

COMPARISONS OF POLYPROPYLENE COMPOSITES: THE EFFECT OF COUPLING AGENT ON MECHANICAL PROPERTIES

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Abstract: The aim of this work is to compare the glass fiber (GF)/polypropylene (PP) and black rice husk (BRH)/black rice husk ash (BRHA)/polypropylene (PP) composites on mechanical properties. Tensile, flexural and falling weight impact test was conducted to investigate the effect of filler content and coupling agent (MAPP) on the mechanical properties of the BRH/BRHA/PP composites. Using a coupling agent, the mechanical properties of glass fiber reinforced material is intended to reach. By incorporating up to 10% (by weight) fillers, the tensile strength of GF/PP increased by 21%, BRH/PP and BRHA/PP were decreased by 20% and 10%, respectively, compare to neat polypropylene. Using MAPP provided to enhance the tensile strength of BRH/PP and BRHA/PP composites. And also the effect of water absorption on GF/BRH/BRHA was investigated. Results showed that increasing BRH and BRHA concentration and increasing water contact time greatly increase water absorption.

Keyword: composites, mechanical properties, FTIR, rice husk, polypropylene

Introduction

In recent years, natural filler have a potential usage for composite production. Cellulosic fibers, like wheat straw, rice husk, flax, wood in their nature, as well as, several waste cellulosic products such as husk, shell flour and wood fiber have been used as reinforcement of different plastic resins. Cellulosic material reinforced plastics, are low cost, light-weighted, have enhanced mechanical properties, and are nonhazardous (Turmanova et al., 2008; Razavi-Nouri et al., 2006; Gupta et al., 2006). Despite the advantages of natural fiber reinforced plastic composites, they have lower impact resistance, lower strength and relatively poor moisture resistance compare to synthetic fiber reinforced composites such as glass fiber reinforced plastics (GFRP) (Lee & Jang, 1999; Rozman et al., 2010). Natural fillers have hydrophilic character in their nature and this cause incompatibility between filler and matrix. Water absorption can cause degeneration of dimensional stability and micro crack can be occurred in structure. This leads to decrease of strength of composite (Arbelaiz et al., 2005; Ershad-Langroudi et al., 2008; Premalal et al., 2002; de Carvalho et al., 2012). Therefore, interface of composite should be improved by chemically or coupling agent. Coupling agent provides to enhance the bonding between filler and matrix. This causes increase of strength and reducing of water absorption value.

Recently publications give information about using of natural fillers as reinforcements in composite applications. Turmanova et al. (2008) studied water absorption and mechanical properties of polypropylene filled raw rice husk and rice husk ash. The mechanical properties like tensile strength and Young's modulus that depend on filler content were determined. They investigated the water absorption and treatment of fillers how changes the mechanical properties. Razavi-Nouri et al. (2006) studied the reinforcing effect of chopped rice husk into polypropylene. And also the effect of coupling agent MAPP was investigated. Arbelaiz et al. (2005) have reported the effects of using different coupling agent for determining the coupling effectiveness for composites. Also they compared the influence of both fiber surface and matrix modification on mechanical properties. Ershad-Langroudi et al. (2008) studied modifying the chopped rice husk reinforced PP by recycled PET. They have investigated the potential usage of rPET on composite production and how can change the mechanical/thermal properties. Two kind of form of natural filler was chosen in this study because of its special properties. Black rice husk and black rice husk ash (obtained burned rice husk). Low cost, low density, high modulus etc. are some of properties that bring about to choose these fillers. In Turkey BRH production is not much but have potential. After harvesting rice husk has no special usage as filler or something like that. Black rice husk is the outer covering of paddy and accounts for 20% of its weight (Ershad-Langroudi et al., 2008). BRHA is form of BRH that is obtained by burning the BRH. It contains higher rate of silica which is usually used as filler. Also using BRHA as reinforcement in certain polymers gives composites with better dimensional stability, toughness, as well as processing properties, and cheap process cost. This characteristic structure may not be used as waste so it should be estimated valuable technical filler.

This study aims to determine physical, mechanical and spectroscopic properties of BRH and BRHA reinforced PP composites. Therefore, tensile, bending tests, water absorption and FT-IR were carried out. The mechanical properties of the composite material obtained with natural fibers compared with neat polypropylene and glass fiber reinforced composite which is quite widely used in composite production. MAPP is used as a coupling agent on natural fiber reinforced PP to enhance the fiber-matrix interface bonding. This study was conducted to determine whether to use natural fibers instead of synthetic fibers. And also which form of rice husk more can be more effective for production compared to glass fiber reinforced composite.

Materials and method

Materials

In this study, the base resin used was S.R.L., a polypropylene homopolymer by ROM Petrol Petrochemicals with density of 0.90 g/cm³ and melt flow index of 9.36 g per 10 min (200 °C per 2.16 kg load cell). Poly-propylene-grafted maleic anhydride (PP-g-MA (Sigma Aldrich), MA content= 1% wt). Chopped glass fiber (E-glass) (PA2-4.5) was supplied by Cam Elyaf A.Ş. It has 10.5 µm diameter and 4.5 mm length (the nominal value from manufacturer's data sheet). And also it was treated by coupling agent silane 0.6%. The lignocellulosic material used as the reinforced filler in the composite was black rice husk (BRH) has been collected from Thrace region in Turkey at 2011 harvest time with moisture content (unseasoned) of 5.88 % according to AACC Method No: 44-15A. Rice husk was burnt in Protherm PLF 120/7 model ashing furnace at 600°C for 6 hours according to AACC Method No: 08-01. Ash content of 77.64% and average particle size of 500 µm were obtained (Anonymous, 2000). As it has been shown table 1, BRHs contain cellulose, hemicelluloses, lignin, waxes, and water-soluble substances (Turmanova et al., 2008).

Composites preparation

Rice husks were dried in a vacuum oven Ecocell 55 at 103 ±2 °C for 24 h to adjust the moisture content to 1–3% and then stored over desiccant before compounding. For this study moisture content decreased from 5.88% to 1.58%. Figure 1 shows drying and burning process.



Figure1. a) Drying process b) burning process

TABLE 1: Typical composition of some of natural fillers (Turmanova et al., 2008; Arbelaiz et al., 2005; Julson et al., 2009).

Natural fibers	Density (g/cm ³)	Cellulose %	Hemicelluloses %	Lignin %	Mineral Ash %	Water Soluble M. %
Flax	1,51	65-85	18-20	1-4	5	1,5
Hemp	1,47	77,5	10	3,7-13	0,8	1,8
Kenaf	1,52	45-57	21,5-23	15-19	2-5	1,9
Sisal	1,45	50-64	10-24	7-11	0,6-1	1,7
Black Rice Husk	0,09-0,15	31-34	22-26	22-23	11-14	7-9
Chemical analysis of Black Rice Husk Ash						
Composition	SiO ₂	K ₂ O	MgO	Al ₂ O ₃	CaO	Fe ₂ O ₃
%	93,19	3,84	0,87	0,78	0,74	0,58

Water causes lower adhesion between filler and matrix. Therefore, it should be removed from composite structure by drying process. Reinforced plastics granules were produced by single screw extruder (L/D= 28). The extruder has four zones with controlled temperature. The setting for these zones was: Z1 = 160°C, Z2 = 175°C, Z3 = 185°C and Z4 = 185°C. The screw velocity used was 55 rpm. Sheets of dimensions 180x180x4 mm³ were prepared using a hydraulic press under a pressure of 150 kg cm². Tensile strength, elongation at break were recorded and calculated by automatically. And elastic modulus was calculated from tensile test data by manually.

Mechanical properties

Tensile, flexural and impact are used for determining mechanical properties of reinforced plastics. In this study, both, tensile and flexural tests were performed using an Instron Universal Testing Machine Model 8501, equipped with a 500 kg load cell, strain-gauge extensometer (Instron, model 2620) after conditioning at 23 ±2 °C according to ISO 527 standard and ASTM D790, respectively. The cross-head speed used for the type IA tensile specimens was 5 mm/min. For the Flexural test (three point bending) a specimen with nominal dimensions of 80x10x4 mm³, a span of 32 mm and a cross-head of 1 mm/min were used (Franco-Herrera & Gonzalez, 2005). Gardner impact test was carried out using a Devostrans Drop Impact Test Machine according to ASTM standard D5420. For the test a specimen with nominal dimensions of 60x60x3.2 mm³, striker diameter 12.70±0.10 mm and support plate inside diameter 16.26 ±0.025 mm. Three specimens of each sample were tested for tensile and flexural tests, and the average results were reported. Gardner impact test was carried out according to Bruceton Staircase Method by 20 samples for each calculated value.

TABLE 2: Formulations of the Composites in Weight Percent

Sample ID	Ingredients (%)			
	PP	GF	BRH/MAPP	BRHA/MAPP
PP	100	0	0/0	0
PPGF10	90	10	0/0	0
PPGF20	80	20	0/0	0
PPGF30	70	30	0/0	0
PPBRH10	85	0	10/5	0
PPBRH20	75	0	20/5	0
PPBRH30	65	0	30/5	0
PPBRHA10	85	0	0	10/5
PPBRHA20	75	0	0	20/5
PPBRHA30	65	0	0	30/5

Water absorption properties

Water absorption tests were carried out according to the ASTM D 570-98 method. Composite samples were immersed in distilled water in Memmert WBN 22 model water bath at 90 °C. After that samples were dried an

oven for 24 h at $103 \pm 2^\circ\text{C}$. The dried specimens were weighed with a precision of 0.0001 g by Sartorius ED224S precision balance. The samples were removed from the distilled water, dried with blotting paper, and weight values were determined. Water absorption percent was calculated using the following formula,

$$M (\%) = \frac{M_t - M_o}{M_o} \times 100$$

Where, M_o and M_t denote the oven-dry weight and weight after time t , respectively.

Spectroscopic characterization

Fourier transform infrared (FT-IR) spectroscopy was used to detect the presence of the functional group that exists in rice husk/rice husk ash/glass fibers. The IR spectrometer (Perkin-Elmer spectrum BX, Perkin-Elmer Canada) was used for detecting of spectra of samples. FT-IR spectra of the samples were collected in the range of 4000-400 cm^{-1} with a resolution of 4 cm^{-1} .

Morphological study

SEM was used for detecting morphology of interface of composite materials. SEM micrographs of the surfaces of impact fractured specimens were taken using a ZEISS Evo® LS 10 scanning electron microscope and FEI F50 SEM. The samples were first sputter-coated with a fine layer of gold under vacuum for 60 sec.

Results and discussion

Tensile properties

Tensile strength and modulus of PPGF composites increased by about 70% and 188%, respectively, according to neat PP at 30% filler content. Gupta et al. (2006) has reported that increase of tensile strength and modulus values reached 50% according to neat PP at same filler content. The increase of tensile strength, as a result of glass fiber incorporation, can be attributed to the good stress transfer to the glass fiber by the glass fiber-matrix interface. This good stress transfer from the polymer matrix to glass fibers leads to increase in tensile strength due to the strength of glass fiber. Figure 2 shows tensile strength and modulus of composites that depends on filler contents. Figure 3 depicts one set of BRH/BRHA reinforced composites' stress-strain graphs which have been obtained from test machine software.

The tensile strength of composites that containing PPBRH decrease by increasing filler content. At 20 % filler content, value of tensile strength reached 42% decrease according to neat PP (Tensile strength, 28 MPa). Tensile modulus of PPBRH composite had a small tendency to increase, at 20% filler content obtained value of 7% increase. Turmanova et al. (2008) has reported 14% tensile strength decrease and 5% tensile modulus increase at 20% raw rice husk content. After using MAPP, tensile strength of the PPBRH increased, and the value was obtained to be lower by 20% compared to the neat PP. The tensile modulus of PPBRH changed the upward and increased by 47% compared to reinforced PP. Tensile strength of PPBRHA decreased 21% and modulus increased 20% according to neat PP, at 20% filler content. Similar results have been reported by Turmanova et al. (2008). After incorporating MAPP, the tensile strength of PPBRHA reached similar value with neat PP. The modulus of PPBRHA decreased by 38% compared to PPBRHA without MAPP.

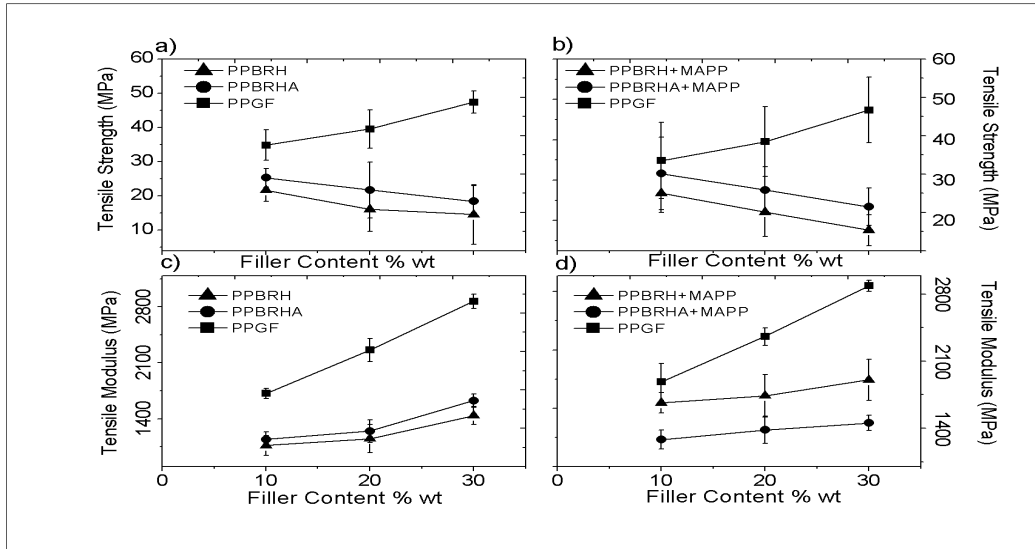
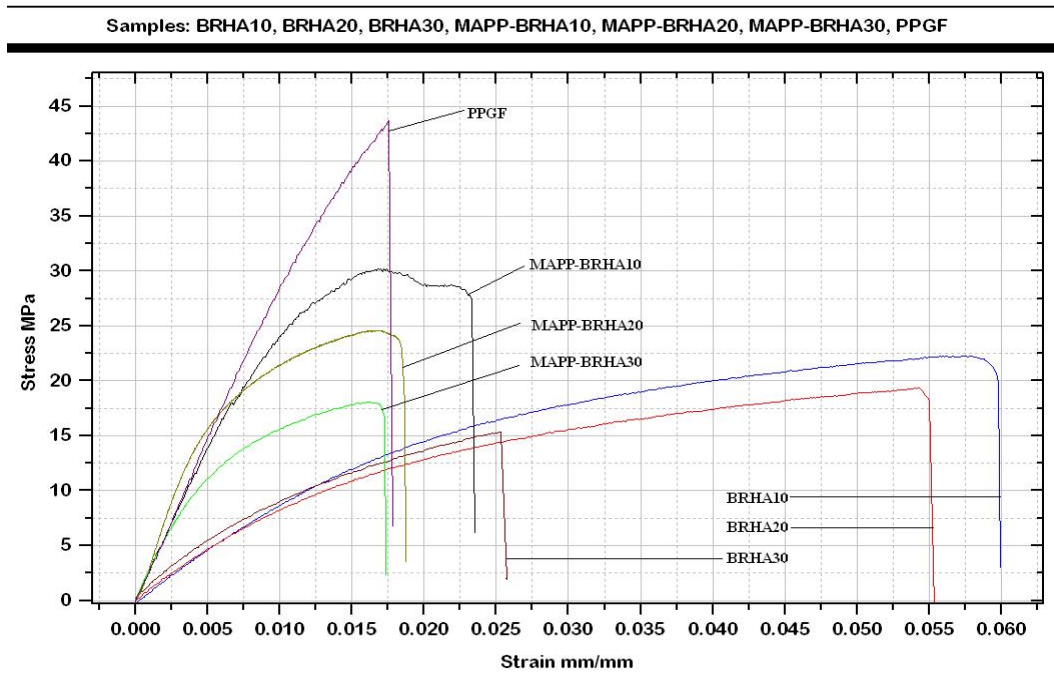
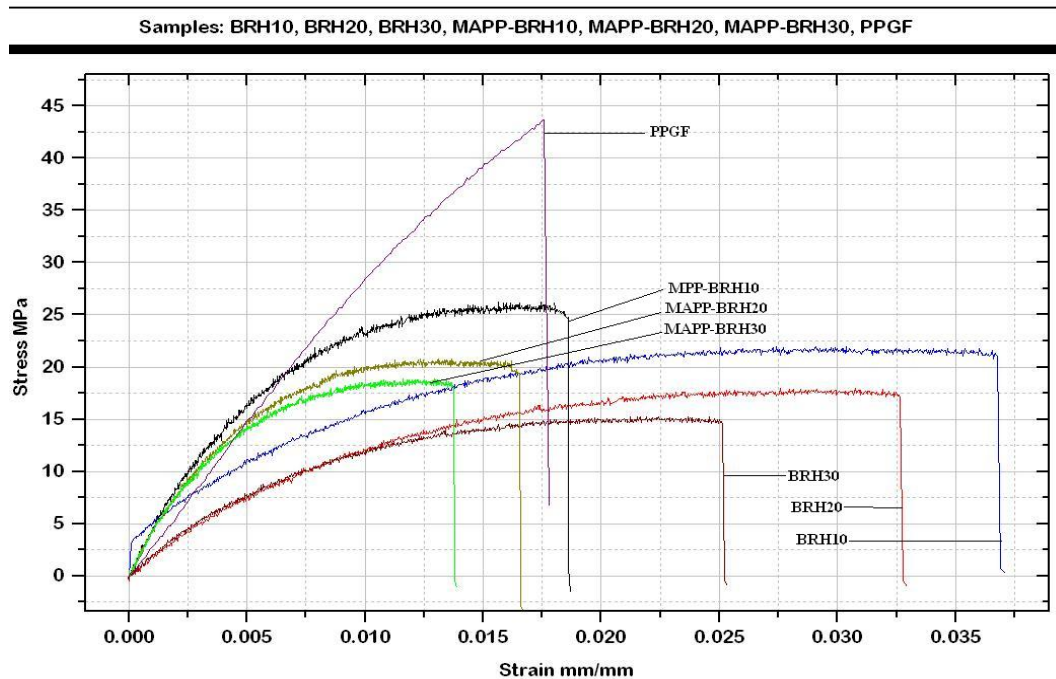


Figure 2. Tensile modulus and strength of composites a) tensile strength of composites depend on filler content, b) tensile strength of composites with %5 MAPP c) tensile modulus of composites depends on filler content d) tensile modulus of composites with %5 MAPP





PPBRH and PPBRHA composites have lower tensile strength according to neat PP and PPGF composite. This mainly attributed to restricting of stress transfer by interface of composite. Weak adhesion force between filler and matrix cause not good interface bonding. It should be noted that BRHA composites have good dispersion because of its smaller size. And this provides higher strength due to better interaction between filler and matrix compared to BRH composites.

Composites had tensile moduli which tend to increase in all composition. This mainly attributed to presence of rigid filler in their structure. Fillers restrict the free motion of matrix and deformation has been avoided. This caused increase of elastic modulus with increase filler content (Crespo et al., 2008; George et al., 2001). It is more likely the rice husk/ash acts as stress concentrators in the PP matrix. Therefore, dispersion of fillers can provide reinforcing affect due to having higher modulus than PP matrix.

Flexural Properties

It was found that although the flexural strength of BRH reinforced PP relatively remained constant, in addition to this PPBRHA increased weakly. The flexural modulus of PPBRH and PPBRHA increased by about 44% and 71%, respectively, comparing to the neat PP. Razavi-Nouri et al. (2006) has reported for BRH composites 10% and 45% increase on flexural strength and flexural modulus, respectively. And also Fuad et al. (1995) reached 40% flexural strength value at BRHA composite.

Figure 4 shows comparison of flexural strength and modulus depend on filler content of composites. PPGF composite showed the highest flexural strength and modulus by about 90% and 153%, respectively, comparison to the others. Gupta et al. (2006) has reported 63% and 258% increase for the 30% filler content. Incorporating glass fibers to polymer matrix increases the stiffness of composite. This is mainly attributed to having higher stiffness according to polymer matrix.

The MAPP is a good coupling agent that is physically stronger and has thermally stable bonds, attributed to hydrogen bonding with BRH/BRHA and chain entanglements and co-crystallization with PP (Khalil, 2008). Using MAPP enhanced the compatibility of fiber-matrix and provided to increase flexural strength and modulus. Flexural strength of BRHPP and BRHAPP increased by 10% and 29% compared to neat PP. The flexural modulus of PPBRH and PPBRHA increased by about 94% and 97%, respectively, comparing to the neat PP.

Tension, compression and shear stress occurs during the flexural loading. Failure is mainly attributed to occurring bending and shearing in bending test. Incorporating of glass fiber provides to resist the shearing of composite and this cause an increase of flexural strength (Gupta et al., 2006; Lee & Jang, 1999).

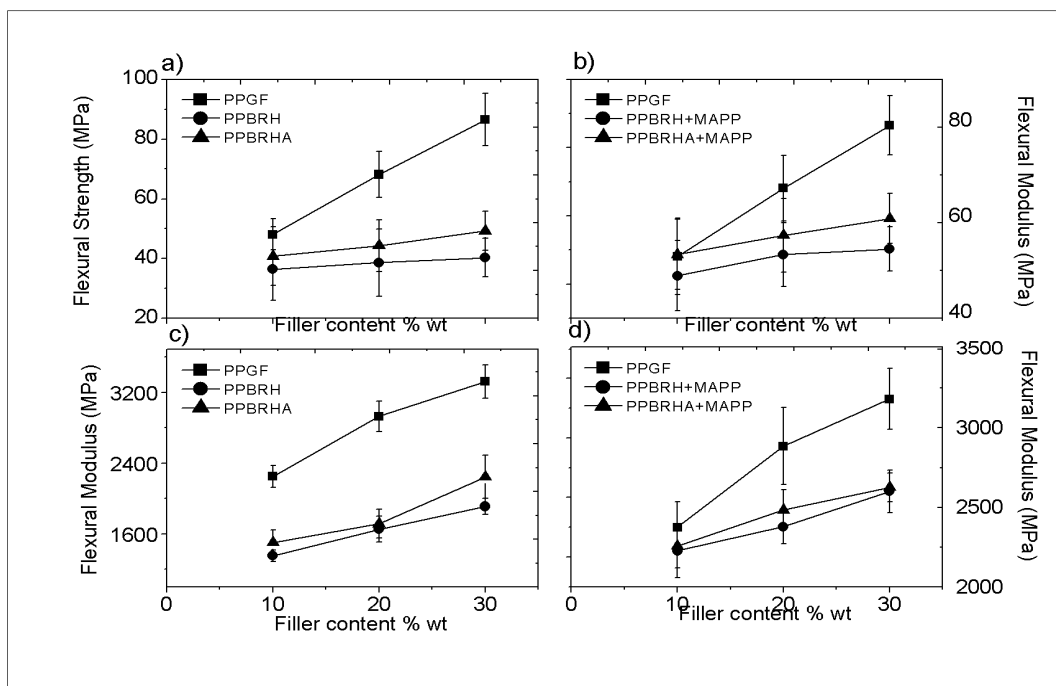


Figure 4. a) Flexural strength depends on filler content b) after using coupling agent, flexural strength-filler content c) flexural modulus without coupling agent d) flexural modulus of composites with coupling agent

Water absorption test results

PP is a non-polar polymer which has less tends to bound with water. In normal conditions, while the PP may contain 0.1% of water; at 90 degrees the water absorption value can reach 0.25 percent. The natural fillers are hydrophilic which restrict of their usage in production. The hydrophilic character of reinforcement in composites causes reduction of mechanical properties. Therefore polymers are required to undergo some operations before the additive into the matrix. Due to the high hydrophilic character of the components of natural fillers water absorption is a severe handicap for some applications of natural fiber polymer composites. Generally speaking, polypropylene hardly absorbs water due to its hydrophobic structure; however, rice husk can absorb water because of its hydrophilic character (Rozman et al., 2010; Nourbakhsh, et al., 2011; Yang, et al., 2006; Starks & Rowland, 2003; Julson et al., 2009; Jacoby et al., 2001; Thwe & Liao, 2002).

The study reports the effect of filler type and content on water absorption value of composite. Figures 5a reveals that the water absorption increases with increase of natural filler content. And also results showed that increasing BRH and BRHA concentration and increasing water contact time greatly increased water absorption, as it can be seen in Figure 5c. Figures 5b and 5d depicts that the water uptake and BRH filler content. Because of the free OH groups contained in cellulose, PP matrix composite acts as hydrophilic structure. Therefore, the water absorption increases with increasing rate of reinforcements. Similar curves were obtained by Turmanova et al. (2008). MAPP use, while ensuring the improvement of the interface between the reinforcement and the matrix, this improvement causes a lowering of water absorption. Reason behind this is considered to be a change in the diffusion mechanism of natural reinforcements. Water molecules are transferred to micro-gaps in the polymer by diffusion mechanism. In the fiber matrix interface, the water molecules are transferred by capillary action due to the lack of wetting. And also micro cracks can be occurred during production process, this cause water transportation to the gaps. Incorporating MAPP decreases the micro gaps due to enhance the bonding mechanism between filler and matrix. So water absorption value reduces by adding coupling agent.

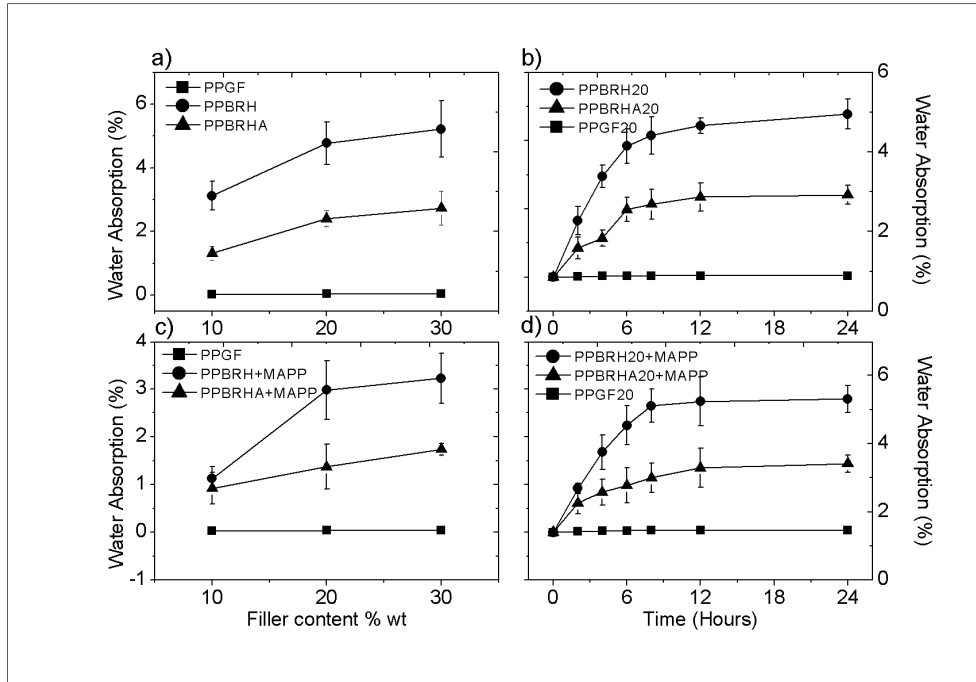


Figure 5. a) Water absorption according to filler content, b) Effect of coupling agent on water absorption, c) water absorption value at 90 °C, d) Dependences of water absorption after using coupling agent MAPP

It can be seen from Fig. 4c-d BRH20 had the highest water absorption value, PPGF20 had the lowest value. It means that the, PPGF is basically a hydrophobic polymer composite.

Gardner impact properties

Impact failures are the result of rapid crack propagation through the material. The crack’s growth rate is inversely proportional to the impact resistance of the material. For a polymer to be considered as having good impact resistance, it should be able to absorb most of the impact energy and slows the rate of crack propagation (Bigg, 1987).

Gardner test is used for determining impact energy required for crack or failure on flat surface. A striker is used for impact by drop weight. Figure 6 shows the impact tester and one of the deformed 20% PPGF sample. The procedure determines the energy (mass x gravity x height) that will cause 50% of the specimens tested to fail. Incorporation of glass fibers provide to increase of impact strength. This is mainly attributed to having higher energy absorb capacity of fibers and it cause less fiber breakage and a higher residual strength to the composite (Gupta et al., 2006). PPGF composite reached maximum impact energy value at 10% filler content according to others. Lee et al. (1999) has reported the maximum impact energy at 20% glass fiber reinforced composite. Increasing glass fiber makes the composite structure more brittle and free motion of matrix chains is restricted by fibers. During the loading, matrix cannot damp the force due to less matrix transport are between fibers. This causes decrease of impact strength of composite. Some others explained that is mainly attributed to increase in the crystallinity orientation factor of PP by GF.

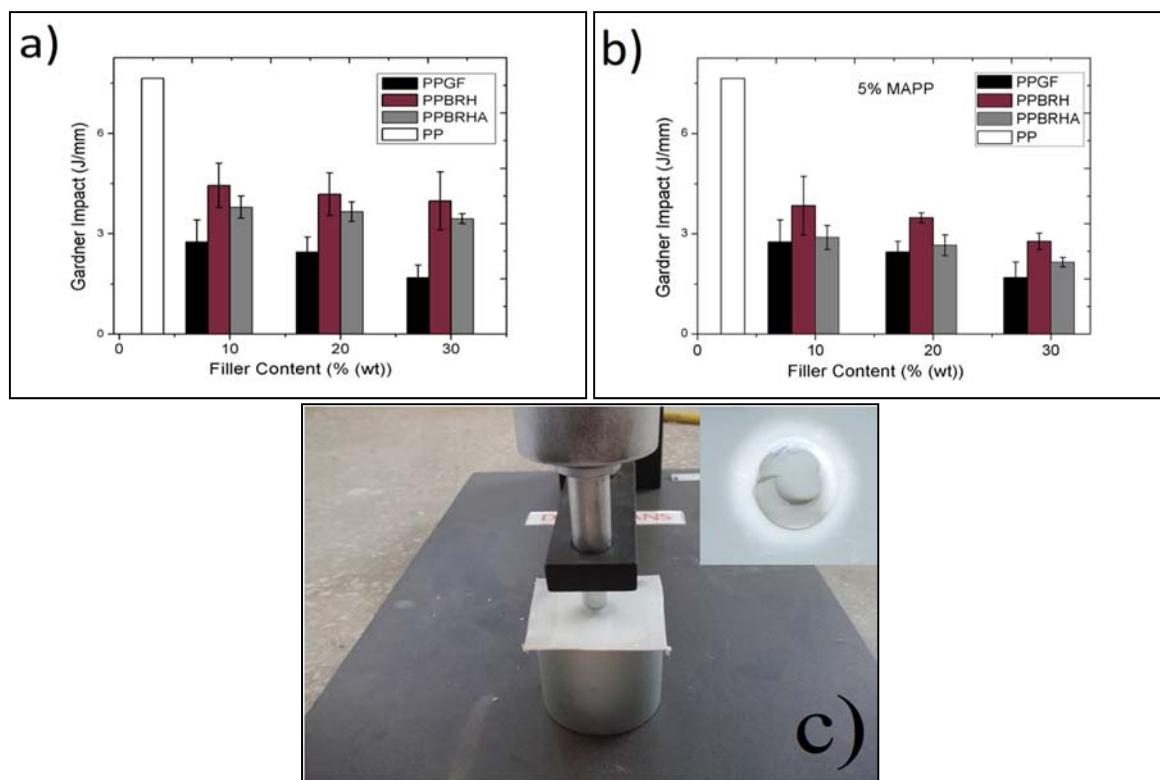


Figure 6. a) Gardner impact energy depend on filler content, b) Gardner impact energy of composites with MAPP c) test set-up

Adding BRH/BRHA filler into the polypropylene causes decrease of the impact strength. Interfacial adhesion effect the impact strength of composite. The weak bonding between filler and matrix cannot handle the energy of impact (Fuad et al., 1995). Crack moves along the weak interface. Polymer matrix cannot block the crack propagation and this cause lower impact strength. Increasing the rate of filler, increases the regions where the weak bonds and cannot prevent crack propagation.

Figure 6a) reveals that the BRH composite have higher impact strength than the BRHA composite. This may be explained higher agglomeration of BRHA that cause restriction of stress transfer to matrix. Agglomeration of the reinforcing particles has been suggested in previous studies by the effect of the adhesion forces within the composite structure (Fuad et al., 1995). BRHA particles make structure more brittle and tend to crack failure. Figure 6b) shows composites including MAPP. Adding MAPP to the structure of composite material makes the structure more brittle. Although, having stronger bond between fillers and matrix, the agglomeration of fillers is not inhibited in the composite structure. During the impact test, external mechanical energy is transferred to brittle structure of matrix and this cause a sudden damage.

Spectroscopic Results

FT-IR gives information about internal structure of composite. These curves present both PP and filler characteristic chemical bands. Figure 7 depicts that the IR spectra of filled PP. 2985-2640 cm^{-1} band give information about symmetric and asymmetric vibration of ethylene, methylene and CH groups (Turmanova et al., 2008). 1436-452 cm^{-1} have number of absorption bands (Hummel & Scholl 1968). The BRH are characterized by band between 3500 and 2700 cm^{-1} . Absorbed water and OH groups can be placed to this band. This band's position gives proof about presence of strong hydrogen bonds. 1600 cm^{-1} and 1500 cm^{-1} band give information about H_2O molecules physically adsorbed onto rice husks and C-H deformation vibrations, respectively. Siloxane bonds (Si-O-Si) bands were placed to peak at 450 cm^{-1} . Si-O network was placed to peaks between 1200 and 700 cm^{-1} . Differences between spectra of BRHA and BRH occurred at the band of 1305 and 450 cm^{-1} . This can be explained with the decrease of organic matter content and its transformation into active carbon (Turmanova et al., 2008). The bands at 1000 and 760 cm^{-1} correspond to the Si-O stretching vibration, and the bending vibration at 450 cm^{-1} appeared sharper as the organic matter was no longer present.

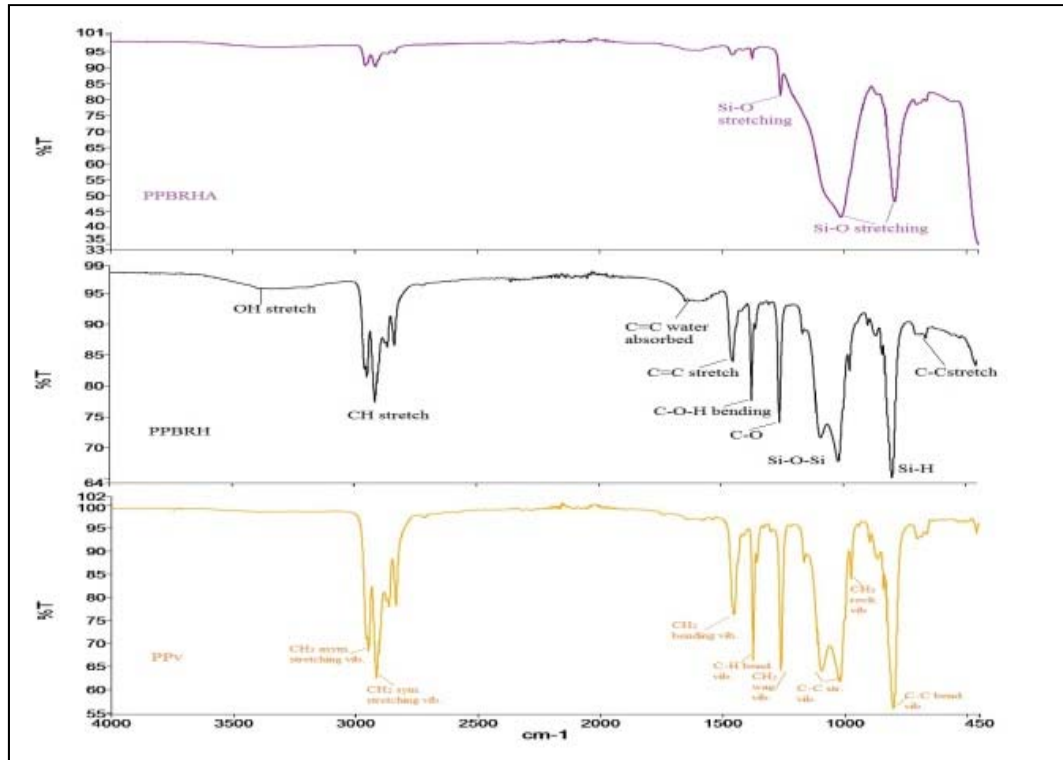


Figure 7. FTIR characterizations of composites

Morphological study of composites

Fig. 8 shows the fracture surfaces of PPGF composite after Gardner impact test. The fracture surface of matrix shows the brittle failure of PP, showing little plastic deformation resulting from fiber end (Fig. 8a). Figure 8b shows that there is a good adhesion between glass fiber and PP matrix. And also location of fibers refers a good dispersion.

Good interfacial adhesion between glass fiber and matrix enhance the stress transfer during the loading. Crack propagation is restricted by glass fiber and this cause increase of strength. Good interface between fiber and matrix also provide increase of energy absorption capacity. During impact loading, fracture propagation can be resisted by fiber-interface-matrix region (Rozman et al., 2010).

Figure 9a shows that BRHA reinforced composite have micro voids between filler and matrix. This causes poor interaction in interface. Fig. 9b reveals that presence of agglomeration on crack surface provides easy crack propagation. And also agglomeration causes lack of interfacial bonding between filler and matrix.

Figure 10a shows location of rice husk in the matrix. Figure 10b reveals that there are micro-cavities between filler and matrix due to the filler have been pulled out from polymer. These cavities result from polar-apolar incompatibility of filler-matrix. This incompatibility cause lower strength and increase of water absorption value.

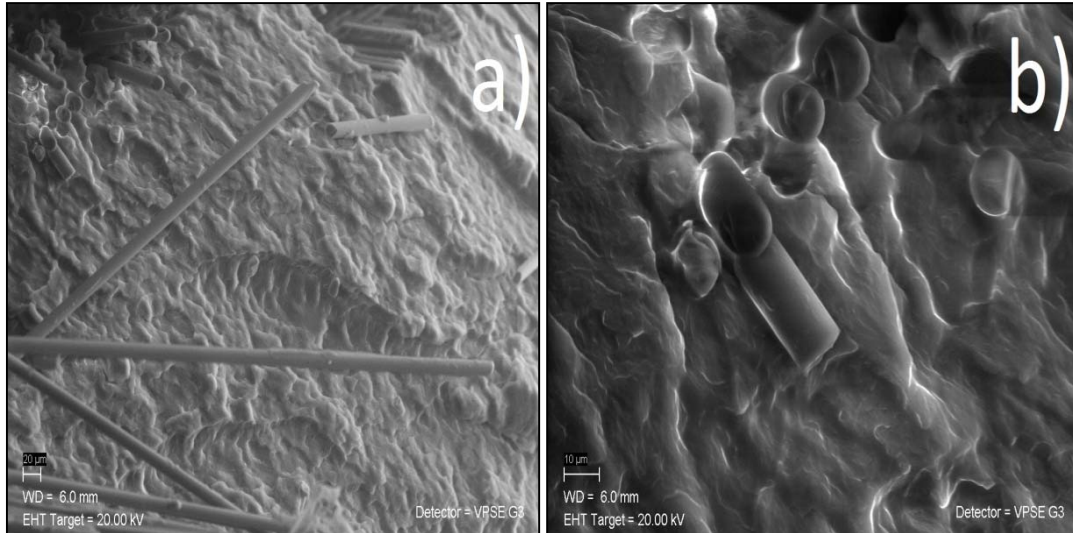


Figure 8. a) Brittle cracking of glass fiber b) Glass fiber and PP interface

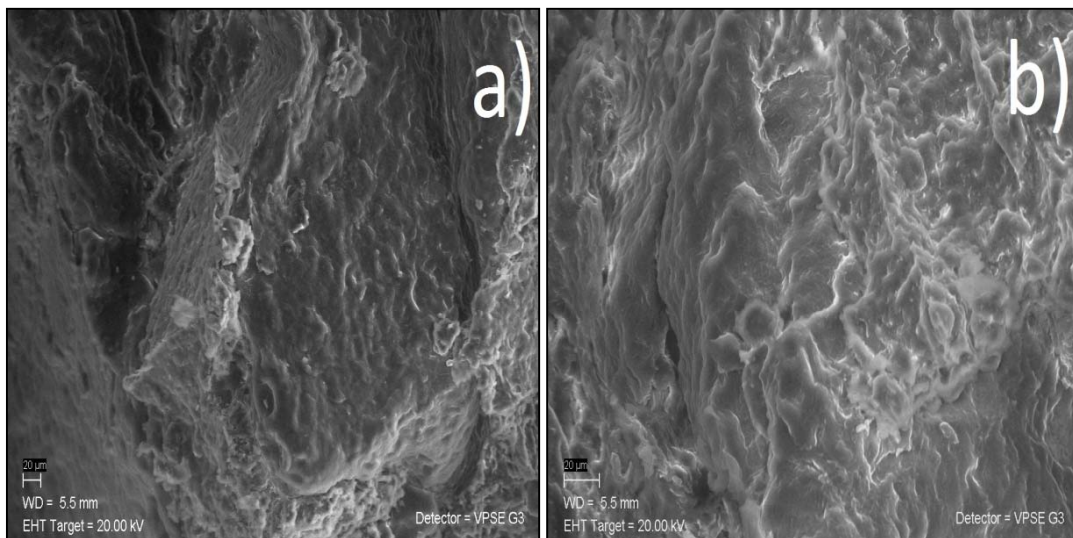


Figure 9. Failure on crack surface of composite b) BRHA agglomeration

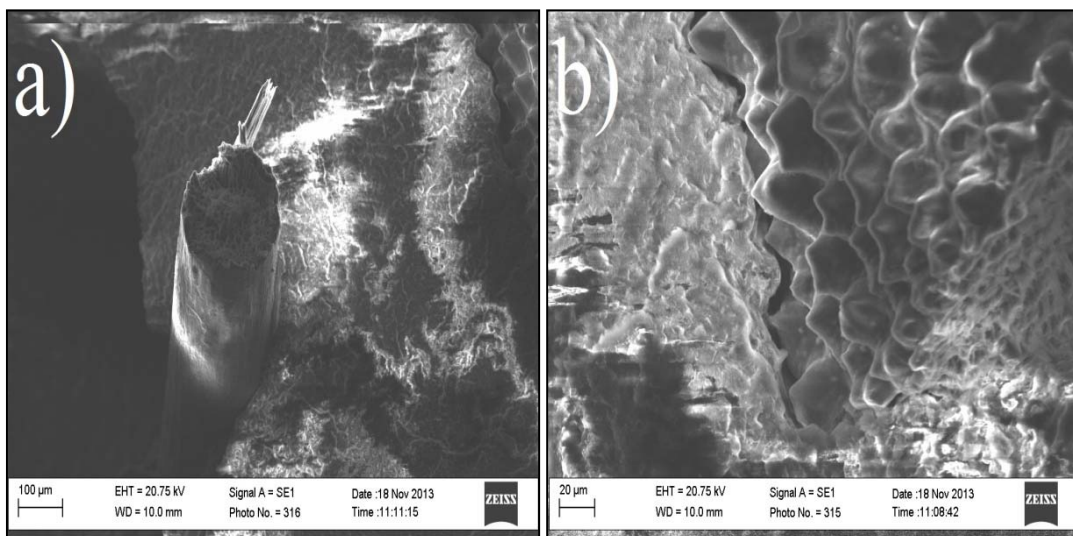


Figure 10. a) Dispersion of rice husk in the matrix b) Cavities between rice husk and matrix

Conclusion

This study reports about usage of glass fiber and rice husk/ash as reinforcement. Composite materials were prepared by incorporating of glass fiber and rice husk/ash to the PP matrix at the weight rate 10 to 30%. MAPP coupling agents have been successfully used in application as a coupling agent for various fillers. It is observed that the mechanical properties, particularly the strength of the composites are increased when MAPP is added to the system. This is because of the improved surface interaction of filler and polymer in the presence of coupling agents and the transfer of stress from one phase to the other.

The tensile and flexural strength of the glass fiber reinforced composite increased up to 30% fiber content but BRH and BRHA composite's strength decreased by filler content. The tensile and flexural modulus of composites increased with filler content. Increase rate of modulus was occurred in composite of PPGF, PPBRHA and PPBRH, respectively. BRH (10% wt) reinforced PP composite had the maximum impact energy value. Energy value decreased with filler content. The highest water absorption values were obtained in PP composites reinforced by BRH, BRHA and the lowest in glass fiber reinforced composite. Water absorption increased with the increased filler content in BRH and BRHA reinforced composites.

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