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Effect of Epoxidized Natural Rubber on Properties of Poly (butylene Succinate)/Rice Flour Composites

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Abstract

Composites made from poly (butylene succinate) (PBS) and rice flour (RF) were successfully prepared in this work. In order to study the effect of rice flour content and epoxidized natural rubber (ENR-50) on properties of composites, PBS/rice flour composites were prepared by varying rice flour contents as follows: 0, 1, 5, 10, 20 and 25 percent by weight with using ENR-50 as a compatibilizer (5 phr). The utilization of epoxidized natural rubber (ENR-50) as a compatibilizer of PBS/rice flour composites was also determined in this study. All materials were mixed by using a two-roll-mill at 165 °C and then processed into specimens using an injection molding machine. Test specimens were characterized phase morphology, rheological and mechanical properties, including tensile properties and impact strength by using scanning electron microscope (SEM), capillary rheometer, tensile testing machine and impact testing machine, respectively. Results indicated that tensile strength, elongation at break, and impact strength of composites were continuously decreased with increasing rice flour content while tensile modulus was increased. The maximum improvement of tensile modulus was achieved at 20 percent by weight of rice flour. This may be attributed to the stiffening effect of rice flour itself. Additionally, using ENR-50 at 5 phr as a compatibilizer of PBS/rice flour composites results in enhancement of shear viscosity, elongation at break and impact strength of the PBS/rice flour composites remarkably, due to the better interfacial adhesion between PBS and rice flour and elastomeric behavior of ENR-50.

Keywords: Poly (butylene Succinate) (PBS); Rice flour (RF); Compatibilizer; Epoxidized Natural Rubber (ENR); Composites

1. Introduction

Today, there are many environmental problems such as pollution, global warming, natural resource depletion and waste disposal as well. The over utilization of resources and creation of synthetic plastics are generating a global crisis of waste disposal. Waste from petroleum based product is one of the major problems that leading to further consumption and pollution of nature. In order to minimize this problem, biodegradable polymer, including poly(lactic acid) (PLA), poly(carpolactone) (PCL), polyhydoxyalkanoate (PHA) poly(butylene succinate) (PBS), have been produced and widely used in the market. Poly(butylene succinate) (PBS) is one of the most popular biodegradable plastic due to outstanding properties such as biodegradability, good chemical resistance and processing capabilities

[1 – 3]. Nevertheless, PBS still has some drawback because their cost are not competitive compared to other plastic materials. Additionally, other properties such as tensile strength, modulus and gas barrier properties, limit its broad application as well. Currently, composites have become an interesting topic because it gives combination of performance between matrix and filler, in order to achieve combined properties that cannot be met by a single-phase material such as high strength and stiffness, light weight, design flexibility, long shelf life and low cost. Generally, filler used in polymer composite can consist of natural and synthetic fiber and also particulate filler such as jute fiber, sisal fiber, glass fiber, rice husk flour, calcium carbonate, clay mineral and starch, etc. There are many works relating to polymer composites. In year 2013, PBS filled with cotton fiber was successfully investigated by Buenaventurada P. Calabia and co-worker [1]. They reported that the tensile strength of composites was improved by up to 78% with the addition of cotton fiber. Thermal behavior, dynamic mechanical properties and rheological properties of PBS/nano-$CaCO_3$ composites were reported by Rong-yuan Chen *et al.* [2]. The addition of nanometer calcium carbonate to PBS matrix tended to improve thermal stability and dynamic mechanical properties.

Rice is a major agricultural product and extensively cultivated in Thailand. Rice flour (RF), a form of powder prepared from finely milled rice and sustainable, biodegradable, inexpensive and renewable source of biopolymer, is a strong applicant to be used as filler for polymer composites. However, polymer composites are incompatible with each other. The limitation especially concerning the difference hydrophilicity between filler and polymer matrix was established. The agglomeration of filler into polymer matrix could be found because of the lack of adhesion between the phases. Thus, the dispersion and distribution of filler play an important role to the properties of composite. The introduction of such specific material can improve compatibility and may promote miscibility of polymer matrix and filler. This material is usually called “compatibilizer”. The best known effect from compatibilizer is the reduction of the interfacial tension and increasing the adhesion between the phases. Evidence from open literature suggests that the properties can be controlled by introducing the compatiblizer or coupling agent to the blend or composite system such as polyethylene-graft-maleic anhydride (PE-g-MA), polypropylene-graft-maleic anhydride (PP-g-MA), silane coupling agent and epoxidized natural rubber (ENR) [4 – 12]. Epoxidized natural rubber (ENR) is achieved from natural rubber by replacing some of the double bonds with epoxide units randomly with 25% or 50% referred to as ENR-50 or ENR-25. There are many advantages of ENR, including oil resistance, excellent damping property, and environmental friendly and improved compatibility with polar polymer also. The utilization of ENR-50 is a particular attractive way of compatibilizing polymer matrix with filler because the polar group of polymer matrix and filler may interact with the polar group of ENR during melt blending. The effect of ENR-50 as a compatibilizer on properties of linear-low-density polyethylene (LLDPE)/soya powder blends was investigated by S.T. Sam *et al*. They reported that the improvement in tensile strength and elongation at break was observed by the addition of ENR-50 because the better interfacial adhesion between LLDPE and soya powder [8]. Therefore, the aim of this work was to study the effect of ENR-50 as a compatibilizer on properties for poly (butylene succinate) (PBS)/rice flour (RF) composites.

2. Materials and methods

Materials

Poly (butylene succinate) (PBS: BioPBS™ grade FZ71PM) supplied by PTT MCC BIOCHEM Co., LTD. Rice Flour (RF) was collected from a local factory in Thailand and screened by laboratory sieves (Mesh 80). Epoxidized Natural Rubber (ENR-50); supplied by Muang Mai Guthrie Public Co., Ltd. All materials were used as recieved.

Compounding, Specimen Preparation and Characterizations

In order to improve the compatibility between PBS (Matrix) and rice flour (Filler), Epoxidized Natural Rubber (ENR-50) was used as a compatiblizer. Composites, prepared from PBS and various amount of rice flour (RF) and using 5 phr of ENR-50 as a compatibilizer. The content of rice flour was used at various amounts of 0, 1, 5, 10, 20 and 25 wt% of composites. Prior to compounding, all PBS and RF were dried at 60 °C for 24 hours. The materials were blended in a two-roll-mill at 165 °C for 10 mins. Composites were dried at ambient temperature, granulated and then shaped into test specimens using an injection molding machine (Boy 22M) at 165 °C. Test specimens were used for phase morphological characterization by field emission electron microscope (FE-SEM 7610F), JEOL Ltd. and rheological testing by CEAST Rheologic 5000 twin-bore capillary rheometer. The tensile properties of the obtained composites were measured by using an Instron Universal Testing Machine at room temperature according to ASTM D638. Izod impact strength was measured using a CEAST Impact tester according to ASTM D256 test procedure method with a 2.70 J pendulum.

3. Results and Discussion

Poly (butylene succinate)/rice flour composites with varying amount of rice flour (RF) from 0 to 25 percent by weight (wt%) were successfully prepared in this study. The rheological properties of the PBS/RF composites are illustrated in Fig. 1. Results indicated that shear viscosity of pure PBS and PBS/RF composites decreased with increasing shear rate, which demonstrates that a pseudoplastic behavior of the materials was observed [13]. Moreover, according to Fig. 1, it could be observed that shear stress and viscosity value at a given shear rate of composites were increased with increasing rice flour loading from 0 to 25 wt% because rice flour restrict the flow of polymer melt. This indicated that the effective of rice flour as a filler lead to increase tensile modulus of the resulting composites. Fig. 2 showed the SEM micrographs of PBS/RF composite with different rice flour content. From SEM micrographs, it was revealed that PBS/RF composites showed similar phase morphology to pure PBS. Even though the overall phase morphology of PBS/RF composites are relatively similar, the roughness of the surface differed, depending on the amount of rice flour added in the composite which affected the properties of the resulting materials. This could be due to the incompatibility between matrix (PBS) and filler (rice flour) as well.

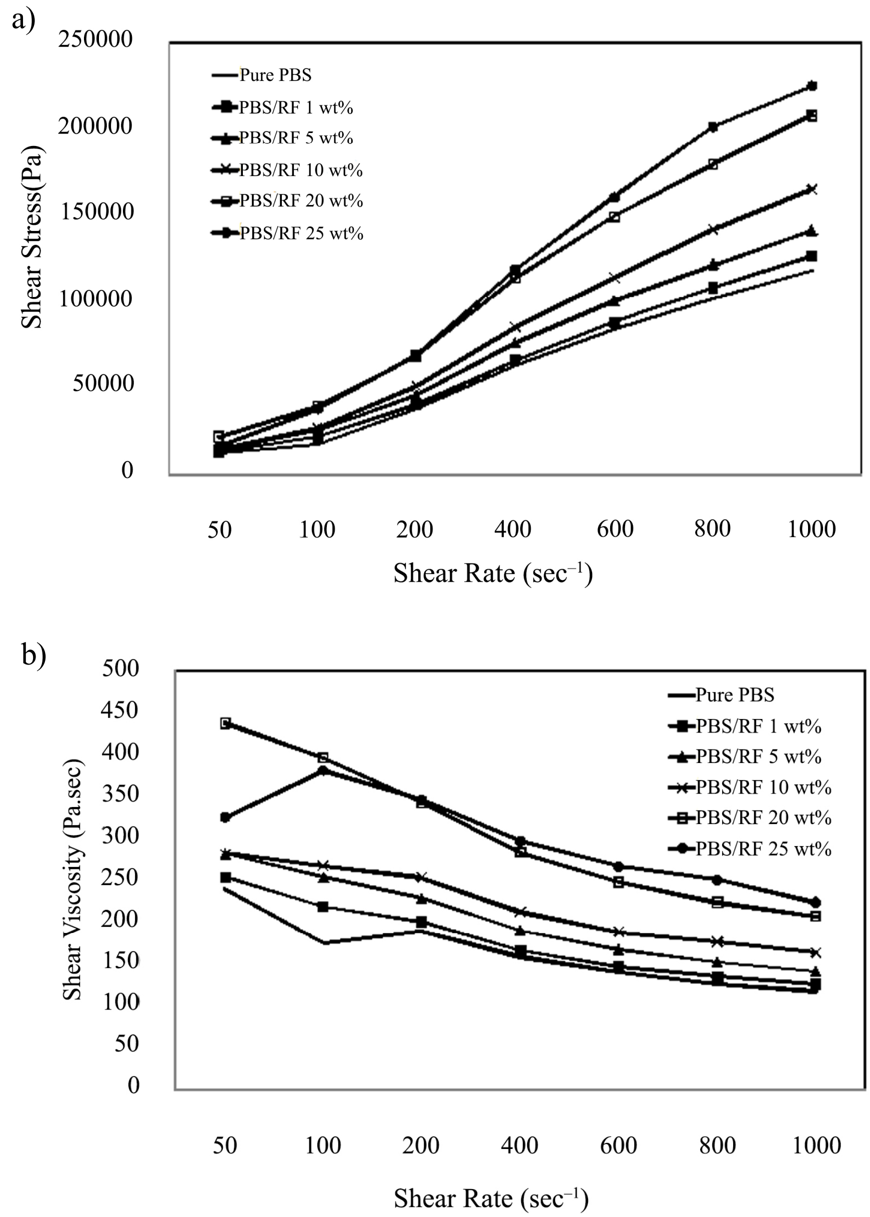

Fig. 1 Melt rheological properties of PBS/RF composites by varying content of RF; a) Shear stress, b) Shear viscosity.

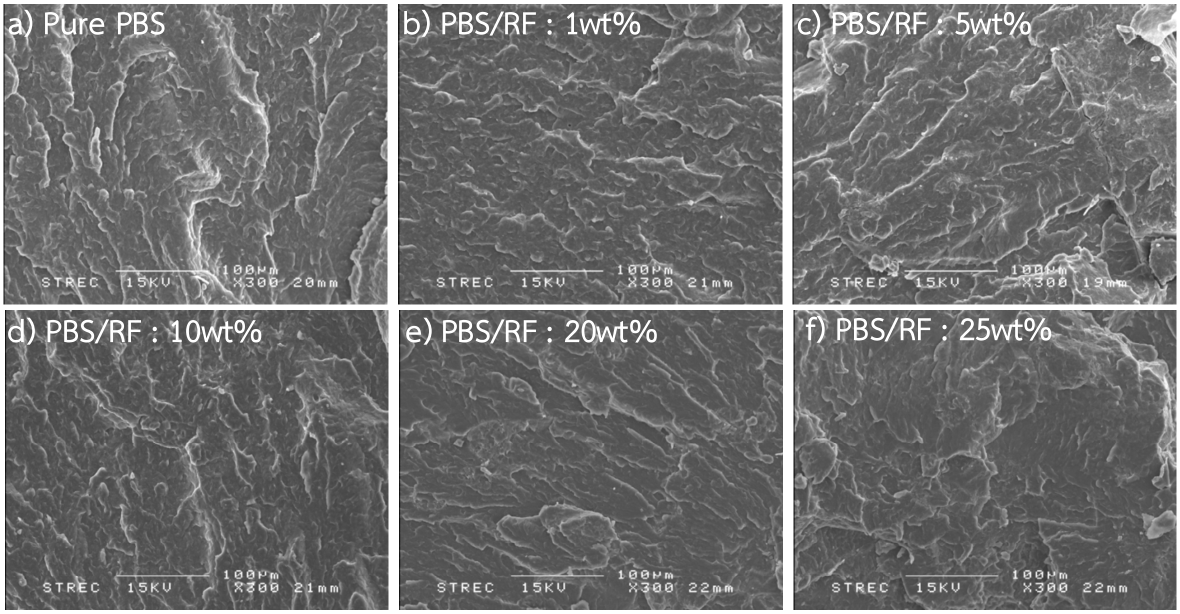

Fig. 2 SEM micrographs of PBS/RF composites.

In order to improve properties of PBS/RF composites, epoxidized natural rubber (ENR-50) was employed as a compatibilizer. The effectiveness of ENR-50 for PBS/RF composites was investigated. The improvement in phase morphology and melt rheological properties of PBS/RF composites was observed. From Fig. 3 and Fig. 4 show the effect of ENR-50 as a compatibilizer on phase morphology and rheological properties, which indicated that the incorporation of ENR-50 into the PBS/RF composites enhance the compatibility, shear stress and viscosity of the composites. The shear stress and viscosity of compatibilized composites exhibited higher value than uncompatibilized one. This could be due to the compatibilization effect of ENR-50 that helped to increased interfacial adhesion between rice flour and PBS, it made polymer chain more restricted to movement. This is in agreement with SEM micrographs of PBS/RF composites with ENR-50 (see Fig. 3), the roughness of the wall surface decreased. The addition of ENR-50 compatibilizer in composite showed that compatibility between rice flour and PBS matrix was enhanced leading to a strong interface.

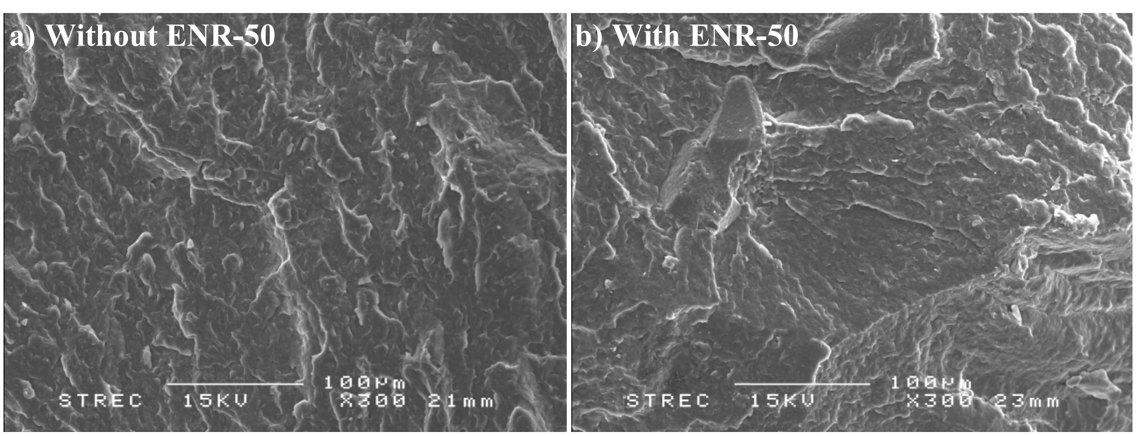

Fig. 3 SEM micrographs of PBS/RF composites; a) without ENR-50, b) with ENR-50 as a compatibilizer.

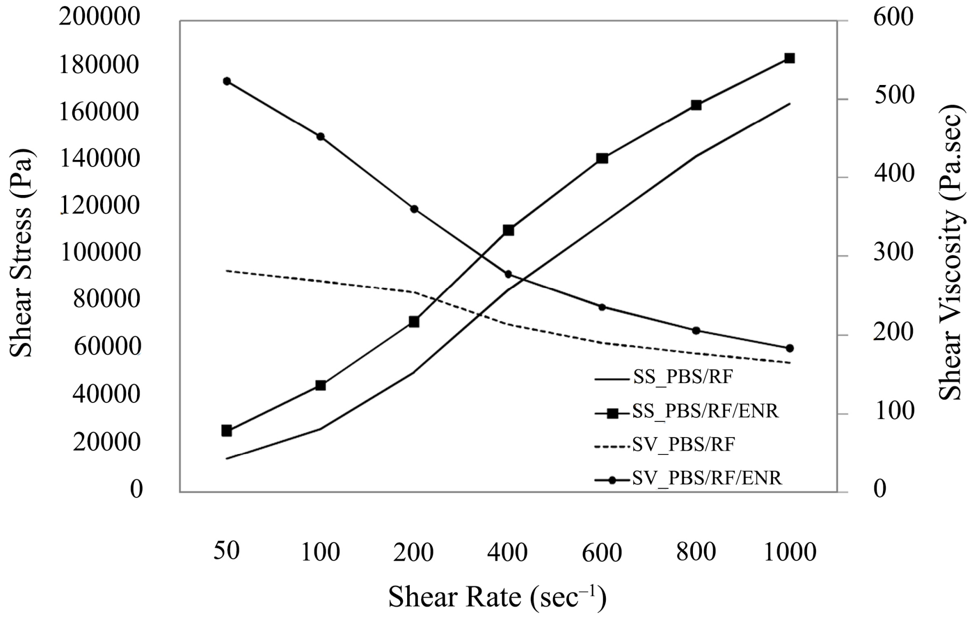

Fig. 4 Melt rheological properties of PBS/RF composites (with and without ENR-50). Shear stress (SS) versus shear rate and Shear viscosity (SV) versus shear rate.

The effect of rice flour loading and ENR-50 on mechanical properties is shown in Fig. 5 and Fig. 6. Tensile strength and tensile modulus of the composite are presented in Fig. 5(a) and 5(b), where as elongation at break and impact strength are listed in Fig. 6(a) and 6(b), respectively. The mechanical properties including tensile strength, elongation at break and impact strength of PBS/RF composites exhibit lower values than those of the pure PBS, where as tensile modulus of PBS filled with rice flour show higher value than pure PBS. This may be attributed to a higher modulus and rigidity of rice flour itself than neat PBS and the incompatible between matrix (PBS) and filler (rice flour). The addition of ENR-50 (at 5 phr) as a compatibilizer into PBS/RF composites increased the elongation at break and impact strength of the composite were clearly due to the reduction of interfacial tension between phases and elastomeric behavior of ENR-50. Similar results were also reported by S.T. Sam *et al.* [8] and G.H. Yew *et al.* [10], who showed that significant improvements in mechanical properties were observed by incorporating ENR-50 as a compatibilizer into composite materials. However, the tensile strength and modulus of PBS/RF composites with ENR-50 (5 phr) as a compatibilizer were decreased. The reduction of mechanical properties was observed when the amount of ENR-50 was too high. This may be due to the rubbery behavior of ENR-50. This is in consistent with the results reported by G.H. Yew *et al.* [10].

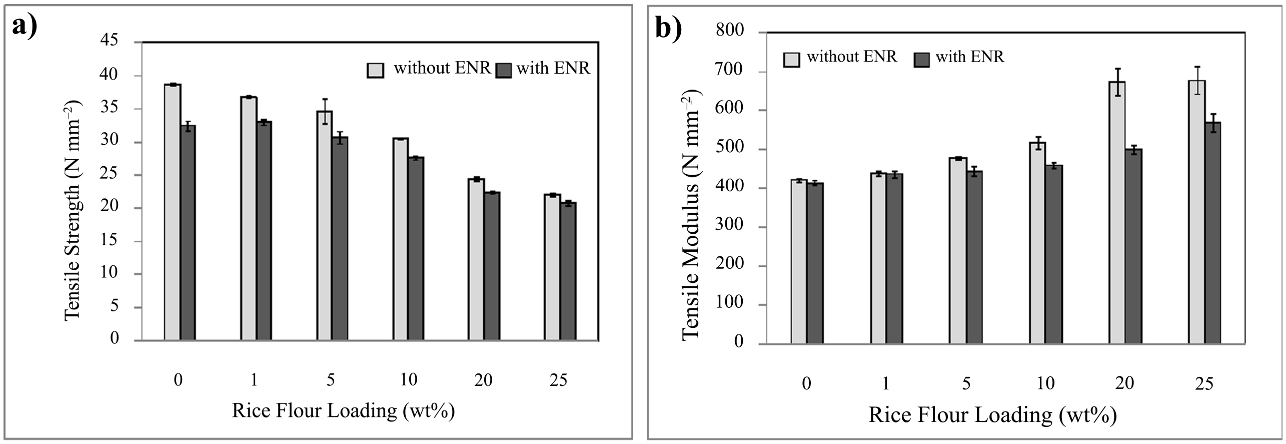

Fig. 5 Tensile strength and modulus of PBS/RF composites (with and without ENR-50).

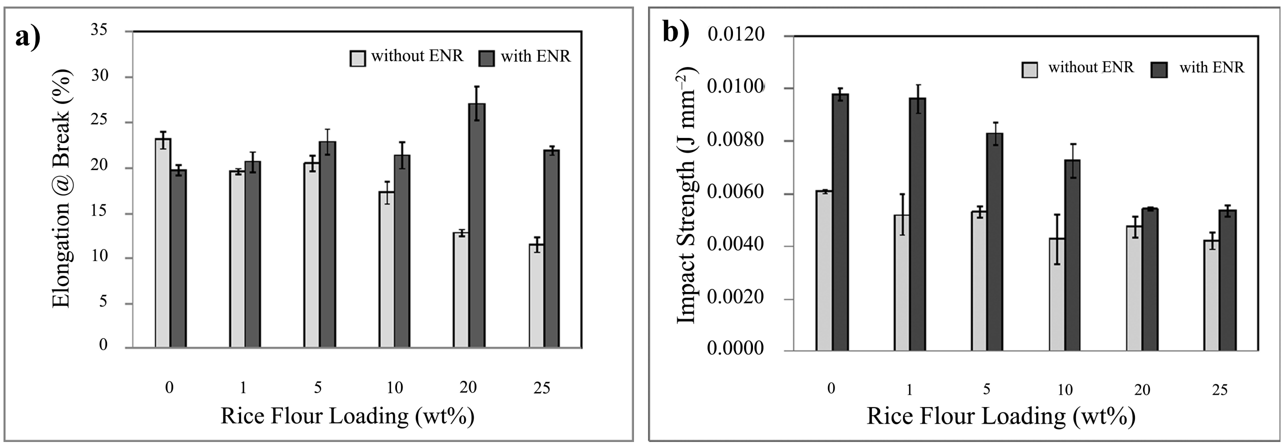

Fig. 6 Elongation at Break and impact strength of PBS/RF composites (with and without ENR-50).

4. Conclusion

Our approach in this research can be contributed to the improvement of properties of poly (butylene succinate)/rice flour composites using epoxidized natural rubber (ENR-50) as a compatibilizer. Tensile Modulus of PBS/RF composites were increased with increasing rice flour content. This may be attributed to the stiffening effect of rice flour itself. The suitable amount of rice flour in composites was 20 percent by weight. Epoxidized natural rubber (ENR-50) was effective as a compatibilizer for PBS/RF composites. The addition of ENR-50 improved compatibility of PBS/RF composites. This could be due to strong interaction between polar materials and epoxide group of ENR-50 which reduced the interfacial tension between phases. The shear viscosity, elongation at break and impact strength of the PBS/RF composites with ENR-50 was higher than that of the neat composites.

5. Suggestions

Although epoxidized natural rubber (ENR-50) was successfully modified PBS/RF composites, some properties, for example, tensile strength and tensile modulus still leave much room for improvement. The reduction of both tensile strength and modulus of compatibilized PBS/RF composites because of the amount of ENR-50 is too great. For further improvement, the suitable amount of ENR as a compatibilizer for PBS/RF composites may be the good answer.

6. Acknowledgement

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7. References

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