Electrical Resistivity, Thermal Stability and Tensile Strength of Rice Husk Flour-Plastic Waste Composites

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ABSTRACT

In this study, the rice husk flour-plastic waste composites (RPC) was produced from polypropylene (PP) and high density polyethylene (HDPE) wastes with 30 and 50% rice husk flour (RHF) contents. RPC was made by melt compounding and compression moulding processes. The electrical resistivity, thermal stability and tensile strength of RPC were determined. The RPC was tested in electrical resistivity and tensile strength according to the ASTM D-257 and ASTM D-638 respectively, while thermal stability was tested using thermogravimetric analysis (TGA) method. From the results, high content of RHF reduces all properties, except for tensile modulus of elasticity (TMOE) in tensile strength test. The ability of moisture absorption and the presence of hemicelluloses, cellulose and silica in RHF reduce the electrical resistivity and thermal stability behaviour of RPC from 50% RHF. The good binding elements and filler agglomeration in RPC from 50% RHF improve only TMOE. Insufficient stress transfer and rigid interphase occurred between RHF and plastics during tensile maximum load and elongation at break (E_b) in tensile strength test. In general, RPC from HDPE indicates better thermal stability, tensile modulus of rupture and E_b (in tensile strength test) compared to PP, based on the good behaviour of thermal conductivity, low water absorption, high molecular weight and good elongation properties of HDPE. However, RPC from PP shows good electrical resistance due to the low thermal expansion coefficient of PP.

Keywords: *electrical resistivity, thermal stability, tensile strength, polypropylene wastes, polyethylene wastes, rice husk flour*

Introduction

Agriculture sector contributes abundant residues such as rice husk. These residue materials can be converted into a new product through the combination with non bio-material such as polymer wastes or recycled polymer. Bledzki and Gassan [1] stated the potential utilization of agricultural wastes as new and inexpensive materials to replace synthetic filler in a composite product. The possible use of rice husk dust as natural filler in a composite product may help to reduce the burden of agricultural wastes and manufacturing cost. According to Hoest [2], rice husk are known to have low lignin (160 g kg^{-1}) and high silica (SiO_2 = 230 g kg^{-1}) contents which

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influence the composite material the ceramic like properties. Mutlu [3] in his study stated that rice husk dust and rice straw dust can be effectively used in the manufacturing of brake pad. The silica particles of rice husk were homogenously distributed in the body of the brake pad composite and at the same time increased the frictional performance of composite brake pad [3].

Apart from agriculture residues, polymer wastes from daily life may also create the environmental problems. These wastes usually disposed by land fill or re-used as alternative containers in the household application. Plastic is one of the non-biodegradable materials that can absolutely increase the environmental pollution when burned in an open area. In addition, recycled polymers have several advantages such as easy to obtain, low cost, good mechanical properties and excellent chemical resistance. Due to the superior properties of these materials, they have been used both with natural fillers and in manufactured composite forms [4]. Recently, the use of natural flour as reinforcing filler with thermoplastic polymer as matrix component has been rapidly increased in automotive and electrical application and in the field of construction. For the purpose of these composite systems, electrical resistivity, thermal stability and tensile strength are among the most important factors that have to be determined.

Based on this rational, the potential composite product from a combination of rice husk flour and polymer waste materials was investigated. The objective of this study was to determine the electrical resistivity, thermal stability and tensile strength of rice husk flour-plastic waste composite (RPC). The RPC was fabricated using different percentages of rice husk flour (30 and 50%) and different types of plastic waste material (polypropylene and high density polyethylene).

Material and Method

The manufacturing process of RPC was conducted at Bio-Composite Technology Laboratory and Polymer Technology Laboratory, Faculty of Applied Sciences, UiTM, Shah Alam, Malaysia. The rice husk was obtained from local rice miller in Sungai Besar, Malaysia. The rice husk was ground to obtain small particle size. The rice husk particle was screened by screening machine to produce rice husk flour (RHF). The size of RHF was ranged from 80 to 100 meshes. The RHF was dried to 4% moisture content for several days in an oven dryer.

Polypropylene (PP) was obtained from plastic container wastes and plastic components that have recycling number "5", such as food container and vehicle components. While high density polyethylene (HDPE) was obtained from plastic container wastes and any plastic components that have recycling number "2", such as medicine bottles, liquid detergent bottles or similar products. The PP and HDPE plastic wastes have to be cleaned first by removing all contaminants, sticker, or any other label. Then the plastic wastes were cut randomly into small size approximately 2 × 2 cm in width and length, respectively. The plastic pieces were crushed by hammer mill to obtain the small size of plastic particles, randomly from 0.5 to 1.0 mm in length.

The RHF was mixed separately with PP and HDPE in a dispersion mixing machine. The polymer materials were molten first in dispersion mixing, followed by mixing process with RHF to produce composite. This composite was prepared with two different percentages of RHF

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content (30 and 50% of overall mass). The compounding of RHF into the polymer materials was accomplished using a dispersion mixer D1-5 with a capacity of 2 kg for each batch. The blending temperature was maintained at 200^{o}C for PP and 180^{o}C for HDPE. After the completion of mixing process, the mixture was rolled on a plate to obtain thin layer of composite. The composite was crushed using a crusher machine to obtain pellet. The pellet was moulded using standard mould with the dimension of 150 × 150 mm in length and width respectively, and 3 mm in thickness. Then the composite was pressed for 5 minutes using a hot press machine under 180 - 200 ^{o}C temperature and 60 bar in pressure to produce RPC. Figure 1 shows the typical RPC produced in this study. The RPC was cooled, trimmed and cut into the required dimension for electrical resistivity, thermal stability and tensile strength test samples. All samples were conditioned at 23±2^{o}C temperature and 50±5% relative humidity in a conditioning chamber for about 40 hours (according to the ASTM D 618-08 [5]) prior to testing.

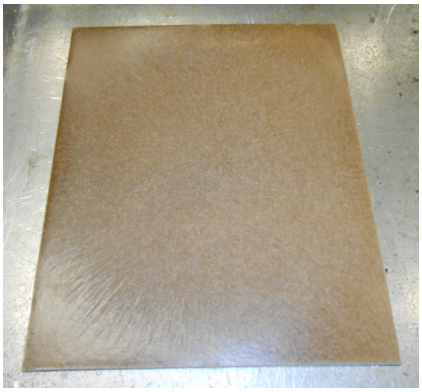

Figure 1**:** Typical RPC produced in this study.

Figure 2 shows the set-up of electrical resistivity, thermal stability and tensile strength tests in this study. Electrical resistivity test was conducted at Electrical Engineering Laboratory, Faculty of Electrical Engineering, UiTM, Shah Alam. Keithley electrometer (Model 6517A, Keithley Instruments) equipped with a concentric ring electrodes test fixture (with applied voltage of 500V for 60 seconds) was used to measure the electrical resistivity of the samples in accordance to the ASTM D-257 [6]. Samples with 48 mm in diameter and 3 mm in thickness were cut into circle profile using a jig saw. The instrument automatically performed the calculation (Equation 1) and displayed the electrical resistivity reading. Average values of 10 samples were recorded for each group.

$$\rho = PR/g \qquad \text{... Equation 1}$$

where:
ρ = surface resistivity (per square)
P = the effective perimeter of the guarded electrode (mm)
R = measured resistance in ohms (V/I)
g = distance between the guarded electrode and the ring electrode (mm)

Thermogravimetric analysis (TGA) was performed using a Perkin Elmer TGA-7 analyzer at Polymer Technology Laboratory, Faculty of Applied Sciences, UiTM, Shah Alam. TGA

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measurement was carried out using 10 – 13 mg of the RPC powder from 30 to 600^{o}C at a heating rate of 40°C min^{-1} in a N_2 atmosphere. The TGA was conducted with the compounds placed in a high quality nitrogen (99.5% nitrogen and 0.5% oxygen contents) atmosphere with a flow rate of 20 mL min^{-1}, in order to avoid unwanted oxidation. The samples were heated to a maximum temperature of 600^{o}C until it turned into ash. Then the graphs indicating weight loss percentage versus temperature were generated by TGA data acquisition system. From the graph, the decomposition temperature was measured using Perkin Elmer Pyris software. Average values of 3 readings were recorded for each group. The weight loss for final temperature was also measured using Equation 2:

$$W = (m_0 – m/m_0) \times 100 \quad \text{... Equation 2}$$

where:
W = percentage of weight loss (%)
m_0 = initial sample weight
m = actual weight loss

Tensile strength such as tensile modulus of rupture (TMOR), tensile modulus of elasticity (TMOE) and elongation at break (E_b) were determined according to ASTM D-638 [7] using the Instron universal testing machine model 5582 fitted with 100 N load cell. Test was performed at room temperature with sample dimension of 127 mm × 12.7 mm in length and width, and 3 mm in thickness. The samples were held in small grips while testing, at a crosshead speed of 5 mm/min until fracture. Average values of 10 samples were recorded for each group. Stress was calculated based on the measured load, while strain was calculated based on the measured extension. TMOR, TMOE and E_b were determined using Equation 3 to 5, respectively:

$$TMOR = P_{max}/A_0 \quad \text{... Equation 3}$$

where:
TMOR = tensile modulus of rupture (N/mm^2)
P_{max} = maximum load (N)
A_0 = cross sectional area (mm^2)

$$TMOE = \sigma/\varepsilon \quad \text{... Equation 4}$$

where:
TMOE = tensile modulus of elasticity (N/mm^2)
σ = stress at proportional limit (N/mm^2)
ε = strain at proportional limit

$$E_b = (L_f – L_0/L_0) \times 100 \quad \text{... Equation 5}$$

where:
E_b = percentage of tensile's elongation at break (%)
L_f = gauge length at break (mm)

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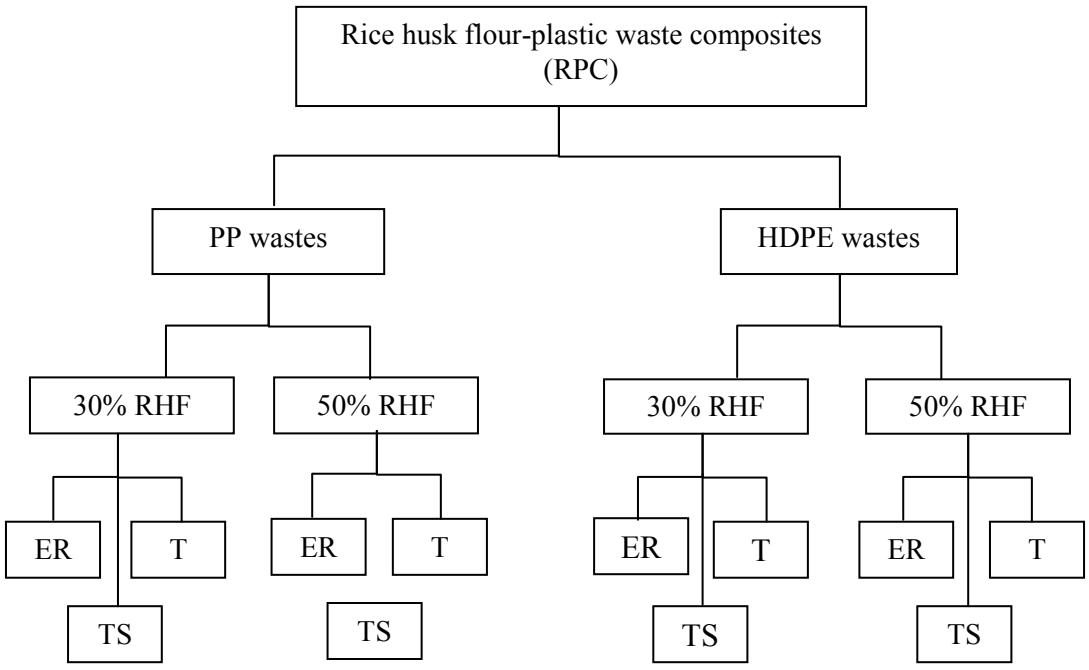
L_0 = original gauge length (mm)

(a). (b). (c).

Figure 2: (a) Electrical resistivity, (b) thermal stability, and (c) tensile strength test set-up.

Experimental design of this study is illustrated in Figure 3. Statistical Analysis Software (SAS) was used to analyze all data collected. Analysis of variance (ANOVA) and t-test method was used to analyze the significance difference of ρ, T_d, WL, TMOR, TMOE and E_b mean values of RPC at different polymer waste materials and RHF contents. The null hypothesis for which was that the plastic waste materials and RHF contents of RPC had no significant effect on all properties.



Note: ER = electrical resistivity test, TS = thermal stability test, T = tensile strength test

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Figure 3: Experimental design.

Results and Discussions

Electrical Resistivity of RPC

Figure 4 shows the mean electrical resistivity (ρ) value of RPC made from different plastic waste materials and RHF contents. ρ of RPC from PP wastes with 30% RHF content was higher (7.39×10^{11} Ohm) than 50% RHF content (4.56×10^{11} Ohm). Similar to PP, the ρ of RPC from HDPE wastes with 30% RHF content was higher (6.28×10^{11} Ohm) than 50% RHF content (2.49×10^{11} Ohm). This was due to the increment of RHF content from 30 to 50% resulted in a significant decrease in electrical resistivity. The increment of biological or natural raw material percentage causes the electrical resistivity to decrease. Being a cellulosic material, RHF has a great tendency to absorb moisture, hence influences the low electrical resistivity. Generally, ρ of RPC from PP wastes was higher than HDPE, especially for 50% RHF content. PP, with its low thermal expansion coefficient as compared to HDPE [8], possesses great bonding energy that affects its hardness of its solid body, thus influences the high electrical resistivity in this study.

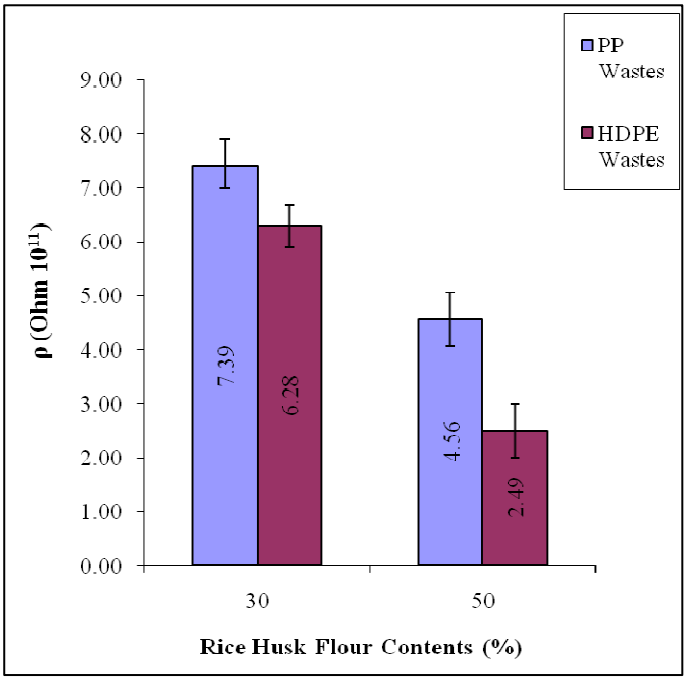

Figure 4: Mean electrical resistivity (ρ) value of RPC made from different plastic waste materials and RHF contents.

Thermal Stability of RPC

Figure 5 shows the mean decomposition temperature (T_d) value of RPC made from different plastic waste materials and RHF contents. T_d of RPC made from PP wastes with 30% RHF was slightly higher (318.3°C) than 50% RHF (317.6°C). Similar to PP, T_d of RPC from HDPE wastes

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with 30% RHF was also slightly higher (323.3°C) than 50% RHF (321.6°C). The increment of flour contents from 30 to 50% has affected the slight decrease of T_d values. According to Yang *et al.* [9], hemicellulose and cellulose are chemically active and decompose thermo-chemically in the range of 150 to 350°C. Therefore, RPC from 50% RHF increased the possibility of high decomposition of hemicelluloses and cellulose of RHF, which reduced slightly the decomposition temperature. As RHF contains high amount of silica [4], the initial ash was formed before the decomposition temperature reached. This initial formation has absaloutely reduced the decomposition temperature of RPC from 50% RHF.

Generally, T_d of RPC from HDPE wastes was higher than PP. Thermal conductivity value of HDPE was relatively higher (0.38 – 0.51 W/mK) compared to PP (0.17 – 0.22 W/mK) [8]. The property has influenced the rapid increases of decomposition temperature of RPC made from HDPE. High rate of heat transfer was occurred in HDPE with high thermal conductivity, due to the temperature dependant of this type of materials.

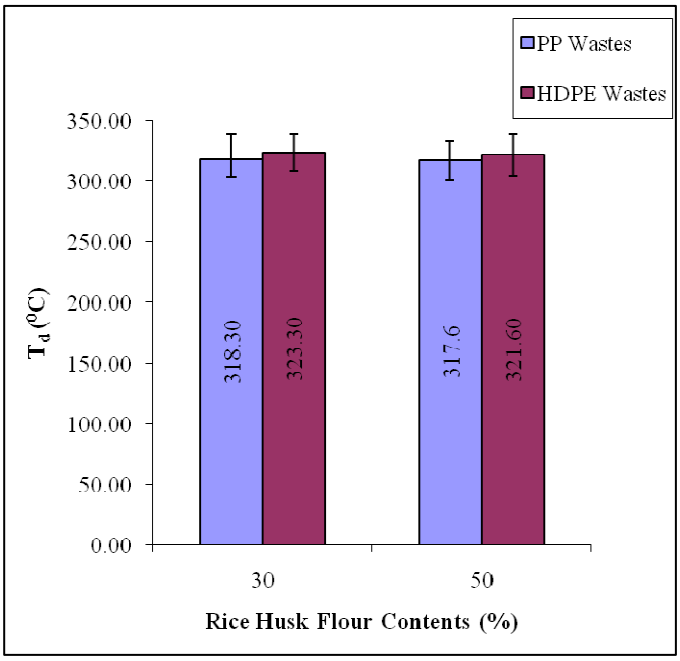

Figure 5: Mean decomposition temperature (T_d) value of RPC made from different plastic waste materials and RHF contents.

Figure 6 shows the effects of different polymer waste materials and RHF contents on the weight loss (WL) of RPC. WL of RPC made from PP as well as HDPE waste with 50% RHF was tremendously higher than 30%. The main source of WL for RPC made of 50% RHF was the formation of degraded holocellulose and lignin, as well as the formation of ash substances from the high silica content in RHF, which affected the high mean WL values at final temperature. PP with its high tendency of water absorption rate (about 0.01 – 0.03% in 24 hours) has also influenced high weight loss compared to HDPE (less than 0.01% of water absorption rate in 24 hours). A small amount of porosity due to the water absorption effects has increased the weight loss rate of PP.

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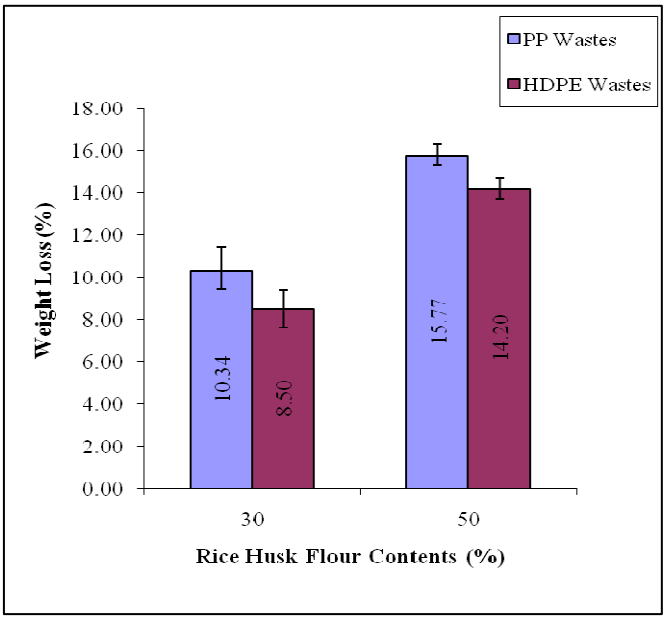

Figure 6: Mean weight loss (WL) value of RPC made from different plastic waste materials and RHF contents.

Tensile Strength of RPC

Figure 7 shows the mean TMOR and TMOE values of RPC made from different plastic waste materials and RHF contents. TMOR of RPC made from PP wastes with 30% RHF was higher (10.20 N/mm^2) than 50% RHF (10.12 N/mm^2). TMOR of RPC from HDPE wastes with 30% RHF was also higher (12.97 N/mm^2) than 50% (11.80 N/mm^2). This is due the increment of flour contents from 30 to 50% that affected the decreasing of TMOR values. Further loading of filler may not be sufficient enough to increase the TMOR due to the insufficient stress transfer within composite microstructure. Jamaludin *et al*. [10] stated that reduced rupture is a consequence of decreased deformability of rigid interphases between fibre and matrix.

However, TMOE of RPC made from PP wastes with 50% RHF was greatly higher (1265 N/mm^2) than 30% (995 N/mm^2). Similar to PP, the TMOE of RPC from HDPE wastes with 50% RHF was also higher (1067 N/mm^2) than 30% (851 N/mm^2). Similar trend was also found by Mohanty and Nayak [11], Yu *et al*. [12] and Yasin and Zuhal [13]. Yang *et al.* [9] reported that the Young's modulus increased with increasing carbon black content due to the greater matrix stiffness of carbon black with respect to polymers. The high amount of RHF (50%) as filler in this study has possibly improved the binding element between filler and plastic material, thus

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improved the mean TMOE values of RPC. The probability for filler agglomeration is also increased at higher filler loading. Lower TMOR and higher TMOE mean values are obtained for RPC containing larger quantities of RHF. According to Bengtsson *et al.* [14], increased in fibre or filler loading increases stiffness, but at the same time reduced the TMOR (toughness).

In general, RPC from HDPE shows good TMOR compared to PP. HDPE has high molecular weight to provide the great toughness (TMOR) needed for structural application [7].

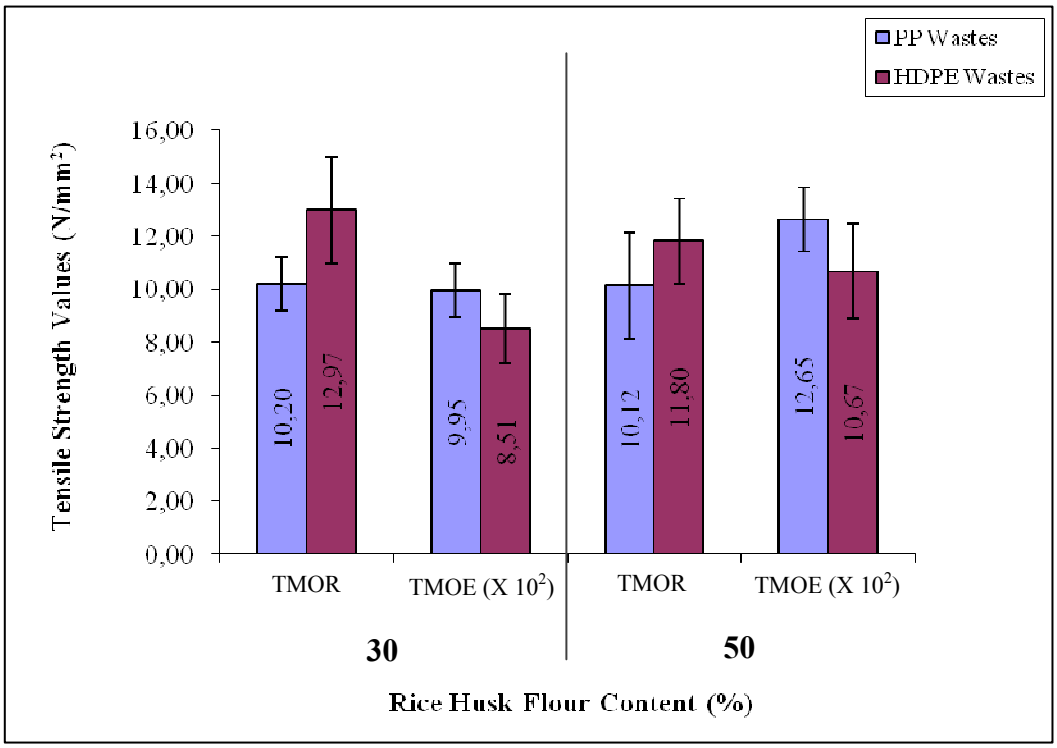

Figure 7: Mean tensile modulus of rupture (TMOR) and tensile modulus of elasticity (TMOE) values of RPC made from different plastic waste materials and RHF contents.

Figure 8 shows the effect of polymer waste materials and RHF contents on the elongation at break (E_b). The figure reveals that E_b of RPC made from PP and HDPE wastes with 30% RHF was higher than 50%. This was based on the increment of RHF content from 30 to 50% that resulted in a significant decreased in mean E_b values. This trend was similar to TMOR. According to Karmarkar *et al.* [15], this was due to the natural flour materials which have low elongation at break and restrict the polymer molecules flowing past one another.

From Figure 8, the E_b of RPC from HDPE wastes was higher than PP. E_b at break in this study was recorded at the moment of rupture of each sample, corresponds to the breaking load or maximum load. HDPE can reach up to 1000% elongation at break compared to PP which only 800% [8], in consequence relation with TMOR results in this study.

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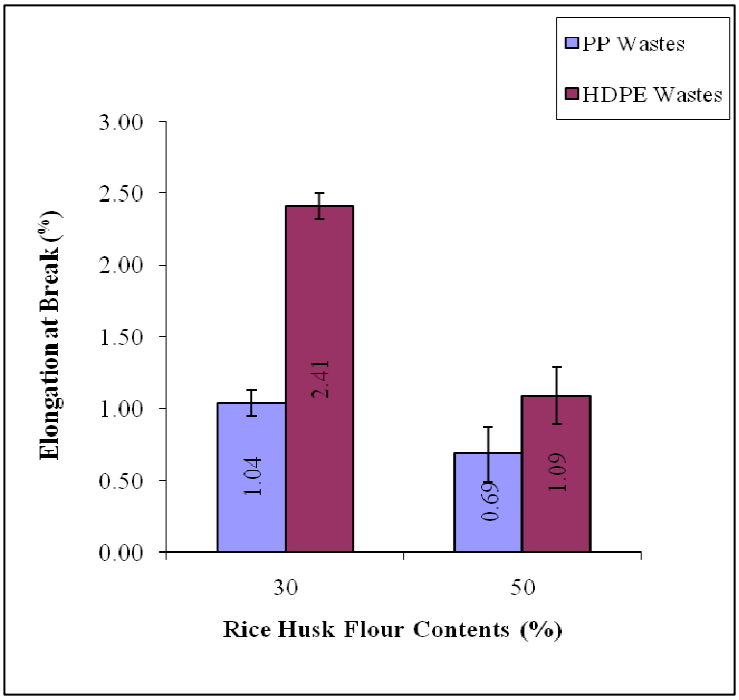

Figure 8: Mean elongation at break (E_b) value of RPC made from different plastic waste materials and RHF contents.

Analysis of variance (ANOVA)

The summary of analysis of variance (ANOVA) on the properties of RPC is shown in Table 1. Polymer waste materials and RHF contents has significant affects on all performance properties of RPC, except for T_d and WL. It is interpreted that these two factors (different polymer waste materials and different RHF content) have significant influence and difference on the of ρ, TMOR, TMOE and E_b mean values of RPC.

Table 1: Summary of the analysis of variance on the properties of RPC.

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Factors	DF	ρ	T_d	WL	TMOR	TMOE	E_b
Plastic waste materials	1	8.96*	2.51^{ns}	0.64^{ns}	12.10*	39.10*	29.94*
RHF contents	3	915.31*	0.75^{ns}	6.16^{ns}	16.43*	33.74*	379.88*

Note; - * = F values are significant at $P < 0.05$, ns = not significant, DF = degree of freedom

Conclusion

RPC composed of PP and HDPE wastes with RHF have successfully been manufactured. The results show that increased RHF content reduces all properties, except for TMOE. RHF has a great tendency to absorb moisture that influences the low electrical resistivity. The presence of hemicelluloses, cellulose and silica in RHF has also reduced the decomposition temperature and increased weight loss at the final temperature. The high amount of RHF content in the composite has improved the binding elements and filler agglomeration that influenced the good stiffness and elastic behaviour for TMOE. At the same time, this binding elements and filler agglomeration may not be sufficient enough to increase the TMOR due to the insufficient stress transfer and rigid interphase between RHF and plastics during tensile maximum load. Also, RHF with its low elongation at break behaviour has restricted the polymer molecules to flow past one another after maximum breaking load which finally reduced the elongation behaviour of RPC with high content of RHF. The high percentage of natural filler may not always indicate a good mixture in a plastic composite based upon the natural behaviour of the filler itself.

In term of polymer waste materials, RPC from HDPE indicates better thermal stability, TMOR and E_b compared to PP. The behaviour of good thermal conductivity and low water absorption of HDPE has influenced the high decomposition temperature and low weight loss. HDPE, with its high molecular weight and good elongation behaviour provides great toughness (TMOR) and elongation at break of RPC from HDPE, which may fulfill the requirement needed for structural composite application. However, RPC from PP shows good electrical resistance due to the low thermal expansion coefficient of PP as compared to HDPE. This behaviour increases bonding energy and hardness, thus influences the high electrical resistivity of RPC from PP.

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