. Meanwhile, 10% rice husk without treatment demonstrated the second highest tensile strength.

TABLE 2

Tensile strength for different loadings of rice husk with different chemical treatments of polymer composites

Rice Husk Loading (wt.%)	Tensile Strength (MPa) for Different Chemical Treatments			
	Maleate	Alkali	Acid	Untreated
10	13.255	7.833	6.740	12.909
20	12.975	6.414	6.452	9.547
30	12.134	6.767	6.543	9.538

Tensile strength decreases with increasing filler content. The highest tensile strength is observed at 10% filler content, followed by 20% and 30% rice husk filler content. MAPE treatment results in the highest tensile strength (13.255 MPa) due to enhanced interfacial bonding between r-HDPE and rice husk, with MAPE acting as a coupling agent [13]. 20% and 30% maleate treatment of rice husk give the tensile strength of 12.975 and 12.134 MPa, respectively. Meanwhile, the untreated set with 10% filler content achieved the tensile strength of 12.909 MPa, followed by 20% and 30% rice husk content with tensile strength of 9.547 and 9.538 MPa, respectively. This is because the proper mixing of polymers and impurities in the matrix definitely achieved strong bonding and thus resulted in high tensile strength. MAPE as the coupling agent does not just modify and adjust the surface of the filler, but also the matrix surface and hence, produced interfacial chemical bridge to achieve strong interfacial bonding. Thus, this proves that the tensile strength of maleate treatment is greater compared to the untreated set [14]. Based on acid and alkali treatments, the results are slightly different from the untreated set and maleate treatment of the filler because the tensile strength increased as the rice husk filler loading increased from 20% to 30%. The tensile strength for acid treatment increased from 6.414 to 6.767 MPa, whereas the tensile strength for alkali treatment recorded 6.452 MPa for 20% filler content and increased to 6.543 MPa for 30% filler content. Alkali and acid treatments remove lignin, hemicellulose, fats, and waxes from the surface of the filler, thus producing rice husk with a rougher surface [15]. This may also be due to insufficient wetting caused by rice husk and thus, this behaviour results in weak stress transfer between them. 10% filler content by NaOH and HCl treatments gives the tensile strength of 7.833 and 6.740 MPa, respectively.

5.2. Elongation at break

According to TABLE 3, the analysis showed that filler content and different chemical treatments can also affect elongation at break. The characteristics of natural plant fillers depend on factors like cellulose content, a high-strength polymer, and microfibrillar angle, indicating the orientation of cellulose fibrils in the major wall concerning the stress axis. The elongation at break results reveals percentages of 7.98% for sugar palm, 1.6-6% for hemp, and 1.16-8% for jute fillers [16]. The break elongation of the composites decreased slightly with the increase of rice husk content. 10% filler loading with maleate treatment indicated the highest elongation with the value of 10.7% compared to alkali treatment, acid treatment, and without treatment of filler loading. 20% and 30% filler loadings with maleate treatment achieved the break elongation values of 8.5% and 5.2%, respectively. This is due to the occurrence of good adhesion between the surface of the matrix and the filler [17].

TABLE 3

Elongation at break for different loadings of rice husk with different chemical treatments of polymer composites

Rice Husk Loading (wt.%)	Elongation at Break (%) for Different Chemical Treatments			
	Maleate	Alkali	Acid	Untreated
10	10.7	8.4	7.6	8.5
20	8.5	5.9	5.7	7.1
30	5.2	4.2	4.1	4.6

Meanwhile, for alkali and acid treatments, the elongation at break did not differ significantly but there is a similarity between them, which is the reduction of break elongation with increasing filler loading. For alkali treatment using NaOH on rice husk filler, 10% filler content produced 8.4% elongation at break, followed by 5.9% and 4.2% elongation at break for 20% and 30% filler loadings of the composites, respectively. From TABLE 3, it is clearly indicated that acid treatment by using strong acid (HCl) treatment produced the lowest reading compared to other treatments. The elongation at break decreased as the interaction of r-HDPE polymer chain and RH overcomes the weak filler matrix adhesion. 10% filler content using HCl treatment achieved 7.6% elongation at break, followed by 5.7% and 4.1% elongation at break for 20% and 30% filler loadings, respectively.

For the untreated, 10% loading of rice husk produced 8.5% elongation at break, and the value decreased to 7.1% and 4.6% for 20% and 30% filler contents applied in the polymer composites, respectively. This result shows that the reading dropped due to the brittle behaviour of the filler and thus reduced the ductile properties in the r-HDPE matrix in composites. From this result, MAPE acts as a coupling agent to improve the break elongation properties between the matrix and lignocellulosic material; therefore, the maleate treatment of rice husk has the highest reading among different treatment samples and the untreated set [8].

5.3. Modulus of elasticity

TABLE 4 present the modulus of elasticity of different rice husk loadings using three types of chemical treatments and also the untreated set. When the rice husk content increased from 10% to 30%, the modulus of elasticity also improved. 10% and 20% rice husk contents achieved the modulus of elasticity values of 644.2 and 767.6 MPa, respectively. Meanwhile, the highest reading (i.e., 937.8 MPa) was obtained for 30% rice husk loading, and this indicates the increasing stiffness of the polymer composites. This is due to the greater stiffness of rice husk and lower stiffness of the r-HDPE matrix. It clearly demonstrates that micro-spaces partially separated as stress was applied to the bio-composites. The greater increase in stiffness is due to the presence of rice husk content and the degree of obstruction also increased [18].

TABLE 4

Modulus of elasticity for different loadings of rice husk with different chemical treatments of polymer composites

Rice Husk Loading (wt.%)	Modulus of Elasticity (MPa) for Different Chemical Treatments			
	Maleate	Alkali	Acid	Untreated
10	644.2	644.1	637.1	573.7
20	767.6	864.2	764.3	578.3
30	937.8	832.5	791.5	625.1

For alkali treatment, the modulus of elasticity values increased from 644.1 to 864.2 MPa for the increase of filler content from 10% to 20%. On the other hand, as the rice husk content increased to 30%, the modulus elasticity decreased to 832.5 MPa. Insufficient wetting of r-HDPE to fibre is the cause for this phenomenon. Based on the analysis for acid treatment, the modulus of elasticity values of 637.1 and 764.3 MPa were obtained for 10% and 20% rice husk loadings, respectively. Meanwhile, 30% rice husk loading produced the highest modulus of elasticity among the three different loadings with the value of 791.5 MPa.

The untreated set recorded the lowest modulus of elasticity readings among the four different types of treatment for 10%, 20%, and 30% rice husk loadings, with the values of 573.7, 578.3, and 625.1 MPa, respectively. This proves that the modulus of elasticity increases as the content of rice husk filler increases.

5.4. Fourier Transform Infrared

FTIR is an important technique or instrument to detect the presence of certain functional groups in a molecule. It is also used to provide the information about organic chemical bonding. This technique is useful to identify the infrared spectrum of the polymer composites in order to study the structure across the functional groups formed in the composites. In this research, different surface treatments such as alkali, acid, and maleate treatment, as well as untreated samples were analysed.

Fig. 1 shows the structural comparison of rice husk reinforced with r-HDPE polymer composites. The FTIR spectra represent the polymer composites of M-20 (maleate treatment), AK-20 (alkali treatment), AC-20 (acid treatment), and C-20 (untreated sample). The absorption band in the range about 3354 cm^{-1} is associated with the acid group in the rice husk reinforced with r-HDPE composites. The spectrum from AC-20 also showed absorbing peaks in the range of approximately 2900 cm^{-1} for the vibration of carbon-hydrogen bond (C-H) [19].

However, for 20% rice husk with alkali treatment (AK-20, shown in blue line), the spectrum displayed a broader peak of hydroxyl group O-H (strong H-bonded) stretching band at 3400 cm^{-1}. The broader peak represents the high concentration of hydroxyl group contributed by NaOH. Meanwhile, a peak at 1655 cm^{-1} is attributed to the H-O-H group [20]. The alkali treatment also shows an absorption band at approximately 2845 cm^{-1} to 2900 cm^{-1}, which are contributed to the carbon-hydrogen bond (C-H) vibration. The peaks in the alkali treatment of rice husk spectrum are present due to the release of natural fats and wax from rice husk, and thus the compounds are increasingly exposed on the filler surface [21].

Furthermore, alkali and acid treatments demonstrate an important role to eliminate hemicellulose, wax, and lignin on the surface of rice husk. For the maleate treatment of rice husk (M-20, shown in red line), a weak and broad band at 1032 cm^{-1} is present due to the occurrence of the grafted maleic anhydride group (MAPE) in the polymer composites of rice husk reinforced with r-HDPE. This represents the stretching of C=O bond, which is absent for acid treatment, alkali treatment, and the untreated set. Anhydride usually gives two stronger bands of C=O group with different relative density between 1855 cm^{-1} to 1800 cm^{-1}, and 1782 cm^{-1} to 1720 cm^{-1}. Cyclic anhydrides always move the absorption to a high frequency. Meanwhile, the spectrum in the region of 960 cm^{-1} to 1320 cm^{-1} showed strong and broad C-O stretch vibration. This is because ester bonds are formed between the anhydride group of MAPE and the hydroxyl group of the filler, and caused esterification reaction. Esterification occurs due to the presence of coupling agents and thus, the peak was not observed for acid and alkali treatments.

For the untreated set, which is C-20, the absorption band in the region between 2849 cm^{-1} and 2911 cm^{-1} is associated with the stretching of carbon hydrogen (C-H) bond vibration. Meanwhile, another peak at 1459 cm^{-1} and in the region of 2849 cm^{-1} and 2911 cm^{-1} represent the vibration of C=O bonds.

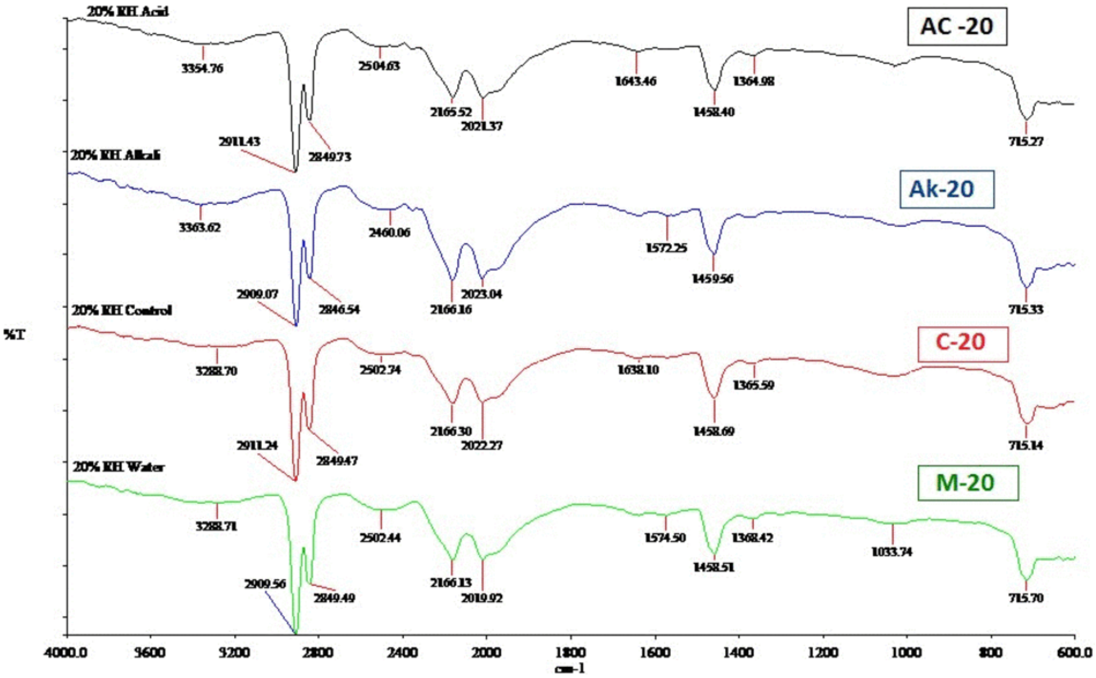

Fig. 1. Structural comparison of rice husk reinforced with r-HDPE polymer composites

The peak at 720 cm^{-1} shows the vibration of O-Si-O stretching, which represents the silica content in rice husk. The relief of silica during chemical treatment and the degradation of cellulose embedded within silica for rice husk can contribute to the low intensity or disappearance of absorption peaks [21].

5.5. Scanning electron microscopy

Figs. 2(a-d) show the morphology of 10% rice husk loadings, and Figs. 3(a-d) present the morphology of 20% with three chemical treatments and an untreated set. These figures illustrate surface fracture morphologies of rice husk reinforced with r-HDPE, examining interactions in different bio-composites. The SEM image of Fig. 2(a) shows a smooth surface on the outside region of the rice husk filler and the inside region is made up of aligned fibre, which is mainly associated with the incorporation of silica onto the cellulose. Fig. 2(b) indicates a smooth surface on both regions and it is clearly observed that fibre is well embedded in the r-HDPE matrix. This represents excellent interfacial adhesion between the rice husk and r-HDPE polymer composites due to the presence of 10% rice husk loading in this sample. Hence, it is proven that rice husk filler is relatively well wetted by the r-HDPE matrix and thus produced composites with higher tensile strength compared to other loadings of similar treatment.

The microstructure fracture surface in Fig. 2(c) represents the untreated set, in which many cavities and pulled-out filler are observed in the HDPE matrix. These phenomena revealed the weak and poor interfacial bond for the rice husk reinforced with the r-HDPE matrix. Besides, the weak dispersion of rice husk fibre in the matrix also proved that the bunch of filler is localised and the patches of matrix occurred. The sample with 10% loading for maleate treatment using water in Fig. 2(d) revealed better and good interfacial phases between the polymer

Fig. 2(a). 10% rice husk by acid treatment

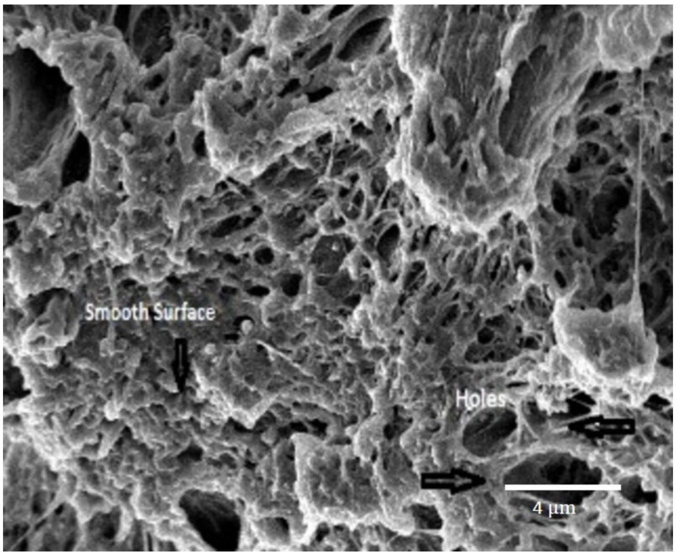

Fig. 2(b). 10% rice husk by alkali treatment

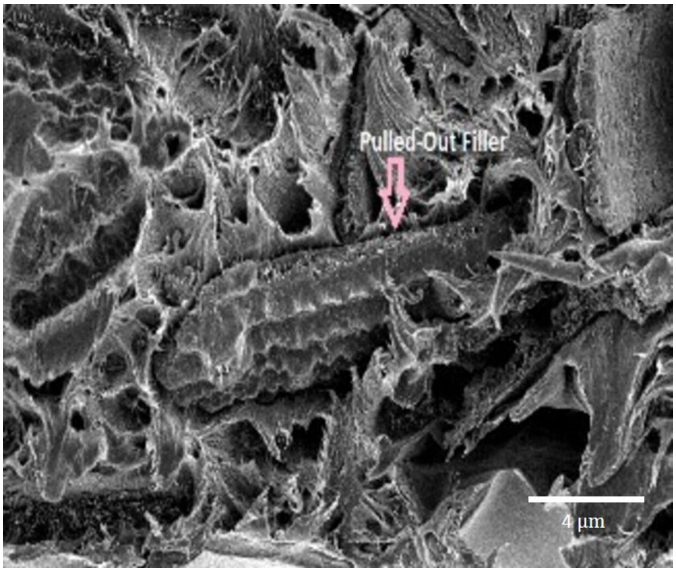

Fig. 2(c). 10% rice husk without treatment

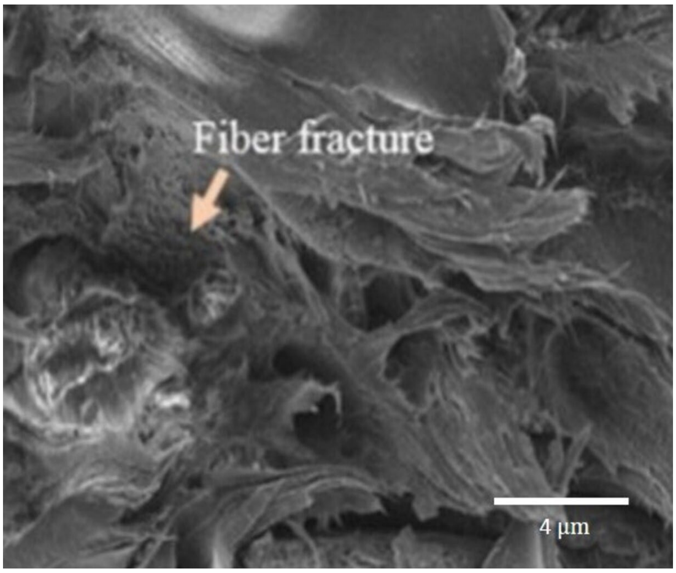

Fig. 2(d). 10% rice husk by maleate treatment composites. The reason is due to removal of impurities by water from the cellulose fibre and thus, the roughness between them increased [21].

Figs. 3(a-d) show the fracture surface morphology of different chemical treatments using 20% rice husk loading. Fig. 3(a) illustrates the small gap between the rice husk filler and the r-HDPE matrix detected the SEM image. This is due to higher loading of rice husk that caused poor and weak mixing between the filler and the matrix during the experiment and thus, led to inefficient bonding for the polymer composites when using 20% rice husk loading. The similar rice husk loading also resulted in relatively insufficient adhesion when applied to the r-HDPE matrix. From Fig. 3(b), the use of NaOH treatment can actually remove natural fats, waxes, lignin, and celluloses from the rice husk filler surface and resulted in the hydroxyl (OH) functional group. NaOH can react with the OH functional group in the cellulose to improve the roughness of surface.

Fig. 3(c) illustrates the untreated set by using 20% rice husk loading without any treatment. The large gap between the filler and the matrix increased as the rice husk filler is pulled out and thus resulted in low tensile strength of the polymer composites. This is because the filler loading is higher and thus caused the weak interfacial region between the polymer composites. Chen reported the increase of contact surface area with increasing rice husk loading and consequently resulted in poor interfacial adhesion between the matrix and the filler due to high weight fraction of rice husk filler applied in the polymer composites [19,21,22].

Fig. 3(d) illustrates the SEM image of 20% rice husk by maleate treatment using water. The greater gap between the rice

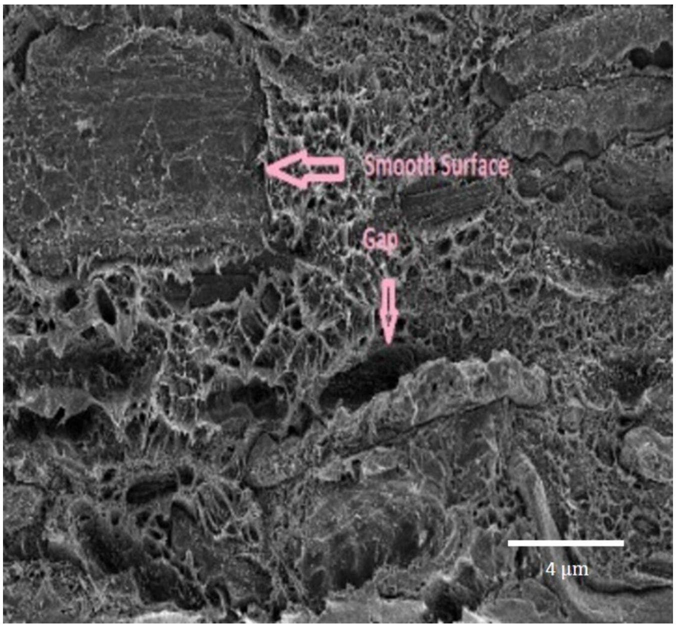

Fig. 3(a). 20% rice husk by acid treatment

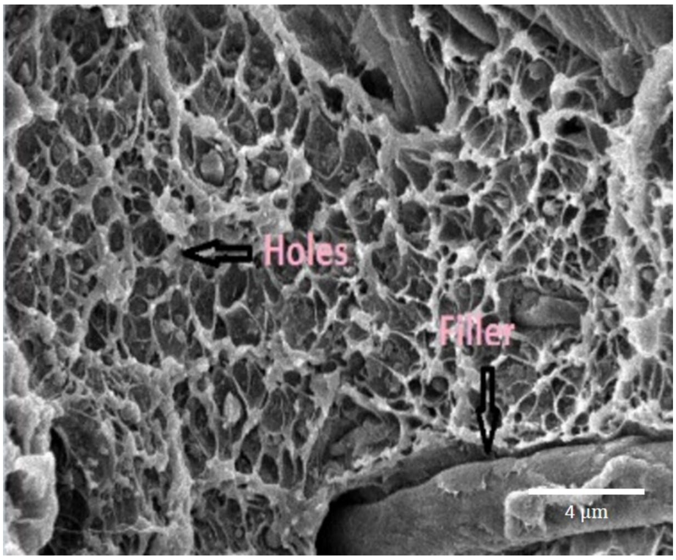

Fig. 3(b). 20% rice husk by alkali treatment

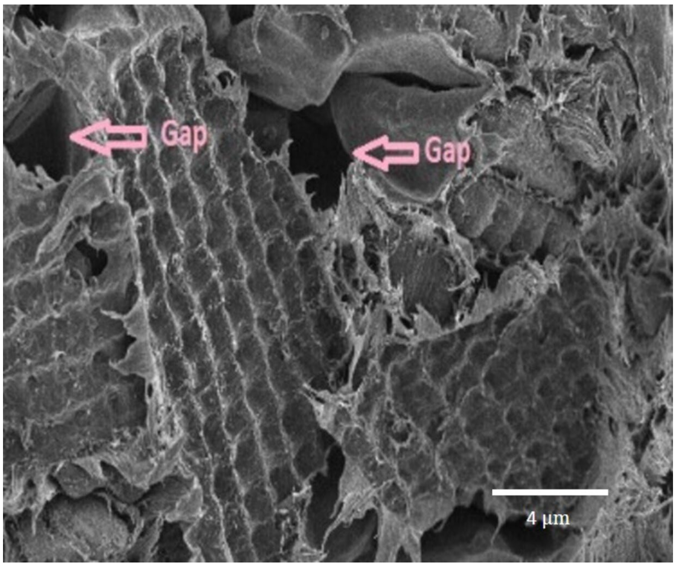

Fig. 3(c). 20% rice husk without treatment

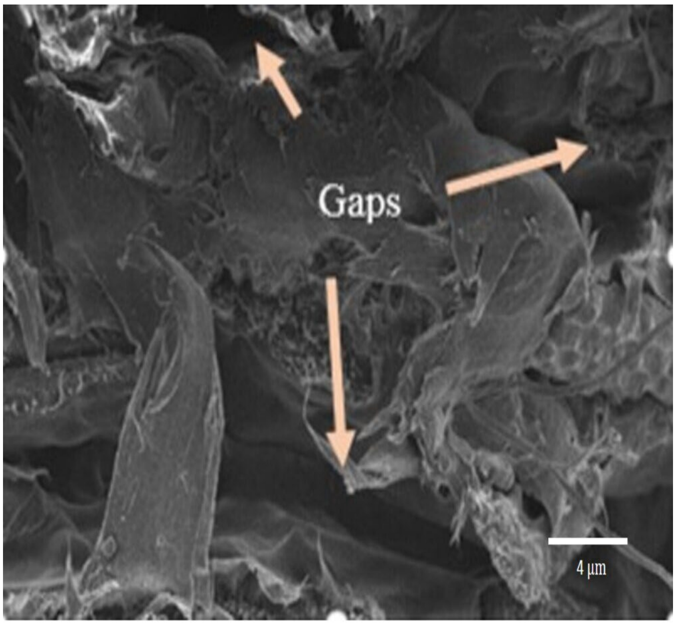

Fig. 3(d). 20% rice husk by maleate treatment husk filler and the r-HDPE matrix is due to the increasing loading of filler applied to polymer composites. This can cause the occurrence of weak and insufficient bonding due to the extent of irregular fracture initiation process and brittle deformation produced in the composites.

Among these treatments, M-10 and M-20 did not differ much due to the function of MAPE as a coupling agent that improves the strong bonding and adhesion between the filler and the matrix. MAPE can increase the high interaction between the rice husk and the r-HDPE matrix and produce composites with high tensile properties. The agent undergoes grafting reaction to react with the OH functional group in the rice husk's cellulose structure. This reaction can subsequently decrease hydrophilic tendency and the C-C bond of the maleic anhydride group is formed with the matrix. The covalent bond between the C-C bonds can create the chemical bridge between the filler and the matrix in order to provide efficient interlocking. Besides, the smooth surface of the sample using NaOH treatment has higher tensile properties than the acid-treated sample to achieve excellent mechanical interlocking of the fibre reinforced with the polymer matrix.

5.6. Flame test

The ignition time, quantity of smoke, smoke colour, and burning rate are presented in TABLE 5. Different loadings of rice husk (i.e., 10%, 20%, and 30%) were used to perform the flame test in order to determine the burning properties of the composites. The presence of MAPE can cause long ignition time, low burning rate, and also produce more black smoke.

TABLE 5

Flammability test

Sample	Time of Ignitions (s)	Quantity of Smoke	Colour of Smoke	Burning Rate (mm/min)
M-10	25.10	More smoke	Black	11.95
M-20	26.48	More smoke	Black	11.32
M-30	26.93	More smoke	Black	11.14
C-10	24.31	More smoke	Black	12.34
C-20	25.22	More smoke	Black	11.89
C-30	25.57	More smoke	Black	11.73

From the results, M-20 and M-30 produced more black smoke with ignition time of 22.48 and 21.58 s, respectively. For the untreated samples, C-10, C-20, and C-30 indicated weak mechanical properties compared to the samples subjected to maleate treatment, and thus contributed to the high burning rate and low flame resistance due to shorter burning time recorded during the flame test. The burning rates of C-10, C-20, and C-30 are 9,096, 10.59, and 10.83 mm/min, respectively.

All the samples released a huge amount of black smoke due to the lignin content embedded in rice husk. The presence of MAPE as the coupling agent in the bio-composites can improve the interfacial bonding and adhesion between the matrix and the filler, and this resulted in excellent flame resistance compared to C-10, C-20, and C-30. Higher rice husk loading will contribute to high flame resistance (i.e., low burning rate) of the composites because the silica content will largely decrease the combustion rate.

6. Conclusion

This study examined the mechanical characteristics of composites made of recycled high-density polyethylene (r-HDPE) reinforced with rice husks and the impact of chemical treatment on these characteristics. The experiment used various formulations of rice husk content in bio-composites made using a twin-screw extruder and a hot press (10%, 20%, and 30%).

Based on the findings and subsequent discussion, FTIR analysis was used to validate the presence of coupling agents such maleic anhydride grafted polyethylene (MAPE) in the bio-composites. Maleate bio-composites had greater tensile characteristics than untreated composites as a result of the ester linkages that formed between MAPE and the hydroxyl groups of the filler. The inclusion of MAPE increased interfacial bonding, particularly in the M-10 formulation, according to SEM analysis.

Although the tensile characteristics of the bio-composites dropped with increasing rice husk loading because the matrix did not sufficiently moisten the filler, this led to greater gaps in the polymer composites. The 10% rice husk content showed the best tensile strength and elongation at break, indicating efficient stress transfer. In contrast, the 10% rice husk component had the lowest modulus of elasticity, which can be attributed to the high stiffness of rice husk and the low stiffness of r-HDPE.

Additionally, the flammability test showed that rice husk-reinforced r-HDPE composites treated with maleate had better flame resistance because higher rice husk loading resulted in longer ignition periods.

Despite these results, there are certain study gaps that should be filled in by other investigations. In order to fully understand the implications on mechanical characteristics and flammability, the inquiry could benefit from examining a larger range of rice husk content. In order to determine if these bio-composites are appropriate for use in practical applications, additional study should concentrate on examining the long-term durability and environmental impact of these materials.

Additionally, it would be beneficial to look at the viability and scalability of producing these bio-composites on an industrial scale, taking things like cost-effectiveness and production efficiency into account.

In conclusion, this study provides important light on the advantages of maleic anhydride treatment as well as the mechanical characteristics of rice husk-reinforced r-HDPE composites. To develop a more thorough understanding of these bio-composites' potential and clear the way for their practical application in a range of eco-friendly applications, future studies should fill up the gaps that have been found.

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REFERENCES

[1] A. Samir, F.H. Ashour, A.A.A. Hakim, M. Bassyouni, Recent advances in biodegradable polymers for sustainable applications. npj Mater. Degrad. **6** (1), (2022).
DOI: https://doi.org/10.1038/s41529-022-00277-7

[2] R. Kumar, B. Rai, S. Gahlyan, G. Kumar, A comprehensive review on production, surface modification and characterization of nanocellulose derived from biomass and its commercial applications. Express Polym. Lett. **15** (2), 104-120 (2021).

[3] T. Gurunathan, S. Mohanty, S.K. Nayak, A review of the recent developments in biocomposites based on natural fibres and their application perspectives. Compos. Part A Appl. Sci. Manuf. **77**, 1-25, (2015).
DOI: https://doi.org/10.1016/j.compositesa.2015.06.007

[4] J.J. Andrew, H.N. Dhakal, Sustainable biobased composites for advanced applications: recent trends and future opportunities – A critical review. Compos. Part C Open Access. **7**, 100220 (2022).
DOI: https://doi.org/10.1016/j.jcomc.2021.100220

[5] A.A.R. Amer et al., Optimizing of the cementitious composite matrix by addition of steel wool fibers (chopped) based on physical and mechanical analysis. Materials (Basel). **14** (5), 1094 (2021).

[6] H.-S. Kim, H.-S. Yang, H.-J. Kim, H.-J. Park, Thermogravimetric analysis of rice husk flour filled thermoplastic polymer composites. J. Therm. Anal. Calorim. **76** (2), 395-404 (2004).

[7] F. Yao, Q. Wu, Y. Lei, Y. Xu, Rice straw fiber-reinforced high-density polyethylene composite: Effect of fiber type and loading. Ind. Crops Prod. **28** (1), 63-72 (2008).

[8] H.-S. Yang, H.-J. Kim, J. Son, H.-J. Park, B.-J. Lee, T.-S. Hwang, Rice-husk flour filled polypropylene composites; mechanical and morphological study. Compos. Struct. **63** (3-4), 305-312 (2004).

[9] R. Arjmandi, A. Hassan, K. Majeed, Z. Zakaria, Rice husk filled polymer composites. Int. J. Polym. Sci. **2015**, (2015).

[10] H.G.B. Premalal, H. Ismail, A. Baharin, Comparison of the mechanical properties of rice husk powder filled polypropylene composites with talc filled polypropylene composites. Polym. Test. **21** (7), 833-839 (2002).

[11] A. Nourbakhsh, F.F. Baghlani, A. Ashori, Nano-SiO2 filled rice husk/polypropylene composites: Physico-mechanical properties. Ind. Crops. Prod. **33** (1), 183-187 (2011).

[12] S.H. Ghaffar, O.A. Madyan, M. Fan, J. Corker, The influence of additives on the interfacial bonding mechanisms between natural fibre and biopolymer composites. Macromol. Res. **26**, 851-863 (2018).

[13] H.-S. Yang, H.-J. Kim, H.-J. Park, B.-J. Lee, T.-S. Hwang, Effect of compatibilizing agents on rice-husk flour reinforced polypropylene composites. Compos. Struct. **77** (1), 45-55 (2007).

[14] J.Y. Tong, N.R.R. Royan, Y.C. Ng, M.H. Ab Ghani, S. Ahmad, Study of the mechanical and morphology properties of recycled HDPE composite using rice husk filler. Adv. Mater. Sci. Eng. **2014**, (2014).

[15] D. Deb, J. M. Jafferson, Natural fibers reinforced FDM 3D printing filaments. Mater. Today Proc. **46**, 1308-1318 (2021).

[16] I. Elfaleh et al., A comprehensive review of natural fibers and their composites: An eco-friendly alternative to conventional materials. Results Eng. 101271 (2023).

[17] M.N. Salleh et al., Comparison between the tensile, water absorption and Flammability properties of recycled high-density polyethylene/rice husk Composite From twin-screw extruder and heated two-roll mill. Arch. Metall. Mater. **67**, 2, 661-668 (2022).
DOI: https://doi.org/10.24425/amm.2022.137803

[18] S. Hamdan, A. S. Ahmed, Effect of chemical treatment on rice husk (rh) reinforced polyethylene (pe) composites. BioResources. **5**, 854-869 (2010).

[19] R.S. Chen, S. Ahmad, S. Gan, Characterization of rice husk-incorporated recycled thermoplastic blend composites. BioResources **11 (4)**, 8470-8482 (2016).

[20] M.H. Ab Ghani, N.R.R. Royan, S.W. Kang, A.B. Sulong, S. Ahmad, Effect of alkaline treated rice husk on the mechanical and morphological properties of recycled HDPE/RH Composite. J. Appl. Sci. Agric. **10** (5), 138-144 (2015).

[21] N.R. Rajendran Royan, A.B. Sulong, N.Y. Yuhana, R.S. Chen, M.H. Ab Ghani, S. Ahmad, UV/O3 treatment as a surface modification of rice husk towards preparation of novel biocomposites. PLoS One. **13** (5), e0197345 (2018).

[22] R.S. Chen, M.H. Ab Ghani, S. Ahmad, M.N. Salleh, M.A. Tarawneh, Rice husk flour biocomposites based on recycled high-density polyethylene/polyethylene terephthalate blend: Effect of high filler loading on physical, mechanical and thermal properties. J. Compos. Mater. **49** (10), (2015).
DOI: https://doi.org/10.1177/0021998314533361