Properties of Rubber Seed Shell Flour-Filled Polypropylene Composites: The Effect of Poly(ethylene co-Acrylic Acid)

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The effect of adding poly(ethylene co-acrylic acid) (PE-co-AA) in rubber seed shell flour (RSSF)-filled polypropylene (PP) composites on processing, mechanical, water absorption, and morphological properties was studied. The addition of PE-co-AA increased the processing torque in PP/RSSF composites. Furthermore, mechanical properties such as tensile strength, flexural strength impact strength, Young's modulus, and flexural modulus show significant improvement compared to uncompatibilized composites, while elongation at break is reduced. The scanning electron microscopy (SEM) of tensile fracture specimens revealed that the dispersion of RSSF filler in the matrix improved with PE-co-AA, with fewer agglomerations. Water absorption ability of the compatibilized composites was reduced due to the improved interfacial bonding, which limits the amount of water molecules to be absorbed. PE-co-AA acted as a compatibilizer in PP/RSSF composites. Improvement in the properties of the composites was contributed by homogenization of RSSF particles in the PP matrix and reduction of voids and cracks due to improved interfacial adhesion and bonding between matrix and filler.

Keywords: Rubber seed shell flour; Polypropylene; Polyethylene copolymerized acrylic acid

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INTRODUCTION

Serious environmental problems are threatened by the usage of petroleum-based or synthetic polymers such as polyethylene and polypropylene to accommodate humans' daily applications. During the past centuries there has been a tendency to overlook the abundance of natural resources, which could potentially serve as a more environmentally friendly source of raw material for daily-used products (Bledzki and Gassan 1999; Al-Kaabi *et al*. 2005; Suryadiansyah *et al* 2007). Somehow, the increment in the usage of natural renewable resources in recent research has been mainly due to the discovery of numerous advantages of these natural resources. Renewable resources such as kenaf, bamboo, starch, and palm fiber are able to produce reinforcement, and most importantly, degradable composites. Advantages include low density, wide availability, high specific strength and modulus, less hazardous nature, and less toxicity and these advantages provide motivations to use such resources to replace synthetic fibers such as carbon fiber, glass fiber, and mineral fillers (Thwe and Liao 2002; Premalal *et al.* 2002; Kim *et al.* 2004). Although fibers from biomass sources provide a way to conserve the environment, there is a corresponding downside in the form of lower mechanical properties, if compared to carbon and glass fibers. Nevertheless, the mechanical properties of these natural-fiber-reinforced composites have sufficient resilience and strength to withstand loads for daily used products including the packaging of food or non-consumable items, and even decorative items.

Little or no attention has been paid to rubber seed, and in particular rubber seed shell (RSS), despite the fact that numerous studies have been performed on natural filler-based composites. On rubber plantations, the main economic interest is the rubber latex, while the usefulness of the by-products such as RSS is often neglected. In previous research work, the usage of rubber tree by-products to improve wastewater treatment has been attempted. In addition, the by-products have been used as extenders in polymer-based products (Guffey and Sabbagh 2002; Li and Sain 2003; Mishra and Shimpi 2005). In fact, RSS has been carbonized to provide the reinforcing effect on polymeric composites (Ekebafe *et al.* 2010b). To the best of our knowledge, not many papers have reported on the usage of RSS in thermoplastic matrix, and in particular, polypropylene.

It is arguable that the content of natural fillers provides reinforcement in polymer composites. For example, RSS itself contains minerals including magnesium, calcium, and potassium compounds. The synergistic effect of these natural and mineral components in RSS further reinforces the composites. It is claimed that due to secondary forces such as hydrogen bonds act among filler particles in a composite structure, thus contributing to the formation of aggregates and agglomerates (Cousin *et al.* 1989; George *et al.* 2001; William *et al.* 2011). On the other hand, when water molecules are permitted to be absorbed in the composites, the result is a weakening of the mechanical properties of natural filler composites. However, cellulosic fillers can be modified and treated chemically and physically to reduce the agglomerations among filler particles. In a different research work, SEM micrographs of fracture surfaces of compatibilized lignin-based polyolefin composites have revealed improvement in interfacial bonding between the hydrophobic polyolefin and filler (Feng *et al.* 2007).

To overcome problems associated with natural filler composites, PE-co-AA was chosen as a compatibilizer in PP/RSSF composites. The effect of adding PE-co-AA in the composites on processing torque, mechanical properties, thermal properties, and water absorption properties were investigated in this research work.

EXPERIMENTAL

Materials

Rubber seed was obtained from local sources in Kelantan, Malaysia. The rubber seed shell was separated from its kernel and dried in the oven for 1 hour at 100 ^{o}C to remove bound moisture before being ground into flour. A high-speed grinding machine was used to obtain the fine particle size of RSSF. RSSF was then sieved, and the granule size was measured by using a Malvern Mastersizer and found to have an average particle size of 100 μm. The composition of RSSF is tabulated in Table 1 (Ekebafe *et al.* 2010a).

PP grade Propelinas 200D with melting temperature of 165 ^{o}C and MFI of 11 g/10mins was obtained from Titan Malaysia Sdn. Bhd. Poly(ethylene-co-acrylic acid) (CAS No. 9010-77-9) with the viscosity of 600 cP was obtained from Sigma-Aldrich, Penang, Malaysia.

The formulations of PP/RSSF composites are shown in Table 2. Weights of the compounding ingredients for uncompatibilized and compatibilized composites are shown in Tables 3 and 4, respectively. From Table 4, the weight of PE-co-AA was reduced with RSSF loading. The amount of PE-co-AA was fixed at 3 php at all loadings to obtain and compare standardize results.

Methods

Sample preparation

The RSSF was dried in an oven for 1 hour at 100 ^{o}C to remove any bound moisture. RSSF then was mixed with PP in an Internal Mixer (Haake Thermomix) (Lab-scale) at 180 ^{o}C and 160 ^{o}C, respectively for 10 min in a closed chamber. PP matrix was melt-blended with different amount of RSSF, ranging from 10 to 40 php with an interval of 10. In all mixing sequences, PP was charged first into the mixer for 3 min, then RSSF was charged after.

Table 1. Composition of Rubber Seed Shell Flour (RSSF)

Composition	Percentage
Ash	0.82
Lignin	2.98
Hemicellulose	24.56
Cellulose	71.64

Table 2. Formulation of RSSF-filled PP Composites

Materials	Composition (php)	
	Without Compatibilizer	With Compatibilizer
Polypropylene (PP)	100	100
Poly(ethylene co-Acrylic Acid) (PE-co-AA)	-	3
Rubber Seed Shell Flour (RSSF)	0, 10, 20, 30, 40	0, 10, 20, 30, 40

Table 3. Weight of Compounding Ingredients for Uncompatibilized Composites

Compound	Weight (g)	
	Polypropylene (PP)	Rubber Seed Shell Flour (RSSF)
1	45.000	-
2	40.909	4.090
3	37.500	7.500
4	34.615	10.385
5	32.143	12.857

Table 4. Weight of Compounding Ingredients for Compatibilized Composites

Compound	Weight (g)		
	Polypropylene (PP)	Rubber Seed Shell Flour (RSSF)	Polyethylene Copolymerized Acrylic Acid (PE-co-AA)
1	45.000	-	-
2	39.820	3.982	1.194
3	36.586	7.317	1.096
4	33.830	10.150	1.0154
5	31.468	12.507	0.944

The mixing time was kept at 10 min, and the torque at the seventh minute was taken as a stabilization torque. After completion of the mixing procedures, the composites were taken out of the mixer and sheeted through a laboratory mill. Samples were then compression molded at 180 ^{o}C, which involves 4 min of pre-heating, 2 min of compressing, and subsequent cooling under pressure for another 4 mins.

Mechanical properties

Tensile properties were determined in accordance with ASTM D638-10, *i.e.*, specimens were placed between grips and pulled until they failed, on a Testometric tensometer M500. Dumbbell-shaped specimens were conditioned at ambient temperature (25±3 ^{o}C) and relative humidity (30%±2) before testing. A crosshead speed of 5 mm/min and gauge length of 50 mm was used. An average of 5 samples were tested and reported as tensile strength, Young's modulus, and elongation at break. Flexural properties reported as flexural strength and flexural modulus were tested using the same conditions and tensile machine according to ASTM D790-92, *i.e.*, a three-point bending system. The specimens' dimensions of 3 x 1.5 x 50 mm (depth x width x length) were tested in each case. The unnotched Izod impact test was carried out according to ASTM D256 by using a Zwick impact-testing model 5101. The dimensions of the specimens were 15 x 5 x 70 mm. Five specimens were tested in each case and reported as impact strength.

Water absorption test

Water uptake measurement was carried out as per ASTM D570. The samples were first dried in an oven at 70 °C for 24 h until a constant weight was attained and then dipped in the distilled water at ambient temperature. After soaking for a specific interval, the samples were then removed from the water, gently dried by wiping with a clean cloth, and immediately weighed to the nearest 0.001 g. The percentage of water absorption was calculated as follows,

$$WA\ (\%) = [(M_1 - M_0)/M_0] \times 100 \quad (1)$$

where M_0 and M_1 were the dried weight and final weight of the sample, respectively.

Scanning electron microscopy

A Zeiss Supra 35 VP, operated at 10 kV accelerating voltage, was used to perform studies on the surface morphology and fracture surface morphology of the composites. The surface of the sample was mounted on aluminum stubs and sputter coated with a thin layer of gold to avoid charging of the sample and to obtain best resolution using a Polaron SC 515 sputter coater.

RESULTS AND DISCUSSION

Processing Properties

Figure 1 shows the processing torque *vs*. time for PP/RSSF composites with the addition of PE-co-AA at varied RSSF loadings. The first significant peak torque appeared upon the charging of PP in all filler loadings, which indicates the high viscosity of PP and decreases as the amount of RSSF filler increases. The torque was observed to decrease after 3 min of processing due to the stabilization of PP after being melted and sheared. Torque value rose at the 4th minute, upon the charging of RSSF. It was observed that the torque value at the 4th minute increased with the RSSF loading. The increment in the torque value is claimed to be due to the dispersion of RSSF in PP, aided with PE-co-AA, which was mixed with PP before the charging. It has been demonstrated that the addition of 3 php of PE-co-AA further improves the dispersion between PP and RSSF (Akhbari and Bagheri 2012). As PP and RSSF were well-blended, the torque values started to decrease and stabilize due to the reduction of composites' viscosity, which ended the mixing sequences after approximately 10 min. Generally, the reduction of the viscosity of the composites will improve productivity in industries as a result of reductions in cost and production time.

Figure 2 compares the stabilization torque of compatibilized PP/RSSF composites with the default PP/RSSF composites at the end of the mixing sequence. The stabilization torque value increased as the filler loading increased due to the possible agglomerations formed in the composites at higher filler loading, which restricts the mobility of composites and results in higher resistance to the revolving rotor in the mixing chamber. Improved interaction between PP and RSSF explained the contrast of stabilization torque value in Fig. 2. Compatibilized composites introduced better interactions and dispersions between matrix and filler that made the composites stiffer and less mobile (Wang *et al.* 2004; Ismail and Supri 2008).

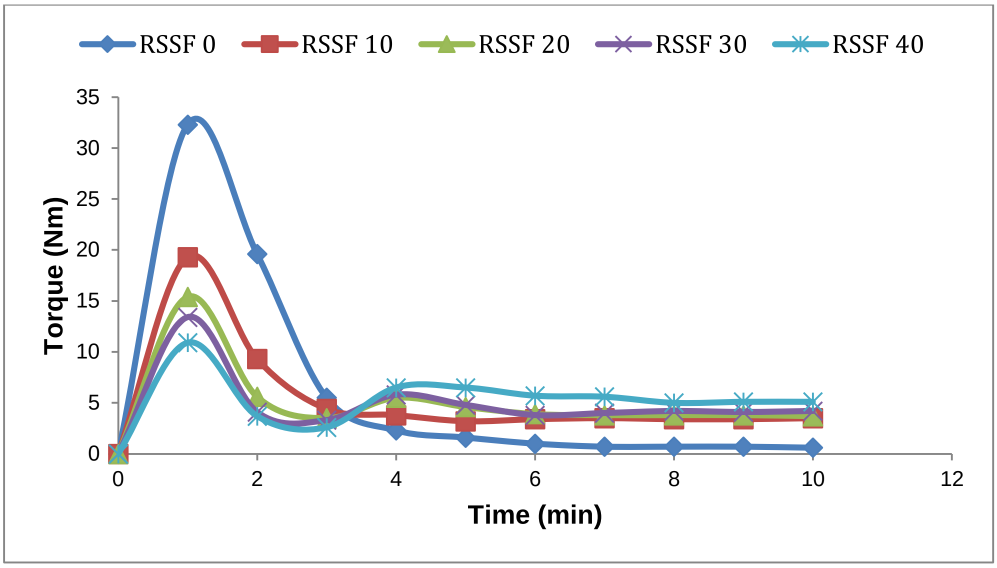

Fig. 1. Torque-time curves of compatibilized RSSF-filled PP composites at different filler loading

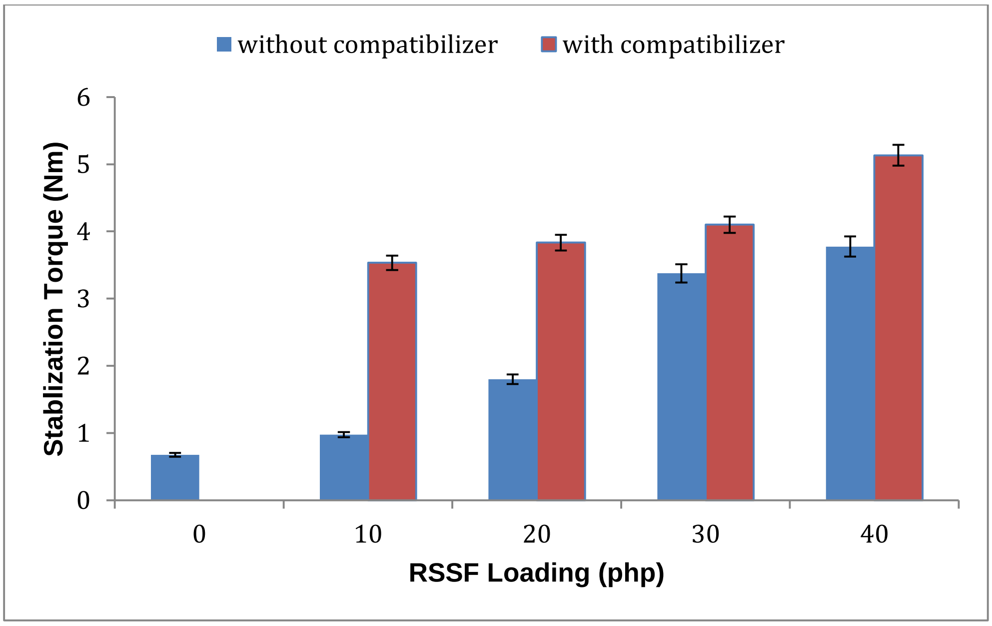

Fig. 2. Effect of filler content and compatibilizer on stabilization torque of compatibilized and uncompatibilized RSSF-filled PP composites

Mechanical Properties

Tensile properties

Figures 3 to 5 show the effect of adding PE-co-AA compatibilizer in PP/RSSF composites on the tensile strength, elongation at break, and Young's modulus, respectively. It can be clearly seen that the tensile strength was decreased with increasing RSSF loading for both uncompatibilized and compatibilized PP/RSSF composites, but compatibilized PP/RSSF composites exhibited improved tensile strength compared with uncompatibilized composites.

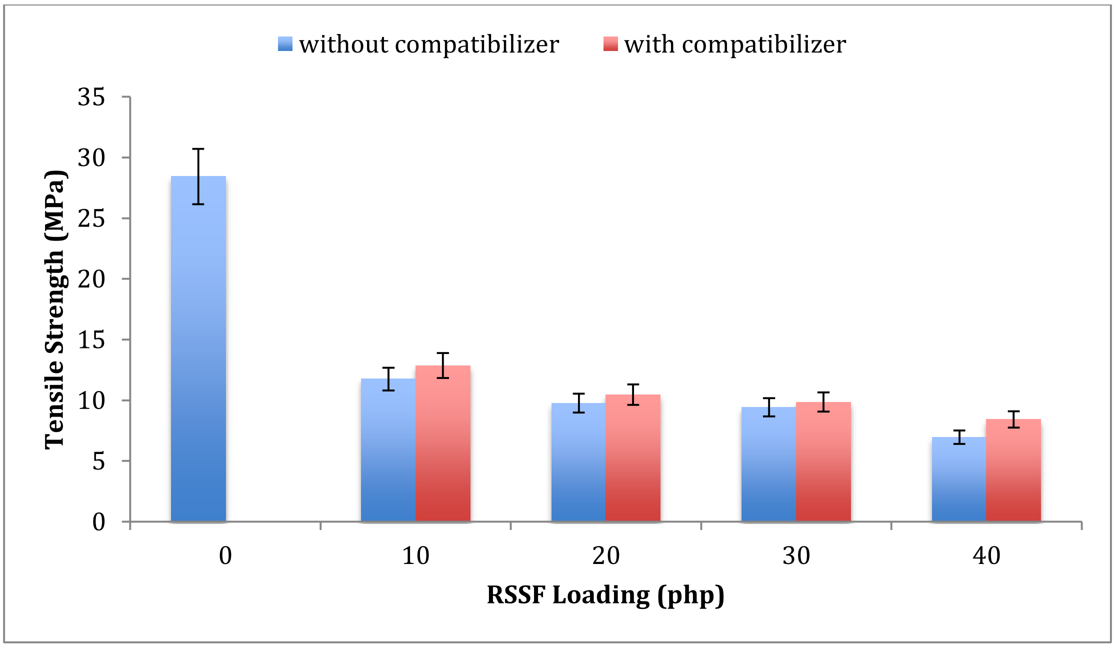

Fig. 3. Effect of filler content and compatibilizer on tensile strength for compatibilized and uncompatibilized PP/RSSF composites

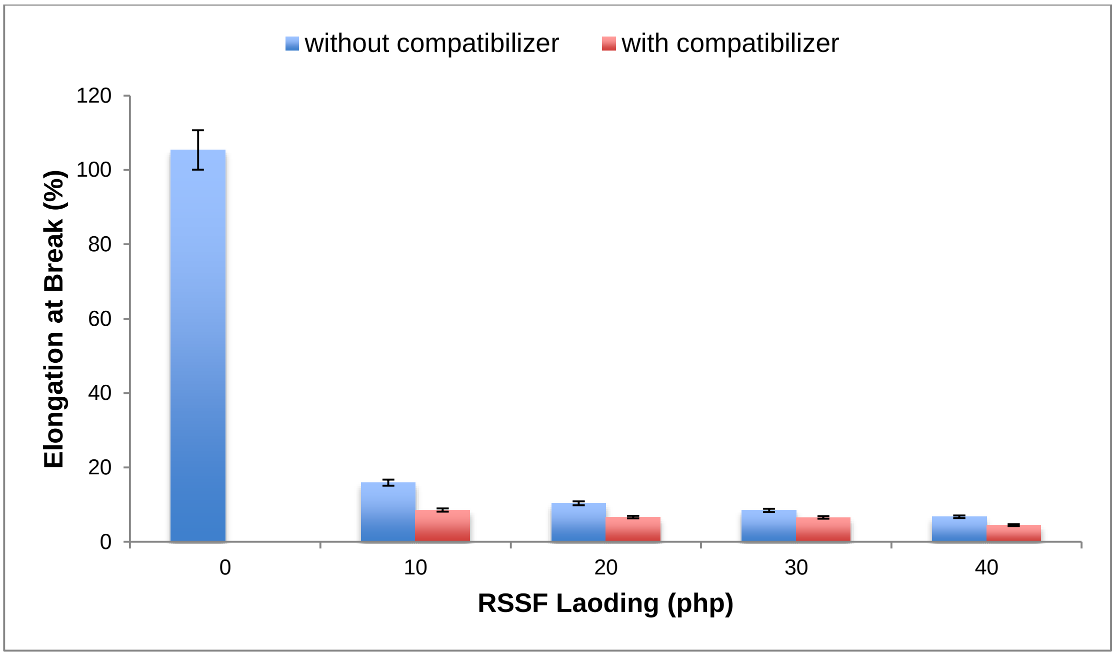

Fig. 4. Effect of filler content and compatibilizer on elongation at break for compatibilized and uncompatibilized PP/RSSF composites

The reduction of tensile strength was mainly due to reduction of matrix effective area and increased stress concentration area. As explained in most findings, the lack of interfacial bonding between the PP and RSSF weakens the strength of the composites (Rozman *et al*. 2001). When the amount of RSSF is increased, weaker interfacial bonding can be found due to the incompatibility of petroleum-based matrix and hydrophilic nature of RSSF. Incorporation of PE-co-AA in the composites improved the tensile strength by approximately 15% in most loadings. The increment is mainly due to improved dispersion and adhesion as well as interfacial bonding. Hydrogen bonding that exists in acrylic acid in PE-co-AA interacts with hydroxyl group of cellulosic content in RSSF (Table 2) (Yang *et al.* 2007). The same findings were reported when incorporating compatibilizers in polymer composites (Abdul Khalil *et al.* 2001).

Figure 4 shows the effect of adding PE-co-AA on elongation at break in PP/RSSF composites at different filler loading. The increasing amount of RSSF filler reduces elongation at break value of compatibilized and uncompatibilized PP/RSSF composites. As interaction between PP and RSSF is improved, the deformability is reduced due to the formation of a rigid interface (Abdul Majid *et al.* 2010). The hydrophilic nature of RSSF and acrylic acid improves the bonding between PP and RSSF, as the acrylic acid is copolymerized with PE first before being used as a compatibilizer in the composite (Gonzalez *et al.* 2002). The polarity (lignin and celluloses content in Table 2) of these materials makes the composite more brittle, such that it easily breaks at lower tensile stress. The ductility of the composite is reduced as the filler content increases and when compatibilizer is added (Suryadiansyah *et al.* 2008). However, compatibilized composites have lower tensile strength and elongation at break than neat PP due to shape and size irregularities of RSSF as presented in Fig. 8. From Fig. 5 it is evident that the modulus of RSSF-filled PP was increased in comparison to unfilled PP, and the modulus was also increased as the filler loading was increased and compatibilizer was added respectively. The ability of both filler and compatibilizer to enhance the stiffness of the composite explained such experimental observations. As compared to uncompatibilized composites, the tensile modulus of compatibilized PP/RSSF composites was improved by approximately 5% on average.

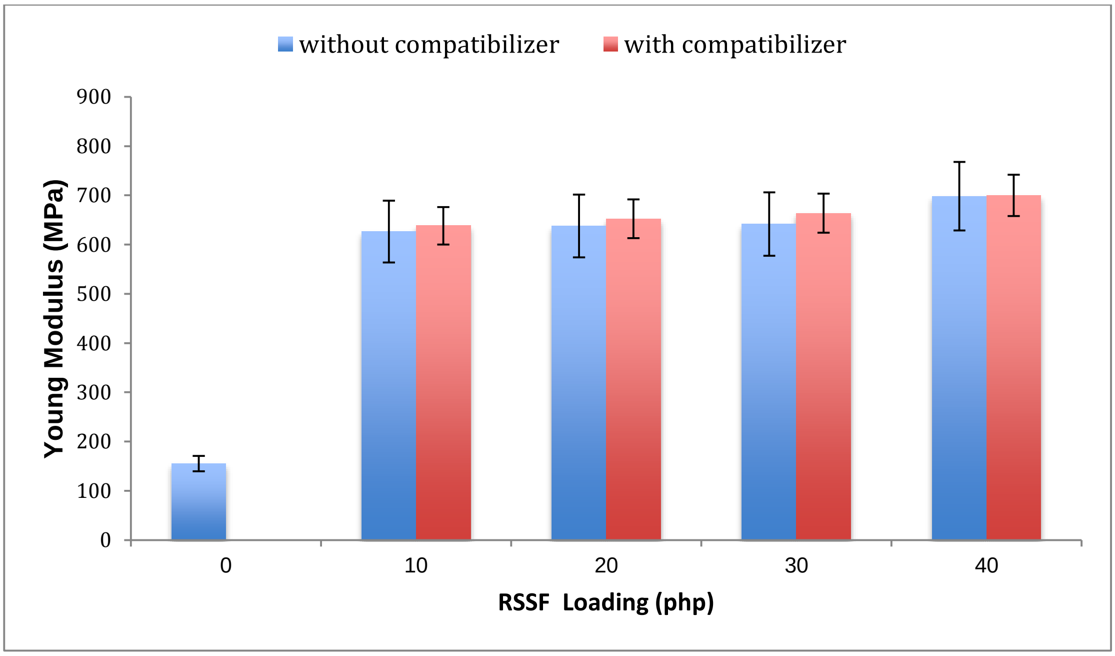

Fig. 5. Effect of filler content and compatibilizer on Young's modulus for compatibilized and uncompatibilized PP/RSSF composites

As discussed by most researchers, Onugebu and Igwe (2012) also suggested that the increase of Young's modulus is due to the stiffening effect that occurs in the composite system. The incorporation of the compatibilizer further stiffens the composite by enhancing interfacial adhesion between the matrix and RSSF particles. Interfacial adhesion prevents the RSSF particles from agglomerating and dispersing RSSF well in the matrix (Torres *et al*. 2007; Rahman *et al* 2009; Nwanonenyi *et al.* 2013).

Flexural properties

Flexural strength and modulus of uncompatibilized and compatibilized PP/RSSF composites at different RSSF loading are depicted in Figs. 6 and 7, respectively. Flexural properties of natural filled polymer composites depend solely on the type of fillers. According to Zabihzadeh (2010), who used a few types of natural filler in polymer composites in his research, poplar-filled composite exhibited the highest flexural properties, while wheat straw composite yielded the lowest value, 40.53 MPa and 36.08 MPa respectively (Yang *et al*. 2006). In the present research, the value for compatibilized RSSF composites exhibited lower flexural strength value than wheat straw composite. Somehow, the incorporation of PE-co-AA in RSSF composite increased the flexural properties, flexural strength and modulus. As discussed before, the improved interfacial adhesion leads to an increase of flexural properties when stress is imposed. The interfacial adhesion that promotes the mechanical properties of natural filled composites depends on the filler shapes and sizes. Smaller particles will reinforce more due to more coating that could possibly happen on the filler particles. Having irregular particle shapes and sizes as depicted in Fig. 8, RSSF-filled composite has lower flexural properties compared to other natural filler composites (Bose and Mahanwar 2004).

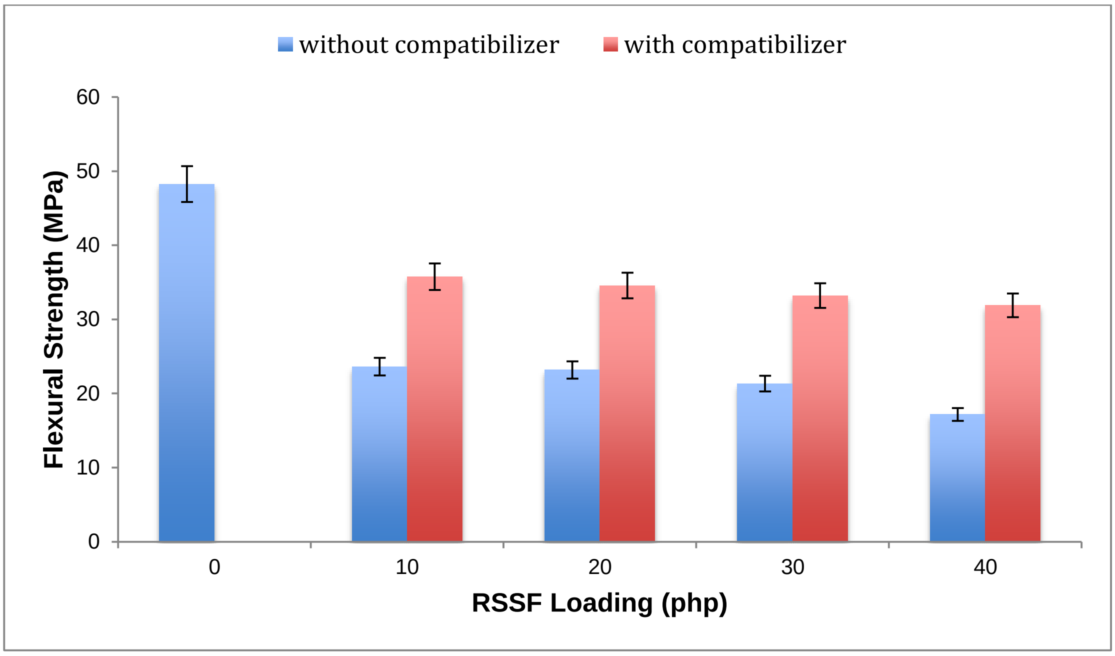

Fig. 6. Effect of filler content and compatibilizer on flexural strength for compatibilized and uncompatibilized PP/RSSF composites

Impact strength

The effect of adding PE-co-AA on unnotched impact strength of PP/RSSF composite is presented in Fig. 9. PP is well known for its ductile behavior. The impact strength of virgin PP was incomparable with filled PP, for both uncompatibilized and compatibilized composites.

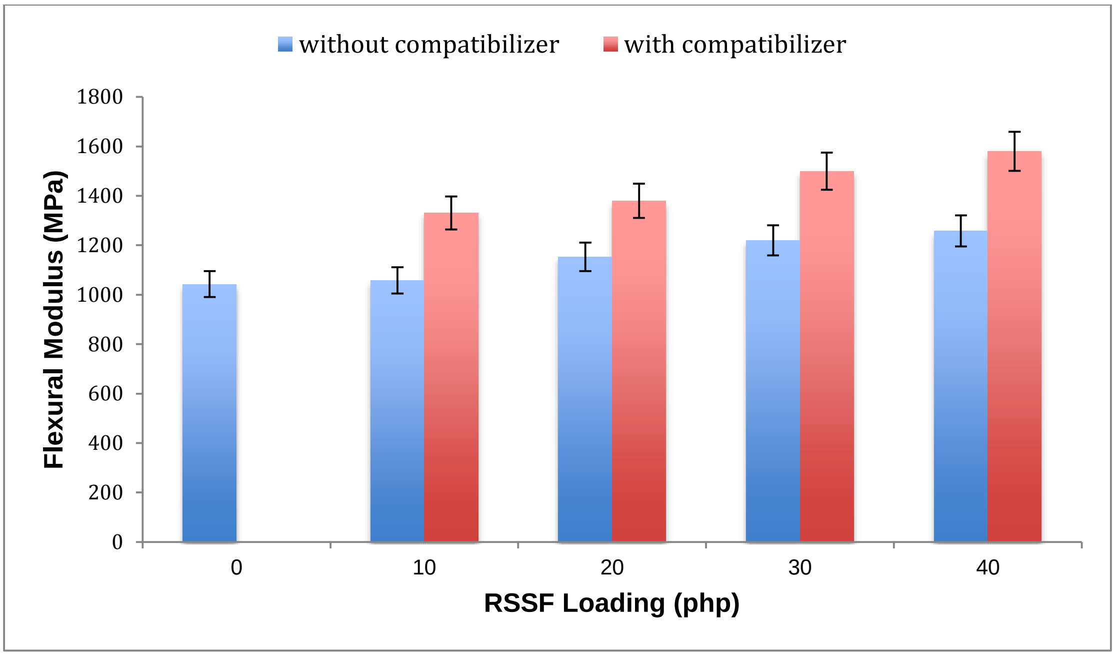

Fig. 7. Effect of filler content and compatibilizer on flexural modulus for compatibilized and uncompatibilized PP/RSSF composites

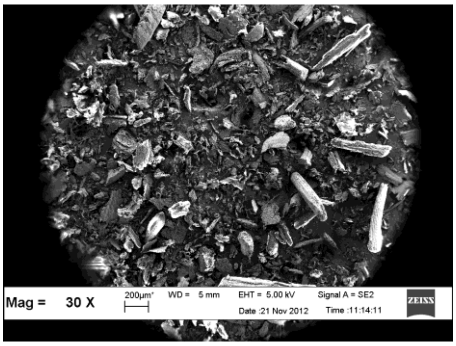

Fig. 8. SEM micrograph of RSSF

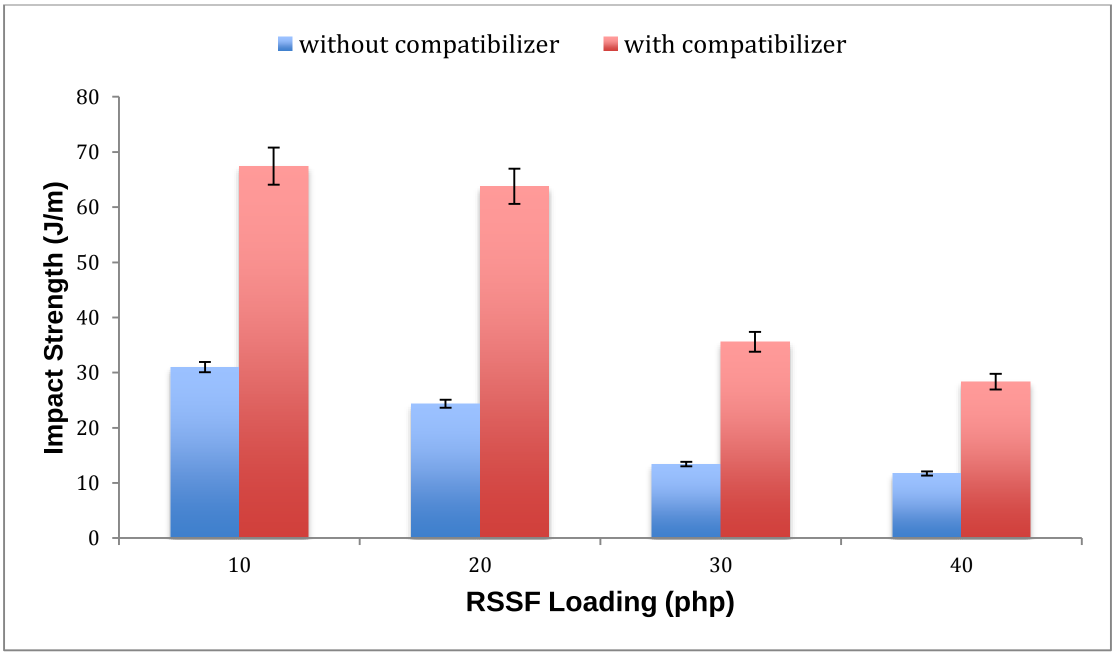

Fig. 9. Effect of filler content and compatibilizer on impact strength of RSSF-filled PP composites

However, for compatibilized PP/RSSF composite, the impact strength was remarkably increased, compared to uncompatibilized specimens, but the drop in impact strength was observed when higher amount of RSSF was incorporated. According to Karimi *et al.* (2007), who measured the impact strength of wood fiber-filled HDPE, this phenomena is commonly reported when incorporating natural fibers in polymers. Approximately 100% improvement of impact strength is reported for compatibilized PP/RSSF composite at all filler loadings. Spaces and gaps that existed between RSSF particles and PP are reduced as the improved interfacial bonding due to the ability of PE-co-AA to interact with RSSF particles and connect with PP.

Morphological Properties

Fracture surface micrographs for compatibilized and uncompatibilized PP/RSSF composites are shown in Fig. 10.

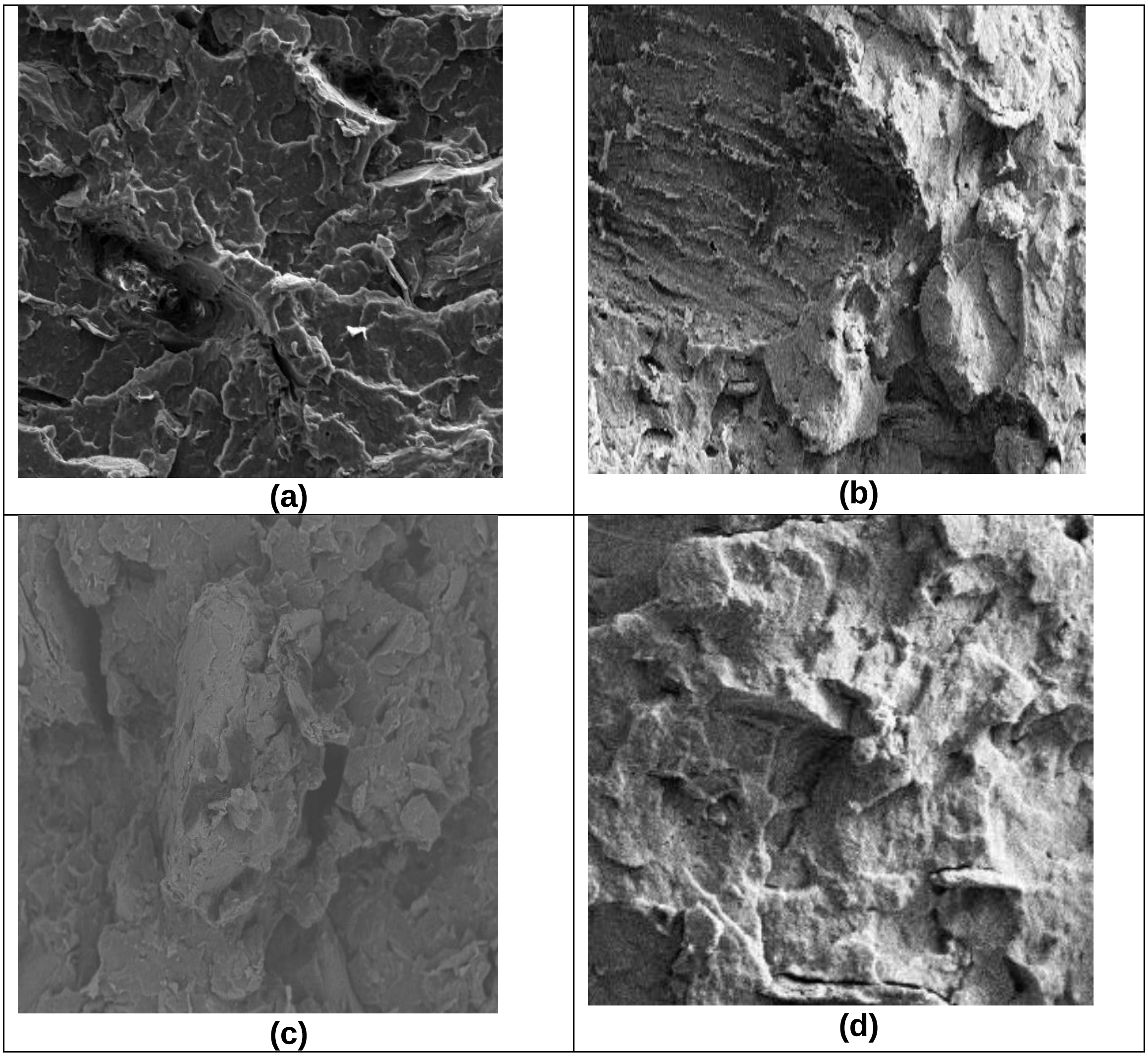

Fig. 10. Fracture surface micrograph for uncompatibilized and compatibilized RSSF-filled PP composites at different filler content

Parts a and b of the figure show the fracture surfaces of uncompatibilized and compatibilized PP/RSSF composites at 20 php filler loading, while Figs. 10c and 10d show the fracture surface of uncompatibilized and compatibilized composites at 40 php, respectively. Fracture surface micrographs of uncompatibilized PP/RSSF (Fig. 10a and 10c) spot areas in which RSSF particles were pulled out from the matrix due to insufficient adhesion between the matrix and filler. Figures 10a and 10c also show that RSSF particles did not have good interactions and dispersion with PP matrix due to different hydrophilic and hydrophobic character of RSSF and PP respectively (Abdul Majid *et al.* 2010). Natural filler such as RSSF contains massive amounts of hydroxyl groups and possibly form hydrogen bonding or ester linkages with acrylic acid in PE-co-AA, which shows an improvement in interfacial adhesion between PP and RSSF, as depicted in Figs. 10b and 10d. In fact, little filler pullouts and detachments can be spotted in Figs. 10 b and 10 d. For both uncompatibilized and compatibilized PP/RSSF composites, brittle failures occurred at higher filler loading, as presented in Figs. 10c and 10d. At higher RSSF loading, more voids can be seen. The usage of PE-co-AA in the composites tend to produce more brittle properties of the composites, as presented in Fig. 5. Young's modulus values for compatibilized composites were slightly higher than the uncompatibilized ones, and also portrayed in the fracture surface micrographs (Fig. 10b and 10d). In a previous finding, an increase of Young's modulus was found in thermotropic liquid crystalline polymer (TLCP)/polypropylene film composites when maleic and hydride-grafted SEBS compatibilizer was introduced. It is claimed that the presence of the active sites of maleic anhydride (MA) in the compatibilizer could form hydrogen bonding in TLCP and further improve the interfacial bonding (Sauvarop *et al.* 1999). The same justification can be applied for PP/RSSF composites in which the presence of acrylic acid in PE-co-AA could form secondary chemical bonding with cellulose sites in RSSF.

Water Absorption

Water absorption properties of compatibilized PP/RSSF composites are shown in Figs. 11 and 12, respectively. In Fig. 11, water absorption of compatibilized PP/RSSF composites increased up to 4% when the composites were loaded up to 40 php of RSSF on the fortieth day of the immersion, regardless the addition of PE-co-AA in the composites. Water absorption of the composites achieved saturation approximately one month after immersion. As reported in previous research findings, the usage of natural filler such as wood flour, kenaf fiber, and palm fiber in polymer composites will promote to the water absorption ability of the composites regardless the amount of compatibilizer added (Ismail and Salmah 2008). The increment of water absorption of the composites is claimed due to lignocelluloses parts that made water molecules accessible (Danjaji *et al* 2002). The lignin and cellulose content of RSSF is shown in Table 2 (Ekebafe *et al.* 2010a).

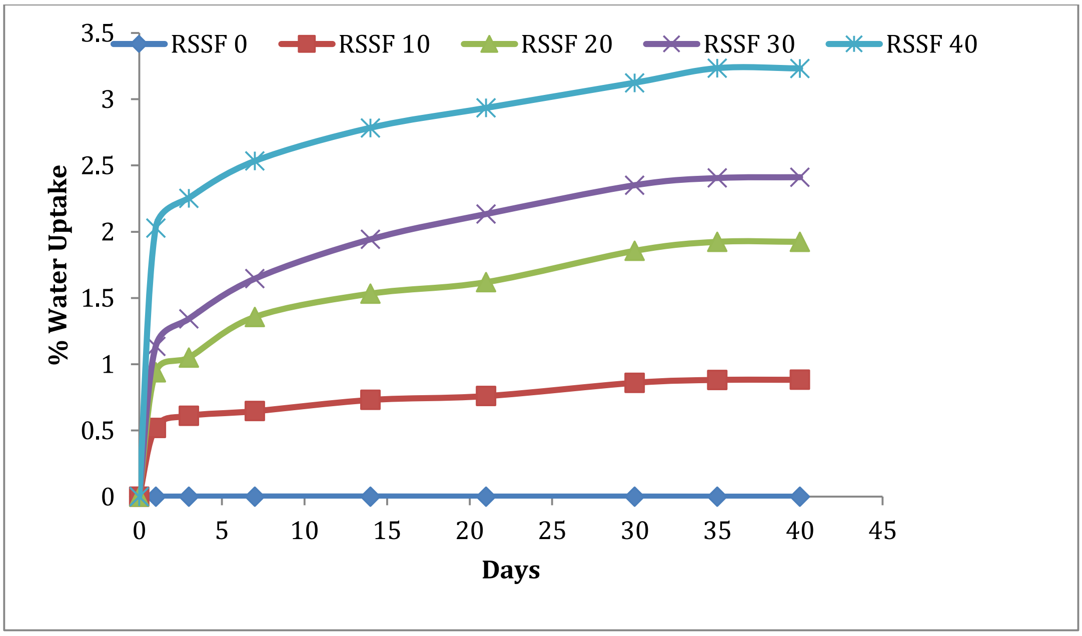

Fig. 11. Effect of filler content on water uptake of compatibilized PP/RSSF composites

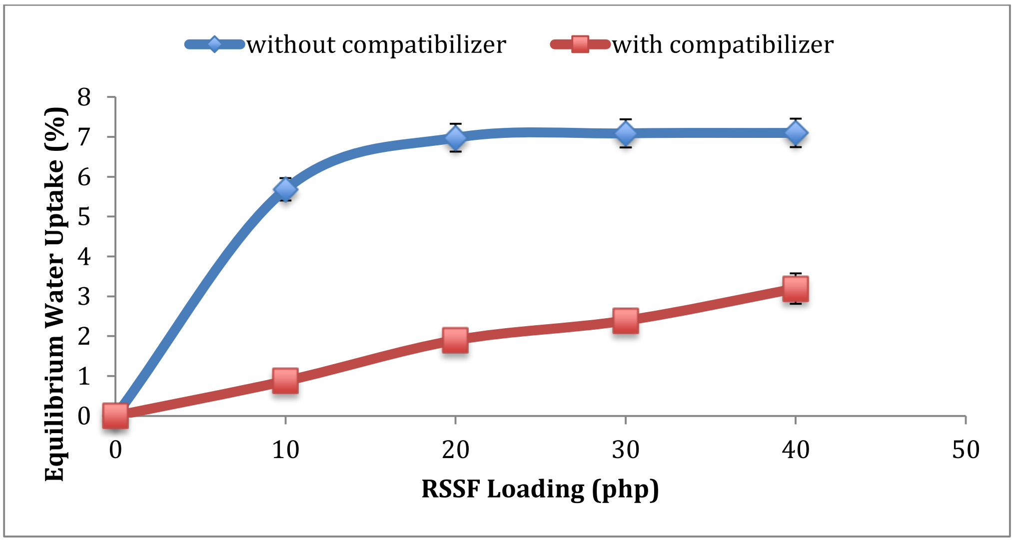

Fig. 12. Comparison on the effect of filler content on equilibrium water uptake of uncompatibilized and compatibilized PP/RSSF composites

The comparison of water uptake of uncompatibilized and compatibilized RSSF-filled composites is depicted in Fig. 12. The compatibilized PP/RSSF composites exhibited lower equilibrium water uptake than uncompatibilized composites. Almost a 50% reduction difference indicated that the incorporation of PE-co-AA reduced the voids and interfacial separation. Voids allow water molecules to access and be absorbed by the composites, assisted by massive amounts of hydrogen bonding that could possibly be formed with the water molecules (Clemons and Ibach 2004; Othman *et al* 2004).

CONCLUSIONS

1. The incorporation of PE-co-AA in PP/RSSF composites increases the stabilization torque and mechanical properties, including tensile strength, flexural strength, impact strength, tensile and flexural moduli, while it deteriorates elongation at break, compared to uncompatibilized RSSF-filled PP composites.
2. SEM Micrographs of tensile fracture surface reveal better interfacial adhesion between PP and RSSF particles and also the ability of PE-co-AA to homogenize the composites.
3. Generally, improved interfacial wetting and dispersion between the filler particles and matrices enhances mechanical properties when stress is applied.
4. At higher RSSF loading, the composite tends to absorb more water. However, the addition of PE-co-AA reduces the equilibrium of water uptake by improving the interfacial adhesion between PP and RSSF, which resulted in the reduction of voids, gaps, and interfacial spaces.

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