Mechanical Properties of Rice Husk-Recycled Polypropylene Composite

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ABSTRACT

The growing amount of plastic and food waste has become a serious problem around the worldwide. As a byproduct of milling rice, rice husk is an agricultural waste that is produced in bulk quantities. Rice husk has been used as filler in polymer composites in a variety of ways. However, there have only been a few reports of using rice husk as reinforcement in composites made of recycled polypropylene. The limited interfacial reaction between natural fibre and polymer is the fundamental issue in natural fibre-based composites. This prompted the development of sustainable composite manufactured from recycled polypropylene(rPP) and rice husk (RH). Thus, we investigated the intermolecular adhesion between rPP–RH composite with maleic anhydride grafted polypropylene (MAPP) as a coupling agent. Varied compositions of RH in the range of 10–40 wt% with 4 wt% of MAPP were fabricated. The rPP, RH and MAPP were blended and executed in injection moulding. The mechanical properties will be analysed through differential scanning calorimetry, rheology, tensile, flexural, impact and hardness tests. The result shows RH agglomeration and limited dispersion in the composite cause a reduction of up to 40% for tensile strength compared to neat rPP. Despite this, it demonstrates improvements in tensile modulus, flexural modulus, impact, and hardness. It is evidence of a good intermolecular between rPP matrix and RH. In conclusion, the ideal RH loading for composites occurs at a filler

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content of 40% and has acceptable mechanical properties for various composite applications.

Keywords: *Recycled Polypropylene; Rice Husk; Rice Husk Reinforced Recycled Polypropylene Composite; Polymer Composite; Mechanical Properties*

Introduction

Water pollution, air pollution, and climate change all negatively affect people and the environment. Scheduled wastes recorded in 2019 exceeded 4.0 million tonnes, according to Malaysia's Department of Statistics [1]. The power plants, metal refineries, chemical industries, and electrical and electronics industries each contributed 57.1% (2.3 million tonnes) of the total scheduled waste [2]. Although this is not a new concern, it remains the world's most significant issue and one of the leading causes of diseases and mortality in people. Environmental pollution impacts the lives of animals and the ecological system's balance. The increase of plastic waste in landfills and the ocean is becoming the most common threat that has been discovered. It harms the environment because plastics will take decades to decompose. It becomes hazardous to humans, animals, and marine life. Therefore, this issue has a bad impact on the food chain, resulting in poisonous food being consumed by humans such as causing digestive issues.

Waste management is critical for the long-term sustainability of the environment. Several programs have been implemented to prevent this problem such as beach clean-up, which intercepts plastic before it reaches the ocean. Recently, there have been new initiatives in separating, cleaning and recycling waste contained in recyclable materials. The most popular program was Coca-Cola, the world’s first bottle made from ocean plastic waste that was recovered from the Mediterranean Sea [3]. Thus, the idea of developing composite with the combination of recycled plastics with agricultural waste has big potential for a sustainable environment. Common processing methods that have been used for natural fibre-reinforced thermoplastic composites are compression moulding, injection moulding, and extrusion [3]-[10].

Rice is the most important crop and produces rice husk as a by-product. The annual output of rice husks is large but most are burned by the peasants. The main components of the rice are identical to those found in wood, including cellulose, hemicellulose, lignin, and silica [4]. Therefore, rice husk may be a potential substitute for alternative wood in wood-plastic composite because of the high amount of cellulose in the rice husk. Meanwhile, the exterior surface of the rice husk has a plethora of organised conical protrusions. It is a combination of lignin and silica that generates a natural protective coating in the form of silicon fibre membranes.

The challenging process of polymerization with fibres is its incompatibility with non-polar matrices. The RH (filler) particles tend to aggregate when combined with a non–polar polymer rPP (matrix), which limits the stress transmission from matrix to filler [11]. Weak interfacial adhesion due to fibre-matrix incompatibility reduces composite impact strength, tensile strength and tensile elongation at break [6]. The mechanical properties of biocomposites are dependent on strong compatibility between the dispersed phase filler and the continuous phase matrix. Modifying the surface of the RH fibre is necessary to increase RH compatibility with non-polar polymers. The compatibility of RH fibre–polymer composite is achieved by using surface treatment processes such as mercerization, ozonolysis, plasma, electron beam irradiation, and coupling agents [12]. Coupling agents have gained significant attention in the production of biocomposite materials due to their ability to improve matrix interfacial adhesion in natural fibre-reinforced polymer composite systems. Simultaneously, the chemical treatment is also preferable to use because its uses simple technology, inexpensive and effective in the modification of the interface between natural fibre and the polymeric matrix. Common chemicals used in the chemical treatment are silane, sodium hydroxide (NaOH), methylene diphenyl diisocyanate (MDI), and MAPP [4]-[5], [13].

Recently, MAPP has been used in producing excellent filler-matrix bridge effects. It was also discovered that rice husk's O–H group and MAPP's maleic anhydride group can esterify each other [13]-[14]. The bi-functional chemical structures of interface MAPP can consume hydrophilic groups (such as O–H groups) on the fibre surface and introduce hydrophobic groups (such as C–H groups in PP) to the fibre surface which results in the higher interface compatibility between hydrophilic fibre and hydrophobic PP, thus make them stronger interactions at the fibre/rPP composites interface [14]. This reduced the moisture absorption of the composite and improved the tensile and flexural strengths [15]. Furthermore, the lower impact properties of the fibre/polymer matrix composite are beyond the point of the improved interaction of the fibre/polymer matrix interface rPP.

This research intends to explore the utilization of MAPP as a coupling agent to improve the surface interaction between RH fibre and rPP matrix composite, and to improve the mechanical, rheological, and thermal properties. This research is focused on various weight percentages of rPP:RH ratios, which are 100:0, 90:10, 80:20, 70:30, and 60:40. This research will promote the circular economy, natural fibre-based composite as an alternative automotive parts, and gain environmental benefits. Simultaneously, this research also adheres well to the sustainable development goal (SDG).

Methodology

The experimentation process for this project starts with the collecting, cleaning, drying, and crushing of material for the feedstock. Following that, the composite was mixed to evaluate five different composition ratios with reinforcement fibre. Rheology and DSC analyses were carried out to comprehend the behaviour of the material before moving on to the injection moulding process. The sample was prepared via injection moulding to analyse the mechanical measurement. Tensile, flexural, impact and hardness tests will be conducted to study the mechanical properties. Thus, the findings of all these tests are revealed and discussed.

Feedstock preparation

Recycled polypropylene with a density of 0.893 g/cm^3 [16] is used to calculate the composition weight of the composite. The recycled polypropylene was collected from a recycling centre in Subang Bestari, Shah Alam, and granulated into pellets before being used as a matrix. The rice husk fibre was supplied by Persatuan Peladang Kebangsaaan (KPK) in Sungai Besar, Selangor, as a reinforcing fibre. Rice husks were blended until they were in the size range of 20−100 mm, then dried in the oven for 3 hours at 105 °C to remove any moisture content [16]. MAPP was purchased from Sigma Aldrich Sdn. Bhd. MAPP weight of 4% remained consistent across the research [13]. Table 1 shows the weight composition ratio of RH, rPP, and MAPP as coupling agents that had been used in the composites.

Table 1: The weight of rice husk (RH), recycled polypropylene (rPP), and MAPP for each composition of composite

Composition ratio	Density (g/cm^3)	Weight (g)		MAPP (g)
		rPP	RH	
rPP/10RH	0.834	345.6	40	14.4
rPP/20RH	0.782	307.2	80	12.8
rPP/30RH	0.737	268.8	120	11.2
rPP/40RH	0.696	230.4	160	9.6

Mixing process

The VT Sigma Blade mixer machine was used to mix the materials at a temperature range of 170−190 °C. The materials were slowly added to the mixing chamber for 20 min, then the mixing was continued for 40 min at a rotation rate range of 30−50 rpm. Then, the mixed composite was taken out from the mixer and allowed to cool to room temperature before being fed into a granulator.

Differential scanning calorimeter (DSC) analysis

Two samples for neat rPP and rPP/40RH composite were examined using the DSC Melter Toledo machine. The material was scanned from 0 to 500 °C at a heating rate of 10 °C/min with a weight of 9.6 mg and under a nitrogen atmosphere. This DSC analysis data is useful in evaluating the permissible temperature range to be considered in the injection moulding process.

Rheology measurement

The rheological characteristics of rPP and rPP/40RH, such as shear stress, shear viscosity, and the flow behaviour index, *n*, are determined using a capillary rheometer machine (model Capillary Bohlin). The feedstock flowability data were created using the Rosand Capillary Rheometer FlowMaster software. The shear rate of the machine was fixed at a range of 10 to 10000 s^{-1}, pressure at 0.1 MPa, diameter capillary die was 1 mm and length-to-diameter (L/D) of 10 was used for this test. This analysis determines the three temperatures with different variances of 165, 175 and 185 °C in order to find the optimal temperature for injection moulding.

Injection moulding process

The injection moulding process was conducted using a compact injection moulding machine (model Boy 22A). The temperatures for the feed zone, trans zone, metering, and nozzle zones are adjusted to 170, 180, 190 and 200 °C, respectively, based on optimum temperatures obtained from the rheology measurement. The injection moulding process parameters that had been used are shown in Table 2. The mould was designed according to the standard sample size for the tensile and flexural tests following ASTM 412-D and ASTM D790 standards, respectively.

Table 2: Injection moulding process parameters

Sample	Volume (mm^3)	Injection pressure (bar)	Holding time (min)
ASTM 412-D	17-19	60	3
ASTM D790	23	65	3

Mechanical measurement

The mechanical properties of fibre-reinforced polymers are determined using tensile, flexural, impact, and hardness tests. A Shimadzu Universal Testing Machine (UTM) (model AG-IC) equipped with a 50 kN load cell was used in this research to perform tensile and flexural tests according to ASTM D412-D and ASTM D790 standards. The crosshead speed was 5 mm/min. The flexural test was span at 50 mm and a crosshead speed of 5 mm/min. Meanwhile, the impact test was performed using a Notched Izod Impact Test Machine following an ASTM D256 standard. The type of weight R1 (2.71) with 0.453

kg was used to perform this test. Next, the sample size of 125×125×2 mm^3 was prepared for the hardness test using the Rockwell Hardness Tester (model INSTRON A654R). Meanwhile, the sample for the impact and hardness tests was modified from a flexural sample produced from the injection moulding process. For each test, five samples were tested for each composition.

Results and Discussion

Differential scanning calorimeter (DSC) analysis

Thermal phase transitions of rice husk (RH) fibre, neat recycled polypropylene (rPP) and rPP/40RH were studied using differential scanning calorimeters (DSC). Figure 1 illustrates the endothermic reactions of rPP and rPP/40RH. The result shows that rPP had a well-defined melt transition at 164.26 °C and the phase transformation occurred between 130−174 °C. The rPP/40RH composite melting behaviour was very similar to that of neat rPP, which melted at 165 °C and underwent phase transformations between 135 and 177 °C. If the temperature increases more, composite materials may degrade. At temperatures between 318−471 °C and 353−495 °C, respectively, the breakdown of rPP to rPP/40RH started to end. However, compared to neat rPP, modified composites showed greater degradation temperatures (both onset and endset). Because filler was added to the composites, there were mechanical interlocking and nucleating effects [17]. The outcome also showed that the decomposition peak for rPP and rPP/40RH happened around 440 and 454.2 °C, respectively. It has the same results as those reported in the previous works, where an endothermic peak at 448 °C may be related to the depolymerization of polypropylene with the production of propylene [18]. The data also showed that for rPP, there was another peak at 226.24 °C, which could be caused by impurities in the recycled material collection. As a result, the information acquired was utilised to choose the ideal temperature, which was above 165 °C when the material started to melt, and the data were used to determine the temperature for rheological study.

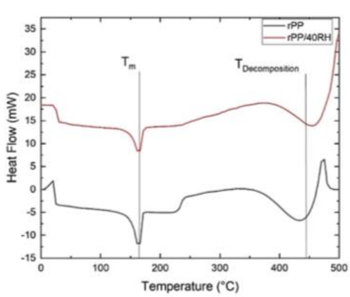


Figure 1: DSC analysis of neat rPP and rPP/40RH composite

Rheological properties

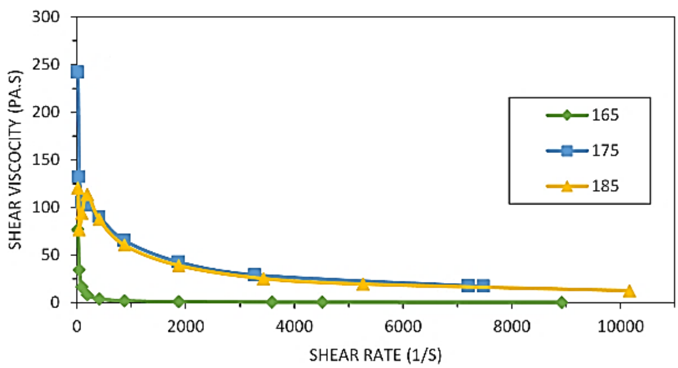
The melt viscosity of recycled polymer blends was evaluated to learn more about the interaction of the polymers in the blend and how compatibilizers affect it. This study was conducted to find the feedstock's stability during the injection moulding process [19]. A feedstock's flow behaviour is preferably pseudoplastic, which means that as the shear rate increases, the viscosity drops. The value of *n* for a pseudoplastic flow must be less than 1. The lower the value of viscosity, the better the flowability of the feedstock. Figures 2a and 2b illustrate the relationship between shear viscosity and shear rate. The data indicated that at 165 °C, the neat rPP and rPP/40RH had the lowest shear viscosity compared to the value of shear rate at 175 and 185 °C. According to recent research, the shear viscosity increased as the temperature value decreased [20]. In this case, the temperature of 165 °C for rPP and rPP/40RH may have been triggered by physical or equipment errors that affect the results of shear rate and shear viscosity. Shear viscosity comparisons between rPP and rPP/40RH indicated that the composite required higher temperatures than rPP, and this may cause the amount of fibre in the composite. The graph showed that the data series for viscosity and shear rate ranged from 0 to 500 Pa.s and 0 to 10000 s^{-1}, respectively. The results indicated that both composite materials had compressive strengths of less than 1000 Pa.s, considering that the materials were acceptable for injection moulding [21].

In this study, the shear sensitivity index *(n)* characteristics can also be evaluated. Previous studies have demonstrated that the optimal *n* value in the pseudoplastic flow phase is in the range of 0.5 to 0.7 [19]. The explanation is because the feedstock viscosity changes with the shear rate more quickly with a smaller *n* value, indicating how a high *n* value may lead to process instability and make it harder to control the sample quality. Table 3 presents the *n* value obtained from the slope of Figures 2a and 2b. The result showed that the lowest *n* value was obtained at 165 °C, while the value *n* in the range of 0.5−0.7 was obtained at 175 °C and 185 °C. Both compositions showed pseudoplastic behaviour at all temperatures. This suggests that as the shearing force increases, molecules that are typically out of alignment start to align their axes in the direction of flow [20]. With less internal resistance, the materials can shear more quickly under each succeeding shearing stress based on this orientation. As a result, the ideal temperature to set during injection moulding is above 175 °C. This information is necessary to keep in the injection moulding stage because it involves the flow of molten feedstock into the mould cavity. Pseudoplastic flow behaviour is the preferred one when using injection moulding [17]. The equation is a Power Law equation that may be used to determine this rheological behaviour (1) where ŋ, K, γ and n are viscosity, coefficient, shear rate and shear sensitivity index, respectively.

$$ŋ = K\gamma^{n-1} \tag{1}$$

Table 3: Flow behaviour index, *n*

Feedstock	Temperature (°C)	Flow Behaviour Index, *n*
rPP	165	0.01
	175	0.6
	185	0.7
rPP/40RH	165	0.01
	175	0.5
	185	0.5



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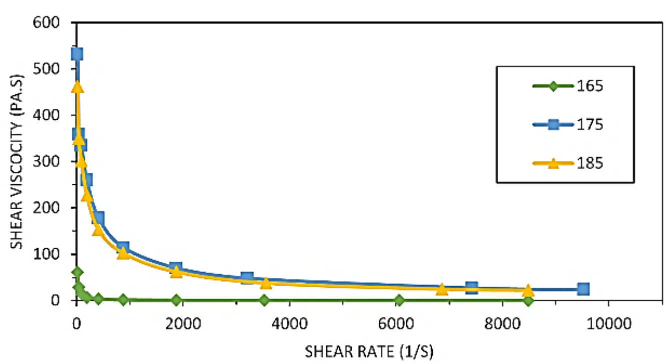


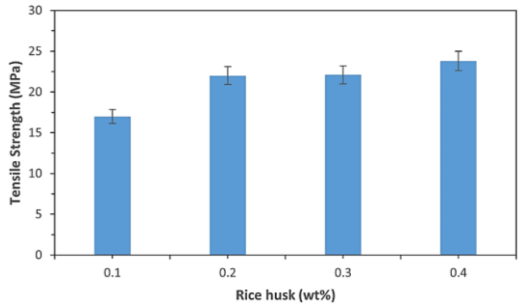
(b)

Figure 2: a) Correlation between shear viscosity and a shear rate of rPP, and b) correlation between shear viscosity and a shear rate of rPP/40RH

Mechanical properties

Tensile properties

A tensile test was conducted on the injection moulded rPP/RH composite to determine its mechanical properties. The tensile tests studied the significant properties of tensile strength, tensile modulus and tensile elongation. The tensile strength, tensile modulus, and tensile elongation of neat rPP and rPP reinforced with varying amounts of fibre loading are shown in Figure 3. It can be seen that the addition of fibre and MAPP to the rPP matrix reduced the tensile strength of the material by 22 to 44% compared to the neat rPP. The same result was also recorded, where the tensile strength of neat polypropylene and recycled polypropylene matrix declined when the rice husk filler percentage exceeded 5% [16]. Due to the small size of the filler particles, there was a large interfacial surface between the polar filler and the apolar matrix [22]. The lack of chemical bonding between the rPP and RH, as well as poor RH dispersion over the rPP matrix due to the substantial differences in surface energy of the rPP and RH fillers, was the potential reason for the reduction of tensile strength when compared to neat rPP. Furthermore, insufficient dispersion caused filler agglomeration, as well as a reduction in tensile characteristics, which may be due to low homogeneity during the mixing process that contributes to this reduction in the MAPP bridge effect between the filler and matrix. Agglomerates' presence can lead to flaws and extra voids between the and the matrix polymer, which lowers the tensile strength. However, according to a previous report, the hydroxyl groups in RH reacted with the anhydride groups in MAPP through an esterification process, which may help to increase the interfacial adhesion between the filler and matrix [13]. The tensile strength increased gradually to the maximum value as the amount of RH fibre was increased to the maximum fibre loading of 40%, as shown by the results in Figure 3a. The results indicated an excellent bridge effect between the filler and matrix, fibre dispersion and fibre−matrix adhesion. Thus, it improved the stress transfer efficiency of the rPP/RH composite. However, the tensile modulus of the rPP−RH composite improved by 33 to 46% compared to neat rPP. The increase in tensile modulus with an addition of RH in the loading of rPP matrix was due to the RH's ability to impart more stiffness to the matrix [23]. This is a typical behaviour when the hard filler is added to softer polymer matrices. Natural fibres have a higher elastic modulus than pure rPP, resulting in increased composite stiffness [12]. Due to this, adding these fillers tends to significantly improve the stiffness of its composites. Figure 3b illustrates the tensile elongation of the composite, which indicates its flexibility of the composite. The results demonstrated a decrease in tensile elongation with the increase in fibre percentage. It can be seen that fibre reinforcement composite with MAPP had a lesser tensile elongation than neat rPP. This occurred as a result of the coupling agent function in igniting the interaction between the filler surfaces and rPP that caused the composite to stiffen. The coupling agent's impact on this property was not immediately apparent in this study. These results also indicated that MAPP decreases the flexibility and elasticity of materials, which is consistent with earlier research [24].



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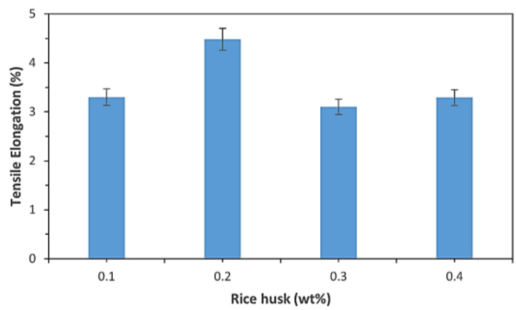

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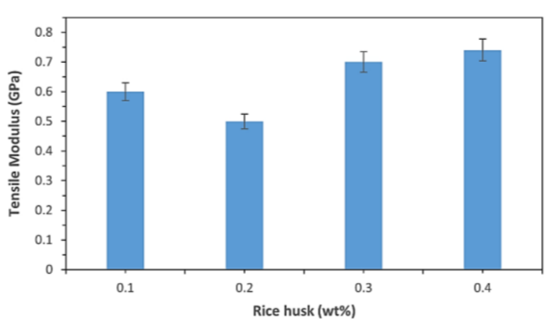


(c)

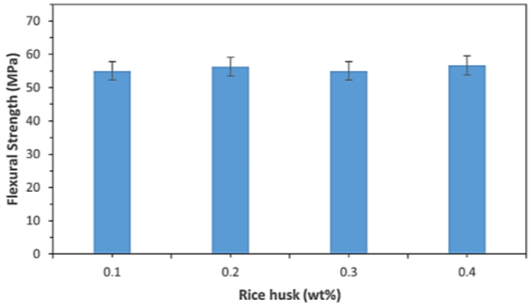
Figure 3: Tensile properties of rPP/RH composite with different percentage of rice husk fibre; a) tensile strength, b) tensile elongation at break, and c) tensile modulus

Flexural properties

The flexural analysis found that the matrix fillers have excellent interfacial bonding in the composite. Figure 4 shows the flexural strength and modulus of rice husk-reinforced polypropylene composites. In comparison to neat rPP, the data showed that 40 wt% fibre has the maximum flexural strength and flexural modulus. This could be attributable to the introduction of filler and coupling agent, which increases due to an improvement in the filler and matrix boundary region. This is further proven by the fact that flexural strength improves when MAPP is added to neat rPP [15]. The same outcome was observed in the earlier investigation, where adding more fibre and MAPP as a coupling agent has been shown to improve flexural modulus by 73% from neat PP [7]. Interestingly, the modulus of composites with coupling agents at high filler loading was much better than that of the composite without coupling agents. The rising pattern could be brought on by the fact that rice husk is much stiffer than rPP. A crystalline layer can develop around the fibres when coupling agents are present because surface crystallisation takes precedence over bulk crystallisation [15]. Crystallite can enhance the polymer matrix's contribution to the composite modulus since they have a greater modulus than amorphous areas.

Impact

Rapid crack propagation via the material causes impact failures [25]. The rate of fracture growth is inversely proportional to the material's impact resistance. To be deemed as impact resistant, a polymer must be able to absorb the majority of the impact energy and limit the rate of crack development. The Izod impact test was used to determine the amount of impact energy required to crack or fail the surface. Figure 5 demonstrates that rPP and rPP reinforced with fibre had similar results at 20, 30, and 40%; however, 10 wt% fibre resulted in low absorbance as compared with neat PP. According to the previous study, the addition of rice husks resulted in a reduction of impact strength because the husks reduced energy absorption ability which led to the increase of fibre breakage and more residual stress in the composite [13]. The addition of MAPP to the fibre-matrix composite structure made it more brittle, and the husks limited the free motion of matrix chains. The matrix dampened the force during loading since there was greater matrix movement between the husks. However, adding rice husks led to a similar impact strength of 1.8 kJ/mm^2. This finding suggests that the MAPP may have improved the interaction between rice husk fibre and matrix.



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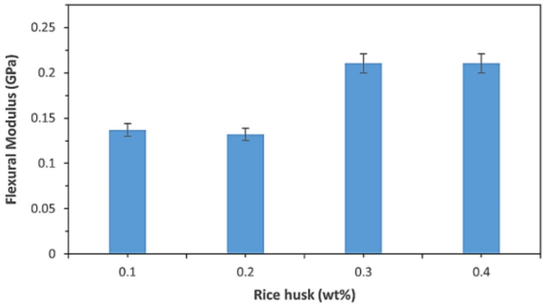

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Figure 4: Flexural properties of rPP/RH composite with different percentages of rice husk fibre; a) flexural strength, and b) flexural modulus

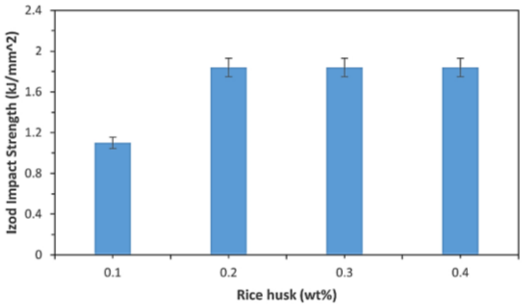

Figure 5: Impact of rPP/RH composite with different percentages of rice husk fibre

Hardness

Figure 6 illustrates the hardness strength with neat rPP and different fibre loading. The optimum hardness was obtained at 40% fibre load. In comparison to neat rPP, the composite can improve by up to 16%. This improvement in the composite can be attributed to the high interfacial adhesion of the rPP−RH and MAPP composite, as well as the RH material's molecular-level dispersion in the matrix, which results in an improvement in how effectively stress is transferred from the matrix to the filler phase [23]. Additionally, when a material becomes more resistant to deformation, its hardness improves. This occurs when additional filler is introduced, making the composite harder and the substance tougher [26]. Better resistance to plastic deformation in the filler's transverse direction is provided by the filler layer.

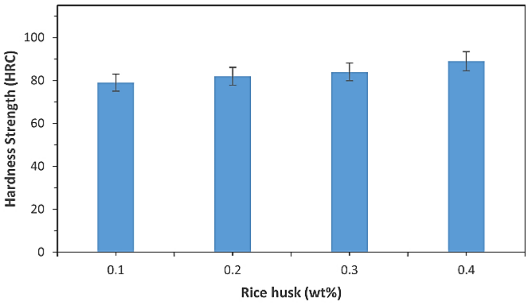

Figure 6: Hardness of rPP/RH composite with different percentage of rice husk fibre

Conclusion

Different rice husk weight ratios have been successfully made and used to characterise the effects of rice husk (RH) as a reinforcement filler on the properties of recycled polypropylene (rPP) matrix with maleic anhydride grafted polypropylene (MAPP) as coupling agents. The determination of the rheology temperature range is set at 165, 175 and 185 °C. Thus, the data indicate that the feedstock has the best flow mouldability at temperatures between 175 and 185 °C, where the *n* value obtained is in the range of 0.5−0.7, which exhibits pseudoplastic behaviour. Therefore, the result demonstrates a decrease in tensile strength and tensile elongation in the composite. However, the finding indicates that tensile modulus, flexural strength, flexural modulus, impact strength, and hardness strength have all improved. In this experiment, the tensile strength and tensile elongation show a decline of 22 to 44% and 33 to 46%, respectively, compared to neat rPP, which shows that the composite lacks chemical bonding and dispersion. According to the flexural study, the best outcome occurs at a fibre loading of 40% weight when a good polymer matrix contributes to the matrix. In addition, when the ratio of RH reinforcement is added to the rPP−RH matrix, the hardness data show an improvement of 3−16% in contact that absorbs high energy impact. Therefore, a composite that incorporates 40% rice husk fibre and MAPP demonstrates greater results among the composites. In conclusion, rice husks and maleic anhydride polypropylene have successfully proven to have better properties of polymer-based nanocomposites and other composite applications as coupling agents.

Contributions of Authors

The authors confirm the equal contribution in each part of this work. All authors reviewed and approved the final version of this work.

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Conflict of Interests

One of the authors, Freddawati Rashiddy Wong, is an assistant managing editor of the Journal of Mechanical Engineering (JMechE). The author has no other conflict of interest to note.

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