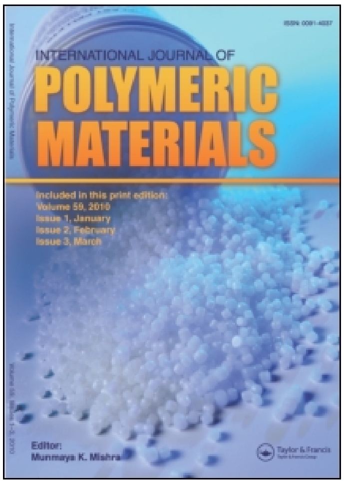

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Rice Husk Ash as Fillers in Polypropylene: A Preliminary Study

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Burnt rice husk ash yields two types of fillers namely black rice husk ash (BRHA) and white rice husk ash (WRHA). These ashes were incorporated into polypropylene using a Brabender thermoplastics mixer. Torques at various filler loadings were investigated. For both types of ashes, the mixing torques increase with filler contents; the increase being more prominent in the BRHA. Polypropylene composites of different filler loadings were compounded using a twin screw extruder and test specimens were injection molded. Thermogravimetry analysis showed a good agreement between analyzed and actual filler content. The composites were found to have a good filler distribution by the same technique. Flexural modulus and density showed an increase whereas tensile strength, elongation at break and impact strength decreased with increasing filler content. Poor adhesion between polymer and filler was indicated by SEM studies.

KEY WORDS Polypropylene, rice husk ash, filler, composite, mechanical properties.

INTRODUCTION

Filler-reinforced polypropylene has been a popular subject due to the versatility of the polypropylene to accept numerous types of fillers and reinforcements.[1-5] Usage of filled polypropylene in electrical and automotive engineering has been on the increase in recent years mainly due to its excellent stiffness property which enables it to substitute conventional materials in demanding engineering applications.[6] Fillers which merely increase bulk volume hence reduce price, are known as extender fillers while those which improve mechanical properties particularly tensile strength are termed as reinforcing fillers. Typical fillers and reinforcements for polypropylene are glass fibres, glass spheres, talc, asbestos, wood flour, calcium carbonate, silica and mica.[7]

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In this study, a relatively new type of filler that is derived from rice husk ashes (RHA) is investigated. Rice husk when burnt in open air outside the rice mill yields two types of fillers. The upper layer of the RHA mound is subjected to open burning in air and yields black ash i.e., carbonised layer. The inner layer of the mound, being subjected to higher temperature yields white ash. Black ash may also be obtained from burners where the rice husk was burnt for a short duration of time as fuel for obtaining energy.

To date, not many publications can be obtained on the subject of RHA. Use of RHA to synthesize zeolite, a type of catalyst, has been carried out by some workers,[8–10] while incorporation of zeolites into polypropylene and nylon was patented by Canard.[11] Earlier work on treatment of rice husk was described by Abbdellatif[12] where the husk was roasted in a controlled condition for less than 60 seconds. Oxygen flow was regulated and roasting occurred at specified temperature, i.e., between 600 to 800°C. The husk was roasted until it has lost 30 to 70% of its original weight. The product, a reactive organic derivative of silicon was then subjected to chemical reactions with specified chemicals to yield products that may be applied in the construction industry. Severe roasting of the rice husk that results in a weight loss of more than 70% will convert the organic siliceous material of the husk into an unreactive mineral silica. On the other hand, moderate roasting where there is less than 30% weight loss, the product will be a mixture of a carbonaceous material and amorphous silica. In relation to the present study, WRHA may be referred to the former product, i.e., mineral silica and BRHA to the latter.

WRHA used in this study has been analyzed to have about 95% silica content. Being a synthetic silica it is likely to have another important property in common with other synthetic silica, i.e., not crystalline like naturally occurring silicas. Synthetic silicas have been shown by X-ray analysis to be amorphous, thereby avoiding silicosis risk.[7] BRHA on the other hand has a lower silica content, typically about 54% and a substantial carbon content i.e. about 44%.[10] It is the aim of this study to investigate the potential of both RHAs as alternative low cost fillers for polypropylene. It is hoped that they will perform similar functions as other commercial mineral fillers, i.e., increasing stiffness, hardness and heat deflection temperature.

EXPERIMENTAL

Materials

Black rice husk ash (BRHA) and white rice husk ash (WRHA) used in this study were supplied by Chemical Technology Unit, SIRIM, Malaysia. Both ashes were ground to fine powder using a ball mill (Model William Boulton Vibro Energy Mill). The chemical and physical properties of both RHA fillers are given in Table I. No surface treatment was applied to the fillers. A locally produced polypropylene homopolymer, Propelinas 600G from Polypropylene (M) Sdn. Bhd. indicated to have a density of 0.90 g/cm^3 and a melt flow index of 12 g/10 min was chosen to be the matrix material. For comparison purpose, two commercial talc-filled poly-

TABLE I

Chemical and physical properties of white and black rice husk ashes

Property	WRHA	BRHA
Chemical Composition (weight %)[a]		
CaO	0.13	0.06
MgO	0.36	0.16
Fe_2O_3	0.05	0.03
K_2O	1.62	1.11
Na_2O	0.09	0.06
Al_2O_3	trace	trace
P_2O_5	trace	trace
SiO_2 (silica)	96.2	54.1
Loss on ignition (LOI)	1.62	44.48
Physical Properties		
Mean Particle Size (μm)[b]	13	20
Surface area (m^2/g)[c]	2.7	97.8
Density (g/cm^3)[d]	2.2	1.8

[a] Characterized using atomic absorption spectroscopy and thermogravimetry techniques.

[b] Determined by a Coulter Counter.

[c] Determined by BET method using Micrometritics FlowSorb II 2300.

[d] Measured based on ISO 8962 using a glass pyknometer.

propylene resins at 20 and 40% filler content obtained from Sumitomo BWH52 and Nagase Calp 4600G, respectively, were used.

Mixing Study

Mixing study was carried out using Brabender W50E mixer. WRHA and BRHA were oven dried at 120°C for 24 hours prior to mixing. The RHA and polypropylene were weighed and mixed proportionally to prepare samples equivalent to 10, 20, 30, 40 and 50% filler loadings. Mixing were carried out at mixer temperature of 180°C, rotor speed of 40 rpm for a period of 5 minutes. Torque versus time curve was obtained for each sample.

Processing

The RHA was compounded into polypropylene by means of a Brabender twin screw extruder (type DSK 42/7) having barrel temperatures of 190, 200 and 210°C, respectively, for the three temperature zones. The compounds were extruded through a twin 4Hmm rod die into a water bath and subsequently pelletized by a granulator. The compounded samples were then prepared into test specimens by injection molding technique. The machine used was a 20 tonne Battenfeld BA 200 CD Plus with UNILOG 400 control system. Moldings were prepared using 130 kg/cm^2 injection pressure, barrel temperature profile ranging from 175 to 195°C and cooling time of 17 seconds. A test specimen mold from Mastermould Inc. having cavities for tensile specimen according to ASTM D638 Type 1 and a rectangular bar, 125 mm × 12.5 mm × 3.13 mm, was employed.

Samples Characterization

Density of the composites was determined according to ASTM D792, Method A, i.e., by water displacement method. An electronic densimeter, AND ED-120T was employed to measure the density of the injected samples accurately to the third decimal place.

Filler content in the molded samples was analysed using Mettler TG 50 thermogravimetry analyzer. The WRHA composites samples were scanned from 50 to 550°C, at a heating rate of 20°C/min, in air with a flow rate of 200 ml/min. The BRHA composites and unfilled polypropylene samples were scanned at a same temperature range and heating rate but in an atmosphere of nitrogen with the same flow rate. The RHA residue level was calculated using STEP analysis available in Mettler Graphware data evaluation software. To check filler distribution within the final composite samples, the RHA residue levels were analyzed at three different positions along the rectangular bar specimen. These positions were at the gate, mid point and end of the bar. To study the effect of the RHA fillers on the thermal degradation of the polypropylene matrix, onset of degradation analyzes were performed on the thermogravimetry curves obtained from scans in nitrogen.

Dispersion of fillers and the adhesion between polymer matrix and filler were studied with the help of the scanning electron microscopy of the fractured composites. Scanning electron microscope (SEM), model Hitachi S0-2500 was used. Prior to scanning, samples were coated with platinum using Hitachi E102 ion sputter.

Mechanical Tests

Tensile tests were carried out on a Lloyd L1000S machine at a cross head speed of 5 mm/min. An Infrared non contacting extensometer was used to determine the elongation. The test was conducted based on ASTM D638 using type 1 specimens. Tensile strength, percentage elongation at break, means and standard deviation values were calculated by the instrument software. Five specimens were tested for each sample.

Flexural test was carried out according to ASTM D790-86, Test Method 1, Procedure A, i.e., three-point loading system. The support span was 50 mm; the diameter of the loading nose and supports were 20 mm and 10 mm, respectively. Test was conducted at a test speed of 2 mm/min with a 100 N load cell on the same machine. Lloyd software was used to detect maximum force and loads at specified strains, i.e., 0.7 and 1.0% for subsequent calculation of flexural modulus.

Izod impact test was performed on a Ceast 6456/000 Izod tester. Test specimens were prepared from the injected rectangular bar. Notching (45°) was carried out on the impact specimens using Davenport notch cutting apparatus. The test was conducted based on ASTM D256-88. A total of 10 specimens were tested for each compound.

All the mechanical tests described above have been carried out under standard laboratory condition of 27°C and 65% relative humidity, i.e., in accordance to ISO 291 standard on conditioning and testing atmospheres for tropical countries.

RESULTS AND DISCUSSION

Mixing Study

Torque versus time curves were obtained for polypropylene and its composites having various loadings of fillers. Torque values after 5 minutes running times of the mixture were recorded by the mixer evaluation software. A typical curve is shown in Figure 1. Peak torque is due to charging of sample; being cold and unmelted yet; the mixing torque is very high. Gradually as polypropylene begins to melt, the torque decreases and finally after about 4 minutes it levels off. A

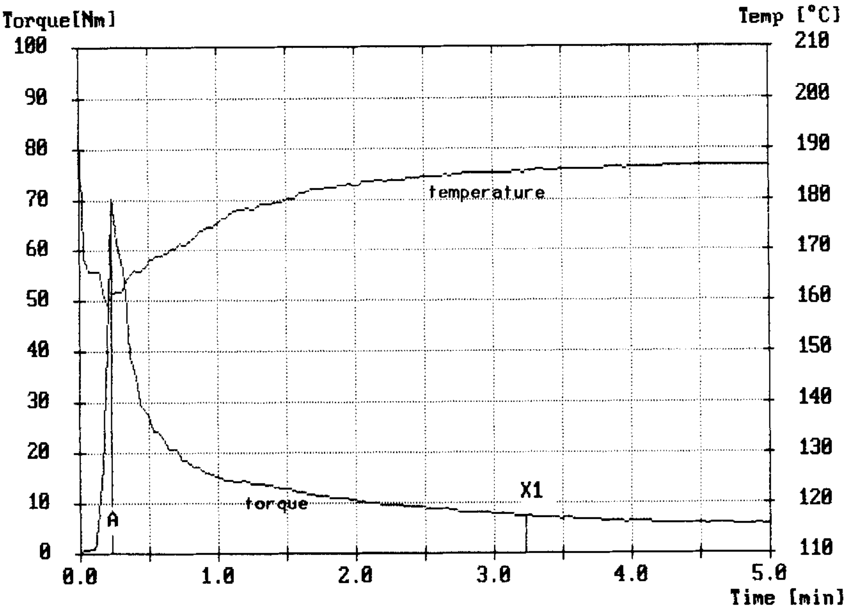

FIGURE 1 A typical torque-time curve obtained during compounding of RHA fillers in polypropylene.

slight correction was applied to the mixing time. The 5 minutes mixing does not appear to be appropriate as the actual mixing does not start at zero time but gradually as the polypropylene begins to melt. Thus the mixing time was adjusted by means of the software to be the time from the initial peak in the curve. The torque after 3 minutes from the peak is taken as the value of interest.

Figure 2 shows plots of mixing torque against filler content for both white and black RHAs. As the filler content increases, the mixing torque rises steadily. As the fine filler particles get into the polypropylene matrix, the mobility of the macromolecular chains of the polymer is reduced, thus offering more resistance to flow, i.e., high torque values. The increase is more pronounced in the black RHA composite indicating considerably more energy is required to mix the black RHA than the white RHA. This may be attributed to a few reasons. The bigger size of the BRHA particulates may be a valid reason though the contribution may not be much. The nature of the particle surface and consequently the interfacial interaction between the discrete and continuous phases may likely be the main factor. Earlier work carried out by Nielsen[13] has proved that the dispersion of filler in the polymer matrix is not perfect. Similarly as in the case of most fillers, the white and black RHA particulates are probably not very well dispersed, too. The fine RHA particulates tend to combine together to form bonded aggregates which in turn may unite to form larger structures known as agglomerates.[14] The higher torque in BRHA composite may be an indication of a higher degree of agglomeration of the particulates. Figures 3 and 4 are SEM micrographs which show some evidence of agglomeration in the RHA particles.

Samples Characterization

Table II gives the values of density for the various composites that have been determined by the water displacement technique. As expected, the density of the

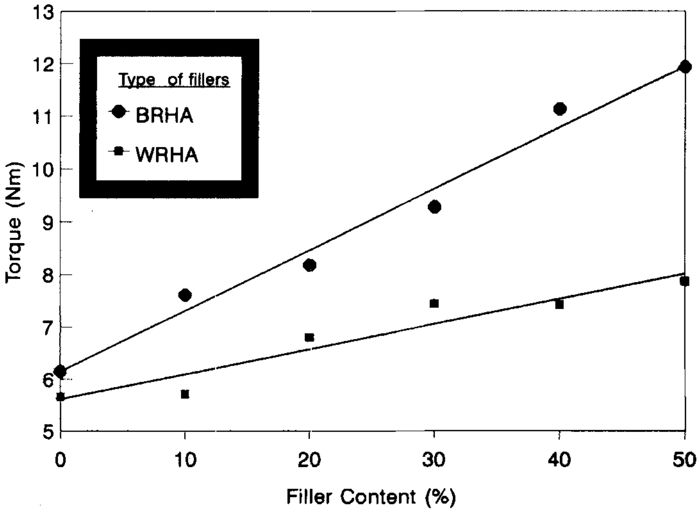

FIGURE 2 Effect of RHA fillers at different filler loadings on mixing torque after 3 minute mixing.

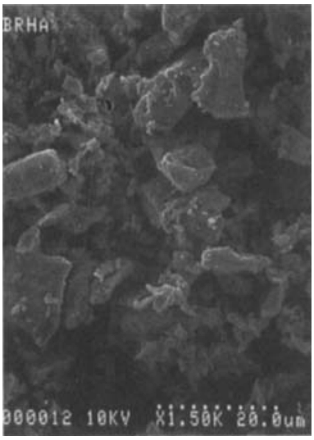

FIGURE 3 SEM micrograph of BRHA at 1500× magnification.

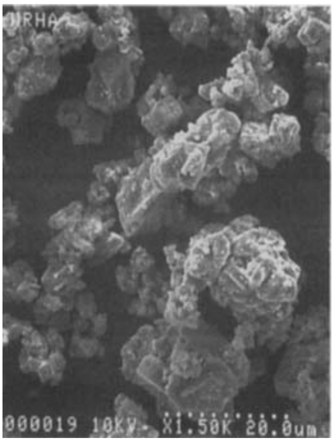

FIGURE 4 SEM micrograph of WRHA at 1500× magnification.

composites increases with increasing filler content. Since both RHA fillers have higher density relative to polypropylene, incorporation of the filler increases the density of the composites. As the WRHA is more dense than the BRHA, it follows that the WRHA composites have relatively higher density than the BRHA composites. The theoretical density of a composite may be calculated from the following equation:

$$\rho_c = [(W_f/\rho_f) + (W_m/\rho_m)]^{-1} \tag{1}$$

where ρ_c, ρ_f and ρ_m are densities of composite, filler material and polymer matrix, respectively, while W_f and W_m refer to weight fraction of filler and matrix in the composite.

Values of 2.2 and 1.8 g/cm^3 were taken as the densities of WRHA and BRHA,

TABLE II

Comparison of measured and calculated density in RHA composites

Composite Sample	Filler loading (%)	Density (g/cm^3)	
		Measured	Calculated
BRHA	10	0.96	0.96
	20	1.00	1.00
	30	1.06	1.07
	40	1.13	1.13
WRHA	10	0.97	0.97
	20	1.03	1.03
	30	1.11	1.10
	40	1.19	1.19

TABLE III

Analysed filler content in RHA composites as determined by thermogravimetry analysis

Theoretical Filler Content (%)	**Analysed Filler Content (%)**		
	WRHA	**BRHA**	**TALC**
10	10.4	10.1	-
20	18.4	19.6	20.5
30	29.9	28.1	-
40	38.8	37.2	37.2

respectively (Table I). Table II compares the values of the calculated density to the measured density. Interestingly, the calculated values agree very well with the measured values for both RHA composites.

Thermogravimetry technique was used to study the filler content and filler distribution within the final composites. The analyses reveal that the determined filler contents in both white and black RHA composites are very close to the percentage of the incorporated fillers (Table III). The technique also confirmed excellent distribution of filler particles within the matrix as filler contents determined at 3 different locations of the specimen (at the gate, mid way and at the end positions), show very close agreement. Details are as shown in Table IV. WRHA composites samples were analyzed in air to burn off the polypropylene matrix; the residue

TABLE IV

Filler content of composites (40% loading) at different locations in a molded sample to show good filler distribution

Sample	Theoretical	Measured	Filler	Content (%)
	Filler Content	Position	of	Sampling
	(%)	Gate	Midpoint	End
White RHA	40	39.4	39.6	38.8
Black RHA	40	37.6	37.8	37.8

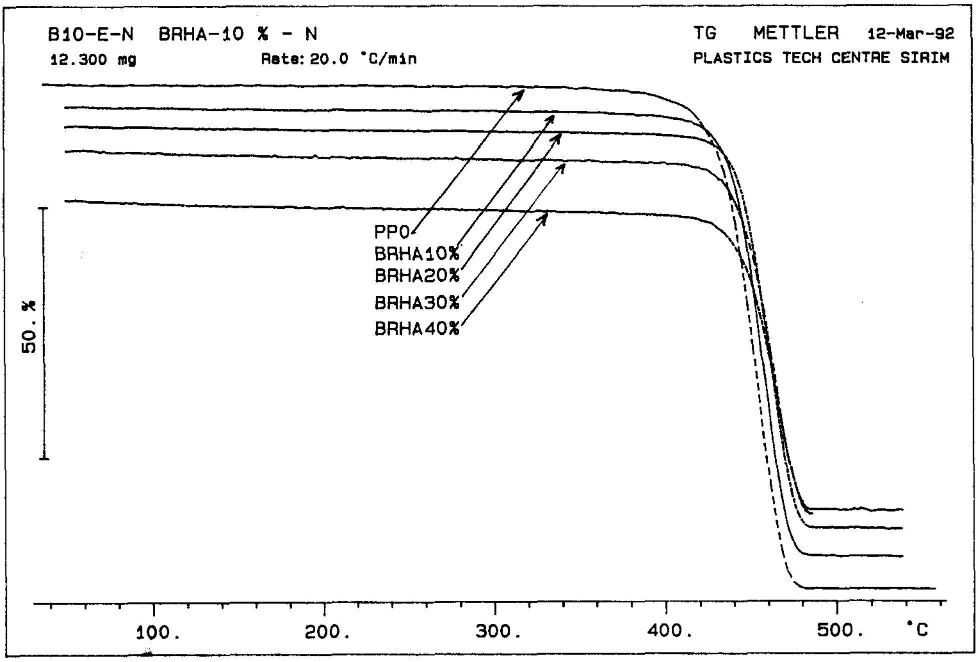

FIGURE 5 Thermogravimetry scans of BRHA composites, with filler contents ranging from 0 to 40%.

being WRHA which consist predominantly of silica. BRHA composites were analysed in oxygen-free nitrogen (OFN) to burn off the polypropylene matrix but without oxidising the carbon content of the RHA. Figure 5 shows typical thermogravimetry scans of BRHA composites.

The effect of the RHA fillers on the degradation of the polypropylene in an atmosphere of nitrogen was investigated too. Onset of degradation of each sample was analysed from the respective thermogravimetry curve. Plot of the onset of degradation temperature with respect to filler loading is given in Figure 6. It was noticed that the degradation temperatures of the BRHA samples increase with

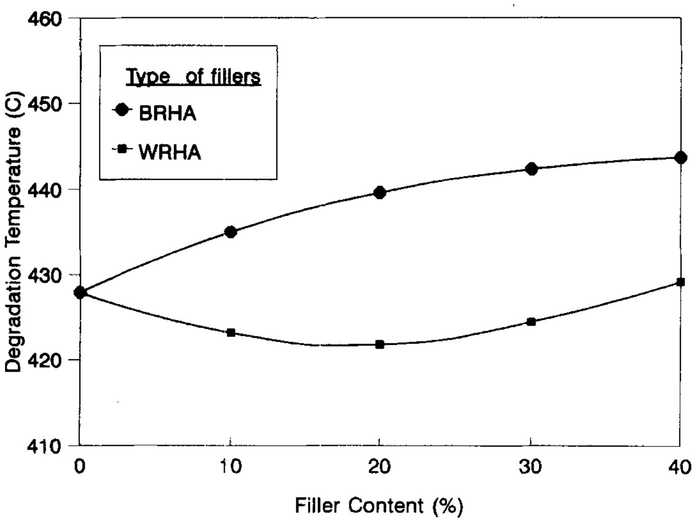

FIGURE 6 Plot of onset of degradation temperatures versus filler loading for WRHA and RHA composites.

filler loading while the WRHA samples showed no significant changes. Probable explanation for this behavior may be offered in terms of the heat absorption capacity of the RHA fillers. BRHA particles being black in colour are excellent absorbers of heat energy. As their quantity increase, more heat is taken up by the RHA component of the composite. Thermal degradation of the polypropylene matrix on the other hand, occurs only after a certain amount of heat energy has been absorbed by the material. The heat initiates the degradation process and breaking down of the matrix structure by causing molecular chains rupture.[15] With increasing filler content, more heat is being absorbed by the BRHA particles. Higher temperature is therefore required to supply the threshold energy for the commencement of the degradation process. Thus as BRHA filler content increases, there is a gradual favourable shift of the degradation temperature upwards.

Mechanical Tests

The overall results for the mechanical tests are summarized in Table V. The detail analyses of tensile, flexural and Izod impact tests results may be elucidated as follows.

Tensile properties

Fibre type fillers normally improve tensile strength of the composite as the fibres are able to support stresses transferred from the polymer via the interfacial region.[16] For irregularly shaped fillers, strength of the composites frequently decreases due to the inability of the fillers to support stresses transfer from the polymer matrix.[2,4] In this study, the tensile strength of both BRHA and WRHA composites decreases steadily with increasing filler loading. SEM micrographs in Figures 3 and 4 have indicated that RHA fillers are of irregular shape and have a tendency to form

TABLE V

Results of mechanical tests on various polypropylene composites

Property	Filler content /%	Unfilled Polypropylene	WRHA	BRHA	TALC
Tensile Strength (MPa)	0	34.5			
	10		31.2	28.9	
	20		27.0	26.9	30.5
	30		24.9	23.9	
	40		21.8	23.5	28.0
Elongation (%)	0	>600 %	>600 %		
	10	>300 %	26		
	20	127	11.2	66.6	30.5
	30	50.6	6.4		
	40	22.1	1.7	3.3	28.0
Flexural Modulus (MPa)	0	1579			
	10		1639	1897	
	20		1936	2306	2692
	30		2292	2639	
	40		2544	3124	3610
Izod Impact Strength (J/m)	0	110			
	10		72.6	45.8	
	20		68.4	42.3	92.7
	30		54.4	28.5	
	40		50	24.8	53.2

agglomerates. These factors plus the fact that no coupling agent was present in the composites, result in a poor adhesion of the RHA particulates to the polypropylene matrix. Weak interfacial regions imply that the transfer of stress from the polymer matrix to the filler will not be efficient. Hence the drop in the tensile strength is not unexpected. The commercial polypropylene talc composites do appear to have superior tensile strength.

Nicolais and Nicodemo[17] have proposed the empirical equation to estimate the lower bound of tensile strength of a filled polymer. The tensile strength of the composite, σ_c is related to the matrix tensile strength, σ_p by:

$$\sigma_c = \sigma_p(1 - a\phi^b) \qquad (2)$$

where ϕ is the volume fraction of the filler, a and b are constants related to stress concentration and geometry of the filler, respectively. For spherical fillers with no adhesion to the polymer matrix, the value of a has been found equal to 1.21. For the case of some adhesion, a becomes smaller than 1.21. The constant b is equal to 1 if the material fails by planar fracture and 2/3 if failure is by random fracture. While both modes of fractures have been observed, random fracture is more common.[2] For the purpose of calculation, if the RHA fillers are assumed to be spherical and the composites failing by random fractures, the lower bound curves (LBC) for each composite will be as shown in Figure 7. The discrepancy observed between the theoretical lower bound curves and the experimental curves is probably due to the assumption that the RHA particulates are spherical and exist as discrete particles while in reality they have irregular shapes and are in the form of agglomerates.

The upper bound of a tensile strength of a polymer filled with spherical particles has been forwarded by Leidner and Woodhams[18] as:

$$\sigma_c = (\sigma_I + C\tau_m) + \sigma_p(1 - \phi) \qquad (3)$$

where σ_I represents the interfacial tensile strength between the polymer and filler, C is a constant (determined to be 0.83) and τ_m the shear strength of the polymer. However, the upper bound response is not examined in this study since the response

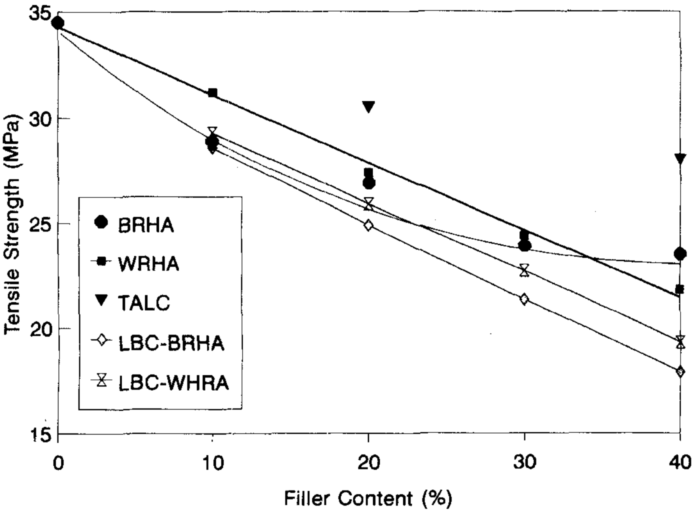

FIGURE 7 Effect of filler loading on the tensile strength of RHA filled polypropylene. Talc represents commercial polypropylene talc composite.

assumes strong adhesion between polymer and filler. Such assumption is not valid in the present RHA composites for two reasons. Firstly, no coupling agent has been incorporated into the RHA composites to improve the adhesion between the RHA fillers and polypropylene matrix. Secondly, SEM micrographs show no adherence of matrix on the surface of the RHA fillers indicating poor adhesion. Further discussion of the SEM micrographs will be dealt with later.

Figure 8 shows the effect of filler loadings on the elongation at break, ε_B for the various RHA composites. The ε_B for the unfilled polypropylene and WRHA composite at 10% filler loading samples are not shown in the figure because these samples did not break till the maximum limit of the testing machine. They have ε_B values of more than 600%. For the rest of the RHA composites, the expected trend is observed, i.e., the ε_B decreases rapidly with increasing filler content. The drop in the elongation at break is much more apparent in BRHA composites; where even at 10% filler loading, the ε_B was reduced from excess of 600% to 26% only. At maximum loading of 40%, the BRHA composite has an extremely low ε_B of 1.7%.

Some idea as to the mode of failures can be obtained from the SEM studies on the appearance of the fractured surface of the specimens. Figures 9 and 10 show typical tensile fracture surfaces of BRHA and WRHA composites, respectively. These figures indicate that failure of BRHA composites are brittle in nature while WRHA composites on the other hand show some degree of matrix tearing and have undergone ductile failures. Unfilled polypropylene itself exhibits ductile failures under the present testing condition. It thus appears that the incorporation of the WRHA does not alter the ductile mode of failure of the polypropylene matrix while the BRHA results in a marked transition of the failure mode from ductile to brittle fracture.

When subjected to stress at a low strain rate, the macromolecular chains of the polymer align themselves along the axial direction in line to the applied tensile

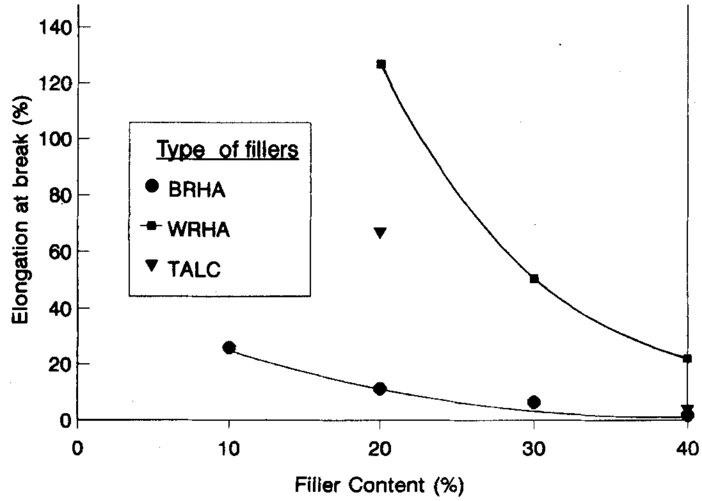

FIGURE 8 The effect of various types of fillers on the elongation at break for polypropylene composites.

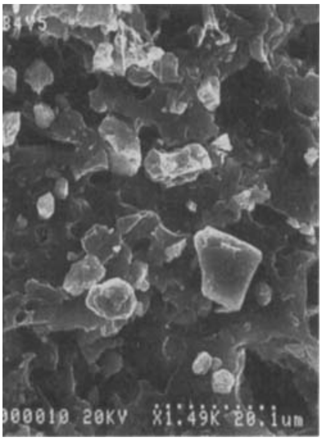

FIGURE 9 SEM micrograph of BRHA composite at 40% filler loading after tensile fracture magnification 1500×.

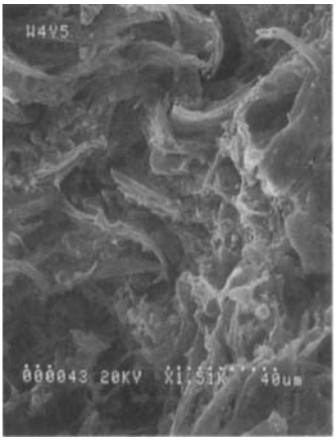

FIGURE 10 SEM micrograph of WRHA composite's fracture surface at 40% filler loading after tensile fracture (1500×).

force. Incorporation of fillers that have poor adhesion to the polymer matrix seems to cause interruption in the alignment process of the chains. This phenomenon may be observed in the SEM micrograph shown in Figure 11. When filler concentration is increased, more weak interfacial regions between the filler surface and polypropylene matrix are formed. As cracks travel easier through the weaker interfacial regions, the composite therefore fractures at a lower degree of elongation with increasing filler content.

Flexural Modulus

The flexural modulus data for all the composite systems investigated are contained in Figure 12. As expected the modulus, which indicates material stiffness increases

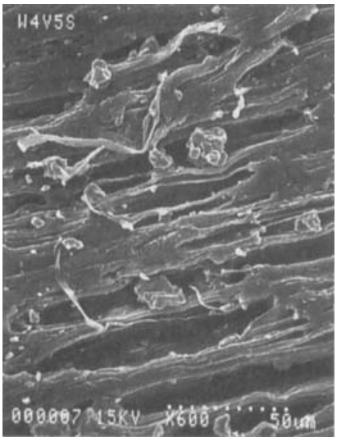

FIGURE 11 SEM micrograph indicating ductile fracture of WRHA composite at 10% filler loading (600 ×), longitudinal view.

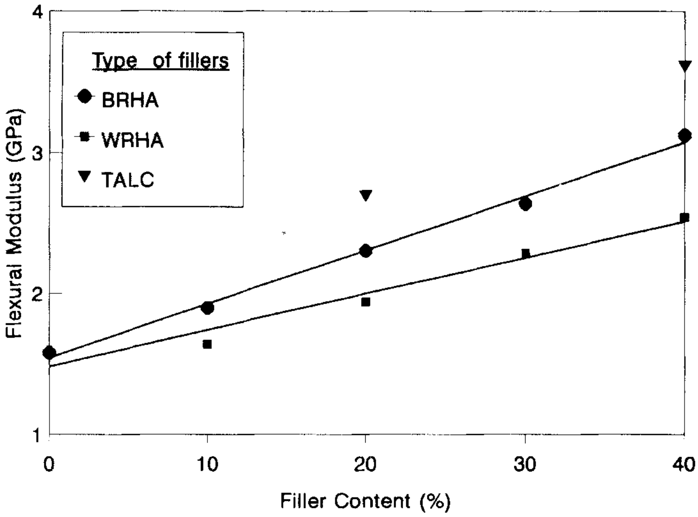

FIGURE 12 Flexural modulus versus filler loadings for polypropylene composites.

steadily with greater filler content. The BRHA composite shows relatively higher stiffness than the WRHA composite; but overall, the modulus is still lower than the modulus of the commercial talc composites. Recently, Riley *et al.*[19] have shown that flexural modulus of polymer composites based on four types of mineral fillers namely, calcium carbonate, clay, talc and mica, increases with both the filler loading and aspect ratio of the filler particles. They noted that the degree of stiffness increases in the following order: mica > talc > clay > calcium carbonate, i.e., mica composite showed the greatest increase while carbonate the least. According to them, the factors affecting the composite modulus were filler modulus, filler loading and filler aspect ratio. High modulus composite requires filler particle of high modulus and high aspect ratio and preferably at high filler loading.

Modulus of composites has been represented by a number of equations, the first and simplest being from Einstein, $E_R = 1 + 2.5\phi$ where E_R is relative modulus (of composite to polymer) and ϕ is volume fraction of filler.[2] This equation is only applicable to materials filled with low concentrations of noninteractive spheres. Padawar and Beecher[20] put forward their theory on filling with planar materials where the effect of particle edge to edge interaction is considered but not flake misalignment. Their theory may be represented by the following equation:

$$E_C = E_P(1 - \phi) + E_F\phi\text{MRF} \qquad (4)$$

where E_C is modulus of composite; E_P is modulus of polymer; E_F is modulus of filler. MRF is modulus reduction factor which is a fraction (<1) which approaches unity with increasing flake aspect ratio and filler loading. According to Riley *et al.*, the equation is only successful in predicting qualitative trends in the increase of modulus with loading of a high aspect ratio filler. For low aspect ratio fillers, the equation from Lewis and Nielsen is more applicable. Lewis and Nielsen put forward the following equation:

$$E_C/E_P = (1 + \eta\xi\phi)/(1 - \eta\psi\phi) \qquad (5)$$

where η is constant related to Einstein coefficient; ξ is related to relative moduli of filler and polymer; ψ is a parameter dependent on the maximum packing fraction of the system. Theoretical calculation for modulus is not possible at this moment because the value of parameter ξ is not available since modulus of the RHA filler is not known yet. Jilken *et al.*[4] obtained good agreement with Lewis and Nelsen equation for their experimental results on dolomite (filler modulus approx. 35 GPa) by letting $\xi = 3$. Likewise reasonably close agreement was obtained when they related their experimental data on mica filler (flakes) with the Padawar-Beecher equation.

Izod Impact Test

Figure 13 shows the effect of filler loading on the notched Izod impact strength of filled polypropylene. Incorporation of RHA fillers into the polypropylene causes a sudden drop in the impact strength as shown by the RHA composites at 10% filler loading. Subsequent addition of fillers results in a more gradual decrease in impact strength. The fall in the impact strength is more prominant in the BRHA composites. Without any coupling agent to modify the RHA surface characteristics, poor wetting of the particles by the polypropylene matrix is expected. This gives rise to poor interfacial adhesion between the filler and the polymer matrix resulting in weak interfacial regions. During the impact test, cracks travel through both the polymer as well as along the weaker interfacial regions. The latter cannot resist crack propagation as effectively as the polymer region, therefore reducing the impact strength.[20] Increasing the filler content merely increases the interfacial regions which exaggerates the weakening of the resulting composites to crack propagation. A similar trend has been observed by Bigg[2] for other particulate filled polymers.

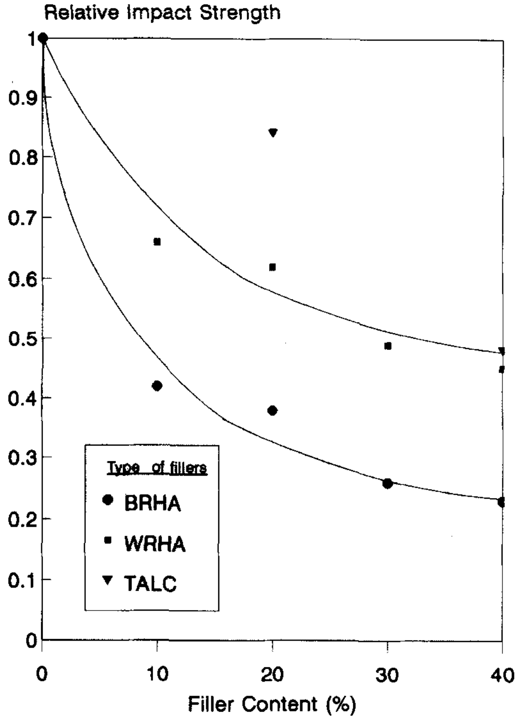

FIGURE 13 Relative impact strength of RHA and talc filled polypropylene.

Figure 5 also indicates that the WRHA composite possess significantly higher impact strength than the BRHA composite. This may be attributed to the greater agglomeration of the BRHA particulates. Ess and Hornsby[14] suggested that there are several adhesion forces holding the particles together in agglomerates viz. interlocking, electrostatic, van der Waals, liquid bridge and solid bridge forces. These combined interparticle forces are, however, weaker than chemical bonding of the polypropylene matrix and are therefore easier to be overcome by external mechanical energy (impact energy in this case) during fracture. Consequently BRHA particulates which are believed to have a higher degree of agglomeration yield a more brittle composite and is more susceptible to cracking. It is also interesting to note that the trend in the fall of the impact strength of the RHA composite is quite similar. And, as expected, the impact strength for the commercial talc composites is higher than that for the RHA composite.

CONCLUSION

The work presented in this paper is preliminary in nature. The main intention of the study is to gauge the possibility of extending the application of the low-cost rice husk ashes (RHA) as alternative filler materials in polypropylene. It has been shown that even without any coupling agent, the mechanical properties of the RHA composites are not much inferior when compared to the commercial talc-filled polypropylene materials commonly used in automotive applications. The Izod impact strength of the WRHA composite at 40% loading for example is very close to that of the commercial resins while its elongation at break is superior to that of the latter. In terms of stiffness, the BRHA composite exhibits higher modulus than the WRHA composite and the moduli at 20 and 40% filler content are not far below the respective values of the commercial composites. Although the tensile strength of both composites are lower than the commercial grades, the gap is not substantial. With a suitable coupling agent, it is hoped the interfacial adhension between the RHA particulates and polypropylene matrix can be improved and thus lead to superior mechanical properties.

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References

1. C. D. Han, T. H. Kwack and H. L. Luo, *J. Appl. Polym. Sci.*, **29**, 2599 (1984).
2. D. M. Bigg, *Polym. Comp.*, **8**, 115 (1987).
3. M. Bramuzzo, A. Savadori and D. Bacci, *Polym. Comp.*, **6**, 1 (1985).
4. L. Jilken, G. Malhammar and R. Selden, *Polym. Test.*, **10**, 329 (1991).
5. T. Kawalewski and A. Galeski, *J. Appl. Polym. Sci.*, **32**, 2919 (1986).
6. A. Weber, *Plast. Rubber Process. Appl.*, **14**, 65 (1990).
7. A. W. Bosshard and H. P. Schlumpf, in *Plastics Additives Handbook*, R. Gachter and H. Muller, eds., Carl Hanser Verlag, Munich (1987).
8. P. K. Bajpai, M. S. Rao and K. V. G. K. Gakhale, *Ind. Eng. Chem. Prod. Res. Dev.*, **20**, 721 (1981).
9. A. K. Dalal, M. S. Rao and K. V. G. K. Gokhale, *Ind. Eng. Chem. Prod. Res. Dev.*, **24**, 465 (1985).
10. S. S. Chen and A. K. Yeoh, *J. Perindustrian SIRIM*, to appear.
11. P. Canard, United States Patent, 4,420,582 (1982).
12. A. E. D. Abbdellatif, United States Patent, 3,574,816 (1971).
13. L. E. Nielsen, *J. Polym. Sci. Polym. Phy. Ed.*, **17**, 1897 (1979).
14. J. W. Ess and P. R. Hornsby, *Plast. Rubber Process. Appl.*, **8**, 147 (1987).
15. F. W. Billmeyer, *Textbook of Polymer Science*, John Wiley & Sons, New York (1971).
16. M. J. Folkes, *Short Fibre Reinforced Thermoplastics*, Research Studies Press (John Wiley), Chichester (1982).
17. L. Nicolais and L. Nicodemo, *Polym. Eng. Sci.*, **13**, 469 (1973).
18. J. Leidner and R. T. Woodhams, *J. Appl. Polym. Sci.*, **18**, 1639 (1974).
19. A. M. Riley, C. D. Paynter, P. M. McGenity and J. M. Adams, *Plast. Rubber Process. Appl.*, **14**, 85 (1990).
20. G. E. Padawar and N. Beecher, *Polym. Eng. Sci.*, **10**, 185 (1970).
21. K. Kendall, *British Polym. J.*, **10**, 35 (1977).