

Properties of Polypropylene Filled with Chemically Treated Rice Husk

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Received 2 March 2007; accepted 19 May 2008

DOI 10.1002/app.28747

Published online 14 July 2008 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Finely ground rice husk was used as a filler in two commercial grades of polypropylene (PP) in different amounts. Rice husk powder was chemically treated with dilute hydrochloric acid, dilute sodium hydroxide solution, and dimethyl sulfoxide. The mechanical, thermal, and rheological properties of PP filled with untreated and treated rice husk powder were determined. Effectiveness of PP grafted with acrylic acid, PP-

g-AA, as a compatibilizer was examined. Rice husk powder treated with acid showed significant improvement in the flexural modulus of PP and also less water absorption. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 110: 1271–1279, 2008

Key words: poly(propylene) (PP); fillers; mechanical properties; rheology

INTRODUCTION

Polypropylene (PP) is one of the widely used thermoplastic materials due to its low cost, easy availability, and wide spectrum of properties. However, its relatively low stiffness limits its applications. Typical fillers and reinforcements for PP such as calcium carbonate, talc, mica, and glass fiber have been used for many years to overcome this drawback and also to reduce the material cost.^{1–5} In last few years, the utilization of fibers and powders derived from agricultural sources (such as sisal, pineapple, palm oil fruit bunch) has attracted attention of many and it has become a subject of interest in polymer composites, mainly due to their low densities, low cost, nonabrasiveness, high filling levels, recyclability, biodegradability, and above all the availability from renewable sources.^{6–11}

Rice husk is one of the globally available major agrowaste products. Its typical composition is α cellulose 35%, hemicellulose 25%, lignin 20%, and ash 17% (mainly silica, 94%) by weight.¹² Because of high cellulosic composition, the main drawback of rice husk is its high tendency to absorb moisture from the environment. Silica in rice husk is known to be highly pure and amorphous with high surface area and surface reactivity.¹³ Most of the industrial

uses of rice husk pertain to the silica present in the husk. These include the preparation of silica, silica carbide, silicon nitride, silicon, zeolite, etc., from rice husk.^{14,15}

Mechanical and morphological study of PP composites filled with rice husk have been reported in the literature.^{16,17} Ishak et al.¹⁸ incorporated rice husk into PP and determined the water absorption characteristics of the composites. They observed that amount of water absorbed increased with increasing concentration of rice husk and tensile strength of the composites decreased due to water absorption. Rice husk ash has also been studied as an alternative filler to commercial silica. It is a classic example of extraction of inorganic fillers (predominantly silica) from rice husk, by a thermal degradation (or burning) process.^{19–22} Use of two types of rice husk ash, namely, white rice husk ash and black rice husk ash, into PP as a filler has been reported by Faud et al.²³ They have shown that PP composites filled with rice husk ash did not exhibit any attractive enhancement of mechanical properties. Furthermore, tensile and impact properties were significantly inferior to commercial talc filled PP composites, suggesting no particular advantages of using rice husk ash in place of talc in PP.

Rozman et al. and Sun et al.^{24,25} have studied the properties of composites of rice husk powder with polystyrene and with PP. They have observed that chemical modification of rice husk with glycidyl methacrylate enhanced the tensile, flexural, and impact properties of polystyrene.²⁴ They have attributed this effect to the increased interaction at the interfacial region between glycidyl methacrylate-

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Contract grant sponsor: General Electric (GE) Fund Scholarship Program.

modified rice husk surface and polystyrene. This modification also improved the dimensional stability of the composites. Rice husk treated with acrylic acid improved the tensile strength of the PP-rice husk composites, but the changes were not significant.

Although rice husk is available abundantly around the globe, there is no work on treating powdered rice husk chemically to remove portion of hemicellulose, lignin, or silica and using the same as a filler in thermoplastic matrix. Therefore, this work is aimed to study effect of chemical treatment of rice husk on the mechanical, morphological, rheological, and thermal properties of PP filled with such differently treated rice husk.

EXPERIMENTAL

Materials

Commercial grades of PP (MFI 11 g/10 min and 3 g/10 min at 230°C/2.16 kg) were used (Reliance Industries, Mumbai, India). Rice husk (RH) was procured from a local rice mill, and it was pulverized into fine powder by ball mill. Bulk density, density, and particle size of rice husk powder were 0.54 g/mL, 1.43 g/mL, and 100–140 mesh (105–150 μm), respectively.

PP grafted with acrylic acid (PP-g-AA) was used as compatibilizer (Uniroyal Chemical, Middlebury, CT; Grade Polybond 1002). A wetting agent (FINALUX G-3, an oleochemical derivative of complex ester blend) was supplied by Fine Organics (Mumbai, India). Dispersing agent (LUWAX^R, A PULVER, BASF) was also used in small quantity.

Treatment of rice husk powder

Acid treatment

Rice husk powder was mixed with dilute hydrochloric acid (10% concentration by weight). The amount of dilute acid used was 5 L/kg of rice husk powder. The mixture was maintained at 90–95°C for different periods of time with mild stirring. After cooling the mass, the mixture was filtered. The residue was washed with water several times to free it from acid. The acid free powder was then dried in air circulating oven maintained at 100–110°C. The acid treatment would hydrolyze and remove the low molecular weight hemicellulose present in rice husk.

Alkali treatment

Rice husk was treated with 2% NaOH in similar fashion as in acid treatment. With alkali treatment, a portion of silica and lignin from rice husk is

removed. Some minor quantities of low molecular weight cellulose may also become water soluble.

Acid and alkali treatment

Rice husk was first treated with 10% HCl (90 min) followed by 2% NaOH solution (90 min) as mentioned earlier. This would remove low molecular weight cellulose (acid treatment) and a part of silica and lignin (alkali treatment).

Acid and dimethyl sulfoxide treatment

Rice husk was treated with dilute 10% HCl acid solution (25 min) followed by extraction with dimethyl sulfoxide (DMSO) at 90–95°C for 30 min. Treatment with DMSO would remove part of lignin but not the silica as was the case in acid and alkali treatment.

COMPOUNDING

Rice husk powder was dried in air circulating oven at 105°C for 6 h prior to blending with PP. For all the compositions, PP, wetting agent (0.2%), dispersing agent (1%), and rice husk with an appropriate composition were tumble mixed for 30 min, and the mixture was melt blended using corotating twin screw extruder (Model MP 19 PC; APV Baker, Peterborough, UK) having *L/D* ratio of 25 : 1. The screw speed was maintained at 60 rpm and the temperature profile for compounding was 150, 170, 190, and 210°C for four zones and 220°C for the die zone. The extrudate from the screw compounder was water cooled and pelletized. The extruded pellets were dried and molded into standard ASTM specimens at a temperature profile of 190, 210, and 230°C for the nozzle, by using microprocessor based injection-molding machine (Boolani Machineries, Mumbai, India).

Initially, powdered rice husk was incorporated into PP upto 30% on w/w basis. Effect of PP-g-AA as a compatibilizer was studied for one composition, namely, 80 : 20 (80% PP and 20% rice husk). The concentration of compatibilizer was varied at 1, 3, and 5%. Different chemically treated rice husk powder was incorporated without any compatibilizer. The blend composition was 90 : 10.

PP filled with 10% treated rice husk are designated as: A10, A25, A45, and A90, where A stands for acid treatment and numbers 10, 25, 45, and 90 indicate time of treatment; AL90 is for alkali treatment for 90 min, A90AL90 for combined treatment of 90 min of acid treatment, and 90 min of alkali treatment and A25DMSO30 stands for acid treatment of 25 min followed by DMSO treatment for 30 min.

TESTING

Mechanical properties

Tensile strength, elongation at break, flexural strength, and flexural modulus were measured using universal tensile testing machine (LR-50K; Lloyd Instrument, Hampshire, UK), according to ASTM D 638 M-91 and ASTM D 790 M-92, respectively. The crosshead speed was 50 and 2.8 mm/min for tensile tests and flexural tests, respectively. The notch was cut using a motorized notch-cutting machine (Polytest model 1; Ray Ran, Warwickshire, UK). Notched Izod impact strengths were measured at ambient conditions according to the ASTM D 256 method by an Impact tester (Avery Denison, Cramlington, UK) with striking velocity of 3.46 m/s, employing a 2.7 J striker.

Rheological properties

Melt viscosity at 230°C was measured for all samples over a shear rate range of 0.01–100 s⁻¹. Viscosity at low shear rates was measured using a rotational viscometer (HAKKE RT-10, Karlsruhe, Germany) employing the parallel plate (35 mm diameter) assembly. Viscosity at high shear rates (50–5000 s⁻¹) was measured using a capillary viscometer (Rosand Precision, Stourbridge, UK) fitted with two capillaries $L/D = 16$ (diameter 1 mm) and $L/D = 0$ (actual length 0.26 mm). These capillary rheological data were subjected to Bagley and Rabinowitsch corrections.

Thermal analysis

Thermal analysis was carried out using differential scanning calorimeter (DSC) (DSC-7; Perkin-Elmer, Seer Green, Bucks, UK). A scanning rate of 10°C/min was used for both heating and cooling cycle, under a nitrogen purge of 30 mL/min.

Vicat softening point and heat distortion temperature

Vicat softening point and heat distortion temperature (HDT) of all the filled PP as well as virgin materials were measured by Vicat Softening Point Instrument (Davenport, Hampshire, UK). The specimen used for this test was a rectangular bar of dimensions (in mm) 125 × 13 × 6.5 (length × breadth × thickness). The specimen was dipped in a silicon oil bath, which was heated at a rate of 2°C/min. The oil was mildly stirred continuously and circulated, for uniform temperature. Vicat softening point and HDT were measured as per ASTM specification D 1525 and D 648, respectively. The entire operation was computer controlled. Each test was duplicated.

Melt flow index

The melt flow index (MFI) was determined using MFI machine (Davenport). The MFI expressed as g/10 min was determined at 230°C and the load was 2.16 kg.

Water absorption tests

The water absorption was determined by immersing the sample of dimension 2 × 25 × 25 mm³. in distilled water at 23°C for 96 h, periodically measuring the increase in weight of the samples. The water absorption, M_t , at anytime was calculated as

$$M_t = (W_w - W_d) \times 100/W_d$$

where W_d and W_w are original dry weight and wet weight after exposure for a specified period, respectively.

Mechanical properties of PP filled with rice husk were evaluated when water was allowed to absorb and also after redrying.

Morphology

The morphological characteristics of rice husk powder and the filled PP were examined by scanning electron microscopy (Jeol JSM 5800, Tokyo, Japan) and operating voltage was 20 kV. The impact fractured specimens were coated with gold.

RESULTS AND DISCUSSION

Effect of chemical treatment on weight loss of rice husk

Table I shows the weight loss in dry rice husk powder due to various treatments. It is very clear that percentage weight loss has increased with time of acid treatment. It is also apparent that weight loss is rapid initially and then it seems to level off. Maximum hemicellulose content of rice husk reported in literature is about 25%. Therefore, treatment for about 90 min may be adequate for removal of most hemicellulose.

During alkali treatment, there was a change in color of rice husk. Its appearance became lighter in color. The alkali treatment is expected to remove most of the lignin and part of silica.

The loss in weight due to combined treatment of acid and alkali seems to be very close to the addition of loss in weight due to individual treatments. Similarly, when acid treatment was followed by DMSO treatment, additional loss in weight was observed, which could be due to partial removal of some lignin by DMSO.

TABLE I
Weight Loss of Rice Husk by Different Chemical Treatment

Treatment	Weight loss (%)
10 min acid treatment	6
25 min acid treatment	12.3
45 min acid treatment	15.5
90 min acid treatment	22
90 min alkali treatment	26
Acid followed by alkali treatment	44.5
25 min acid followed by DMSO treatment	16.4

Properties of PP filled with untreated rice husk

Table II shows the properties of two different grades of PP filled with untreated rice husk. Tensile strength of PP of 11 MFI has remained unaffected till the concentration is about 10–15%. It decreases marginally when the concentration of rice husk is further increased. Percentage elongation at break has decreased rapidly even when concentration of rice husk is 5%. At higher concentration, it has further decreased marginally. The flexural strength has remained unaffected. Impact strength remained unaffected till the concentration of rice husk is about 10%. At higher concentration, it has decreased very marginally.

Most significant effect is on flexural modulus and also on HDT. The flexural modulus has increased substantially with increasing concentration of rice husk. Similarly, HDT has increased although Vicat softening point has remained same for all compositions.

Effect of rice husk loading on the properties of PP of 3 MFI shows similar trend but the extent of change is different. Although the tensile strength of virgin PP of 3 MFI is somewhat higher than that of 11 MFI, tensile strength of the filled PP are comparable at a given concentration of rice husk. The impact strength of PP of 3 MFI is also higher than

that of PP of 11 MFI. However, impact strength of the composites are comparable at a given concentration of rice husk. The flexural modulus has increased substantially for this grade of PP also. MFI seems to increase slightly at lower loading of rice husk powder for both grades of PP. Slight increase in MFI may indicate noncompatible nature and slip between rice husk powder particle and PP matrix.

It is very clear that flexural modulus of PP has increased substantially due to addition of rice husk. It is also apparent that level of increase of flexural modulus at higher loading, i.e., 30% is not very substantial over that at 20% loading. At higher loading of such filler, one may lose tensile strength or impact strength. Therefore it may be worthwhile to study the effect of compatibilizer at 20% rice husk loading so that possible increase in flexural modulus will be obtained without losing tensile strength or impact strength to any significant level. Therefore, the effect of compatibilizer was studied for concentration of rice husk at 20%.

Effect of compatibilizer

Table III shows the effect of adding PP-g-AA as a compatibilizer as mentioned earlier. These effects are studied for only one grade of PP, namely, 11 MFI, and for one concentration (20%) only. Flexural modulus of PP filled with rice husk increased further when small amount of PP-g-AA was added. Most importantly, the impact strength also has increased. From these results, it seems that concentration of PP-g-AA at 3% seems to be adequate. Remaining properties such as tensile strength, flexural strength, MFI, HDT remained unaffected. Surprisingly, elongation at break reduced further when PP-g-AA was incorporated. Addition of compatibilizer, PP-g-AA, reduced the water absorption value of the sample. This may be due to increase in interfacial interaction between the PP matrix and rice husk.

TABLE II
Properties of PP Filled with Untreated Rice Husk

Samples	Tensile property		Flexural property					
	Tensile strength (MPa)	Elongation at break (%)	Flexural strength (MPa)	Flexural modulus (MPa)	Impact strength (J/m)	MFI (g/10 min)	HDT (°C)	Vicat softening point (°C)
11 MFI PP	32	52	32	1066	34	11.0	64	160
5% RH	32	31	27	945	30	11.5	66	160
10% RH	31	27	28	1225	32	11.7	68	160
15% RH	29	23	33	1497	31	10.7	72	160
20% RH	29	23	37	1665	30	10.5	72	162
30% RH	27	23	31	1728	30	9.5	73	162
3 MFI PP	37	101	36	1091	53	3.0	63	163
3 MFI PP + 10% RH	31	44	28	1134	36	3.7	65	158
3 MFI PP + 20% RH	27	21	32	1632	32	3.2	66	156

TABLE III
Properties of Compatibilized Rice Husk Filled PP (11 MFI)

Samples	Tensile property		Flexural property			MFI (g/10 min)	HDT (°C)	Vicat softening point (°C)	Water absorption value (%)
	Tensile strength (MPa)	Elongation at break (%)	Flexural strength (MPa)	Flexural modulus (MPa)	Impact strength (J/m)				
Virgin PP	32	52	32	1066	34	11.0	64	160	0.010
20% RH	29	23	37	1665	30	10.5	72	162	0.448
20% RH + 1% PP-g-AA	31	16	37	1745	34	10.5	71	160	0.290
20% RH + 3% PP-g-AA	32	14	42	1966	34	10.5	72	161	0.132
20% RH + 5% PP-g-AA	33	14	36	1907	34	10.5	70	160	0.129

Effect of chemical treatment

The increase in flexural modulus is normally observed when hard inorganic fillers are employed. Rice husk contains silica in substantial proportions. The effective concentration of silica in rice husk can be enhanced by removing low molecular weight cellulosic components either by acid or alkali treatment. As alkali treatment will remove lignin also, rice husk was treated separately with acid and alkali. Addition of silica to PP is known to increase flexural modulus. Considering that rice husk contains ~ 25% hemicellulose, removing hemicellulose by acid treatment can increase its silica content considerably. Therefore, one may use lesser quantity of treated rice husk to maintain similar level of silica. Since good properties are obtained by incorporating 20% untreated rice husk, one can use 10–15% treated rice husk in which hemicellulose is removed. Therefore, it was decided to incorporate 10% treated rice husk for this study.

Table IV shows the properties of PP filled with 10% rice husk, which is chemically treated in different manner. Addition of rice husk reduces tensile strength of PP substantially. So all the treated rice husk were added in lower concentration (10%) to get better understanding on effect of chemical treatment of rice husk.

Effect of acid treatment

The flexural modulus of PP has registered significant increase when rice husk was treated with acid (A10–A90). The properties of PP filled with rice husk treated for 25 min seems to give better properties when compared with rice husk treated for different times. More interestingly, effect of acid treatment has increased the percentage elongation at break. As mentioned earlier, acid treatment is likely to remove hemicellulose. The decrease in hemicellulose content will effectively increase the relative percentage of α -cellulose, lignin, and silica in rice husk. As the rice husk is used as a filler, the change of composition at the surface of the rice husk will be more important. The acid treatments for longer times are likely to affect the α -cellulose also. The decrease in the properties due to longer treatment of acid may be due to this reason. When rice husk is treated with acid and then with DMSO, almost all the properties have remained unaffected while slight enhancement of impact strength is observed. Removal of hemicellulose and part of lignin may make surface of rice husk more porous and can enhance the surface compatibility. The acid treatment may even enhance the interaction at the interface as is observed by Rozman et al.²⁴ when they used rice husk treated with glycidyl methacrylate. Increase in percentage elongation

TABLE IV
Properties of PP (11 MFI) Filled with Treated Rice Husk (10%)

Samples	Tensile property		Flexural property			MFI (g/10 min)	HDT (°C)	Vicat softening point (°C)	Water absorption value (%)
	Tensile strength (MPa)	Elongation at break (%)	Flexural strength (MPa)	Flexural modulus (MPa)	Impact strength (J/m)				
10% RH	31	27	28	1225	32	11.7	68	160	0.429
A10	30	35	32	1355	32	11.7	68	160	–
A25	28	32	31	1478	33	11.5	72	161	0.373
A45	30	21	29	1306	32	11.5	70	160	–
A90	29	23	29	1330	34	11.3	70	162	–
AL90	26	28	33	1158	34	11.5	68	159	0.425
A90AL90	26	38	25	1117	35	12.3	68	159	–
A25DMSO30	28	32	28	1450	35	12.0	72	161	–

TABLE V
Mechanical Properties of PP Filled with Rice Husk in Wet and Redried States

Samples	Tensile strength (MPa)		Elongation at break (%)		Flexural strength (MPa)		Flexural modulus (MPa)		Impact strength (J/m)	
	Wet	Redried	Wet	Redried	Wet	Redried	Wet	Redried	Wet	Redried
11 MFI PP	32	32	52	52	32	32	1066	1066	34	34
10% RH	30.5	29.8	35	32	28	27	1086	1122	32	32
A25	28.4	28.2	36	31	30	31	1301	1417	31	30
A90AL90	24.6	24.5	38	33	23	25	1050	1105	34	33
A25DMSO30	27.1	26.8	38	34	28	26	1136	1417	33	33
20% RH	28.5	27.9	27	25	34	35	1537	1609	31	30
20% RH + 3% PP-g-AA	30.5	30	18	14	35	38	1682	1683	32	31

at break may also indicate better interaction between PP matrix and rice husk powder. Water absorption studies were conducted for sample containing rice husk treated with acid for 25 min only, as properties of this composition were better than remaining compositions. It also seems that percentage of water absorption is reduced when acid-treated rice husk is used as acid treatment seems to reduce polar components, i.e., hemicellulose from rice husk. Rozman et al.²⁴ have also reported that modification of rice husk with glycidyl methacrylate reduced water absorption of their composites.

Effect of alkali treatment

PP filled with rice husk treated with alkali does not give properties as attractive as those exhibited by acid-treated rice husk. It appears that properties of PP filled with untreated rice husk are better than samples containing alkali-treated rice husk. Surprisingly, combined treatment of acid and alkali did not improve the properties very significantly. From the data reported in Table IV, it is very clear that alkali treatment does not seem to influence the water absorption characteristics significantly. The water absorption of PP filled with untreated rice husk was 0.429% while that for alkali-treated rice husk was 0.425%.

Ahmad et al.²³ have reported the increase in flexural modulus of PP composites prepared with rice husk ash, which is mainly silica. They produced rice husk ash by burning the rice husk. During such a process natural amorphous silica would undergo uncontrolled thermal changes converting it into SiO₂. The increase in flexural modulus at 20% loading reported in their work was about 20–25%. The improvement in flexural modulus at 20% loading is almost 60% in this work. In this study the low molecular weight hemicellulose is removed by acid treatment, leaving silica in its natural form. In addition, the α -cellulose which is fibrous in nature, is also present. Comparison of these results with those

of Ahmad et al. seems to indicate that presence of both silica and α -cellulose rather than silica alone may be responsible for improving flexural modulus. Also nature of silica may play significant role. Presence of lignin may not improve the flexural modulus but it may improve electrical properties as reported by Kharade and Kale for phenolic resin.²⁶

Effect of water absorption on mechanical properties of PP filled with rice husk

Table V compares the effect of water absorption and redrying on the properties of PP filled with rice husk. As expected, most mechanical properties reduced when water was absorbed by the specimen. Similar results have been reported by Ishak et al.¹⁸ It is interesting to note that elongation at break has increased in specimens which have absorbed some moisture. This phenomenon may be due to swelling of rice husk portion and possible plasticization due to water. Further study is needed in this respect. The reduction in flexural modulus was less when rice husk was treated with acid. Rice husk upon absorption of water becomes soft and hence its flexural properties decrease. Most interestingly, the properties are regained after the specimens are dried. The regain of properties can be commercially attractive.

Rheological properties

Figures 1–3 show the viscous behavior of PP filled with rice husk. For discussion, viscous behaviors of PP filled with 20% rice husk are presented here. Additional data are reported by Bera.²⁷

Virgin PP and all the filled PP specimen exhibit well-defined Newtonian viscosity in low shear rate region while shear thinning behavior in high shear region. Effect of rice husk powder on the viscosity is different for two grades of PP. Thus viscosity of filled PP of 11 MFI is slightly higher than that of base resin in the low shear rate region. However, at

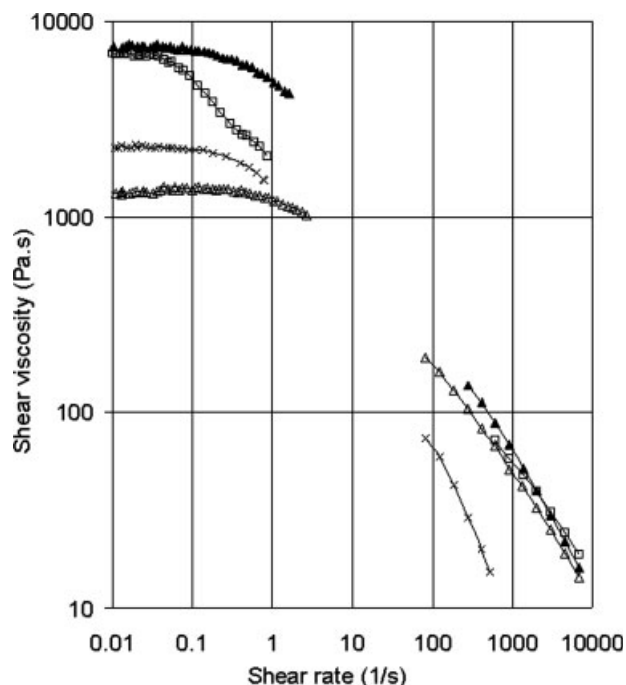


Figure 1 Flow curve for PP filled with 20% rice husk at 230°C; (▲) 3 MFI PP, (□) 20% RH + 3 MFI PP, (△) 11 MFI PP, (×) 20% RH + 11 MFI PP.

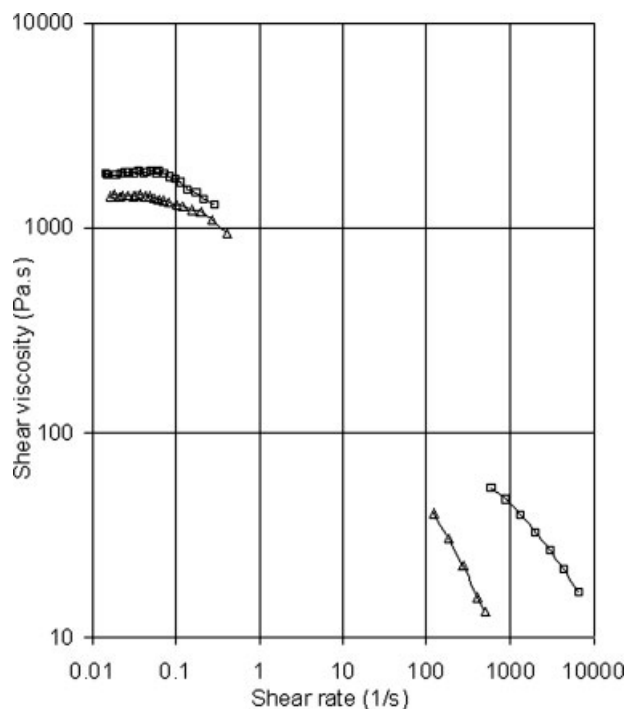


Figure 3 Flow curve for PP (11 MFI) filled with 10% rice husk (acid treated and untreated) at 230°C. (□) A25, (△) 10% RH.

high shear rate region it is substantially lower than that of base resin. When viscous behavior of filled PP of 3 MFI is considered, it shows comparable viscosity at high shear rate and also at very low shear rate with base resin. However, the onset of non-Newtonian behavior is earlier for filled PP. The

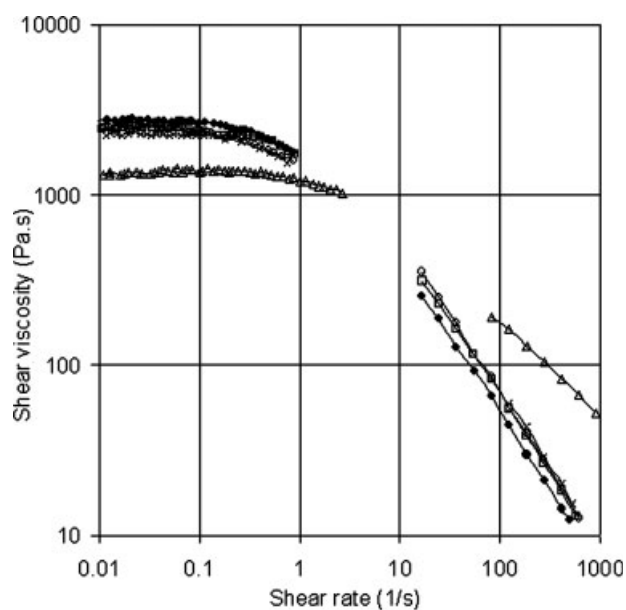


Figure 2 Flow curve for variation of PP-g-AA into PP (11 MFI) containing 20% rice husk at 230°C; (×) 20% RH, (□) 1% PP-g-AA, (●) 3% PP-g-AA, (○) 5% PP-g-AA, (△) 11 MFI PP.

lower viscosity of filled PP may indicate noncompatibility and the slippage. Slightly higher MFI values also indicate the lower viscosity. The lower viscosity can improve the processability of PP.

Figure 2 shows the effect of compatibilizer on the viscous behavior of the filled PP. It is very easy to see that viscosity of the compatibilized filled PP is slightly higher than that of noncompatibilized one and also appreciably higher than that of base resin in low shear rate region. As the viscosity of PP containing PP-g-AA at higher shear rate is comparable with that of base resin, processability of PP and filled PP appear to be comparable.

From the results shown in Figure 3, it is clear that viscosity of acid-treated rice husk filled PP is higher than the filled PP made from untreated rice husk. This suggests better interaction between rice husk and PP matrix at the interface.

TABLE VI
Thermal Properties of PP (11 MFI) Filled with 10% Rice Husk

Samples	Heating cycle			Cooling cycle		
	Onset (°C)	Peak (°C)	ΔH_m (J/g)	Onset (°C)	Peak (°C)	ΔH_c (J/g)
Virgin PP	153	166	89	116	109	-107
10% RH	155	166	72	120	116	-90
A25	160	168	61	126	121	-84

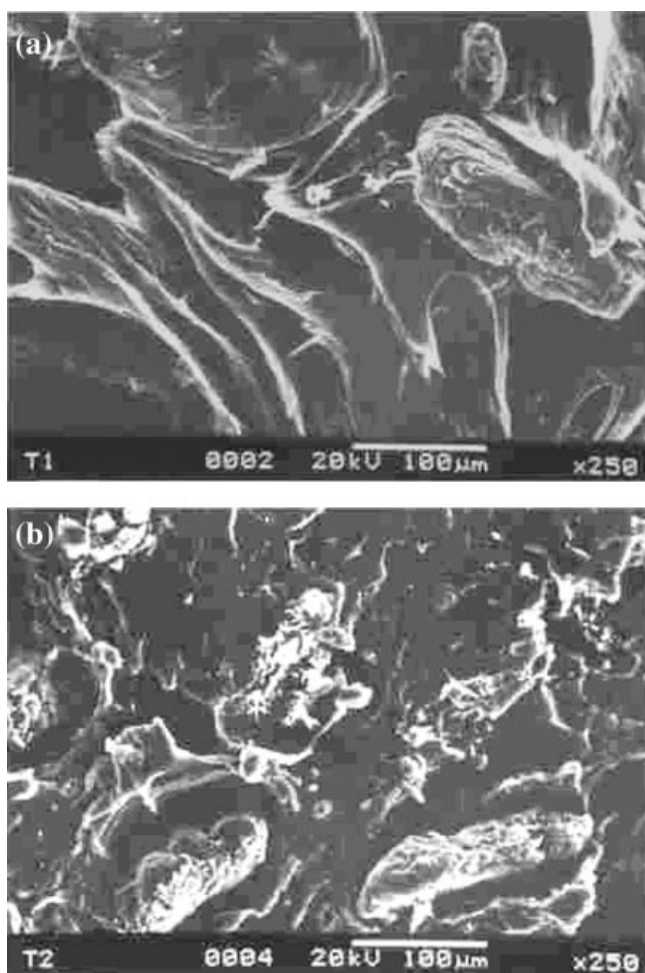


Figure 4 SEM photograph of fracture surface of (a) A25 and (b) PP filled with 10% untreated rice husk.

THERMAL ANALYSIS

DSC thermograph

Thermal properties of PP (11 MFI) filled with 10% rice husk are summarized in Table VI. It seems that addition of rice husk has decreased the degree of crystallinity as observed from enthalpy values. The cooling characteristics have shown very interesting behavior. The temperatures corresponding to onset of crystallization and peak crystallization have increased due to presence of rice husk. These temperatures have further increased due to chemical treatment of rice husk. Thus it seems that addition of rice husk is causing early crystallization of PP. This indicates that rice husk is influencing the degree of supercooling of PP. Hattotuwa et al.¹⁶ also have reported similar results.

Morphology

Figure 4(a,b) shows SEM micrographs of acid-treated and untreated rice husk filled PP, respectively. From

Figure 4, it is very clearly evident that dispersion of treated rice husk is more uniform than untreated rice husk. In addition, the treated rice husk seems to give fibrillar orientation which may be due to presence of α -cellulose at effectively higher concentration, which is fibrous in nature. Also the acid treatment removes the predominant hemicellulose. The fibrillar morphology due to α -cellulose may also be present in PP filled with untreated rice husk. However, because of the presence of hemicellulose, it may not be apparent clearly as in PP filled with treated rice husk sample.

CONCLUSIONS

The incorporation of rice husk into the PP matrix resulted in significant enhancement in flexural modulus without affecting the tensile, flexural, and impact strength. Use of PP-g-AA as a compatibilizer improved the flexural modulus further. Acid-treated rice husk further improved the flexural modulus and also reduced the water absorption. Viscosity of acid-treated rice husk filled PP was higher than that of untreated rice husk filled PP. Morphological data indicate uniform dispersion of acid-treated rice husk. The crystallization characteristics of PP filled with rice husk are modified such that early crystallization is indicated. PP filled with rice husk treated with acid absorbs lower moisture when compared with PP filled with untreated rice husk.

References

1. Leong, Y. W.; Mohd. Ishak, Z. A.; Ariffin, A. *J Appl Polym Sci* 2004, 91, 3327.
2. Diez-Gutierrez, S.; Rodriguez-Perez, M. A.; De Saja, J. A.; Velasco, J. I. *Polymer* 1999, 40, 5345.
3. Sova, M.; Pelzbauer, Z. *J Appl Polym Sci* 1989, 38, 511.
4. Zhao, R.; Huang, J.; Sun, B.; Dai, G. *J Appl Polym Sci* 2001, 82, 2719.
5. Labour, T.; Gauthier, C.; Senguela, R.; Vigier, G.; Bomal, Y.; Orange, G. *Polymer* 2001, 42, 7127.
6. Manikandan Nair, K. C.; Diwan, S. M.; Thomas, S. *J Appl Polym Sci* 1996, 60, 1483.
7. Couto, E.; Tan, I. H.; Demarquette, N.; Caraschi, J. C.; Leao, A. *Polym Eng Sci* 2002, 42, 790.
8. George, J.; Bhagawan, S. S.; Prabhakaran, N.; Thomas, S. *J Appl Polym Sci* 1995, 57, 843.
9. Uma Devi, L.; Bhagawan, S. S.; Thomas, S. *J Appl Polym Sci* 1997, 64, 1739.
10. Rozman, H. D.; Tan, K. W.; Kumar, R. N.; Abubakar, A. *Polym Int* 2001, 50, 561.
11. Oksman, K.; Clemons, C. *J Appl Polym Sci* 1998, 67, 1503.
12. Premalal, H. G. B.; Ismail, H.; Baharin, A. *Polym Test* 2002, 21, 833.
13. Real, C.; Alcalá, M. D.; Criado, M. J. *J Am Ceram Soc* 1996, 79, 2012.
14. Gorthy, P.; Pudukottah, M. G. *J Am Ceram Soc* 1999, 82, 1393.
15. Real, C.; Alcalá, M. D.; Criado, J. M. *J Am Ceram Soc* 2004, 87, 75.

16. Premalal, H. G. B.; Ismail, H.; Baharin, A. *Polym Plast Tech Eng* 2003, 42, 827.
17. Yang, H.-S.; Kim, H.-J.; Son, J.; Park, H.-J.; Lee, B.-J.; Hwang, T.-S. *Compos Struct* 2003, 63, 305.
18. Mohd. Ishak, Z. A.; Yow, B. N.; Ng, B. L.; Khalil, H. P. S. A.; Rozman, H. D. *J Appl Polym Sci* 2001, 81, 742.
19. Ismail, H.; Mega, L.; Abdul Khalil, H. P. S. *Polym Int* 2001, 50, 606.
20. Siriwardena, S.; Ismail, H.; Ishiaku, U. S. *Polym Int* 2001, 50, 707.
21. Suwanprateeb, J.; Hatthapanit, K. *J Appl Polym Sci* 2002, 86, 3013.
22. Chuayjuljit, S.; Kunsawat, C.; Potiyaraj, P. *J Appl Polym Sci* 2003, 88, 848.
23. Ahmad Faud, M. Y.; Shukor, R.; Mohd Ishak, Z. A.; Mahd Omar, A. K. *Rubber Compos Process Appl* 1994, 21, 225.
24. Rozman, H. D.; Lee, M. H.; Kumar, R. N.; Abusamah, A.; Mohd Ishak, Z. A. *J Wood Chem Technol* 2003, 20, 93.
25. Sun, L.; Xiao, M.; Xiao, P.; Song, J.; Wang, W.; Zhang, Y.; Gong, K. *Mater Res Soc Symp Proc* 2001, 661, KK 5.14/1.
26. Kharade, A. Y.; Kale, D. D. *Eur Polym J* 1998, 34, 201.
27. Bera, J. M. Tech. Thesis, UICT, Mumbai, 2004.