Effect of Benzyl Urea on Properties of Recycled High Density Polyethylene/Ethylene Vinyl Acetate/Egg Shell Powder Composites

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Keywords: Composites, Eggshell, Recycled high density polyethylene, Ethylene vinyl acetate.

Abstract. This paper presents the study on the effects of benzyl urea (BU) on the mechanical, morphological, and water absorption properties of recycled high density polyethylene/ethylene vinyl acetate/eggshell (rHDPE/EVA/ESP) composites. The results indicated significant improvement in the tensile strength and modulus of elasticity of the compatibilized composites due to better interfacial adhesion between the rHDPE and EVA. The water resistance properties were increased by the addition of the BU to the rHDPE/EVA/ESP composites. This study may suggest that BU can act as compatibilizer to enhance the interfacial interaction between the rHDPE and EVA matrix in the rHDPE/EVA/ESP composites.

Introduction

Blending is an easy and cost-effective way in producing new materials with new properties combination. EVA is an important industrial material, and is commonly used by blending with other polyolefins such as LDPE, HDPE, and PP [1–3]. Atul Rajan et al. [4] investigated the moisture absorption behavior of EVA/HDPE/OMMT blend nanocomposites, while Othman [5] studied on the effects of VA content and blending ratio of PE/EVA on the properties of PE/EVA blends. Akhlaghi et al. showed that the tensile modulus and toughness of HDPE/EVA/CaCO$_3$ ternary reinforced blends has been increased with the incorporation of EVA and CaCO$_3$. However, most blends achieved by melt-mixing are immiscible and thus show poor properties. Compatibilization is demanded to obtain a blend with desired properties to improve adhesion and compatibility for system containing filler and the matrix in the composites [6]. The agent modified the interface by interacting with filler and polymer, thus forming a link between the components [7]. Therefore, in this article, the effect of benzyl urea as a coupling agent on tensile properties, morphology, and moisture absorption behavior of rHDPE/EVA/ESP composites were investigated.

Experimental

Materials. rHDPE with a melt flow index of 0.7 g/10min (190°C) and a density of 939.9 kg/m$^3$ was used. EVA contains 6.5 wt% VA, with melt index of 2.5 g/10min (80°C; 2.16 kg) and density of 0.93 g/cm$^3$ was supplied from A.R. Alatan Sdn. Bhd., Kedah Darul Aman, Malaysia. The eggshell was obtained from local poultry industry in Negeri Sembilan, Malaysia.

Sample preparation. For the ESP preparation, the eggshell was washed, dried, and grinded to powder using kitchen blender. ESP with average filler sizes of 63 μm was dried in vacuum oven at 80°C till obtain the constant weight as reported in ref. [8]. For composites preparation, the
compounding of the blends was carried out by melt blending in Brabender internal mixer. The rHDPE was first mixed in the internal mixer at 190 °C, 50 rpm for 4 min and then pre-weighed amounts of EVA, BU, DCP, and ESP were added to the mixer for the next 6 minutes. Each of the molten samples was compression molded into sheets of 2mm thickness using a hydraulic press at 190 °C for 2 minutes and cooled under pressure for 4 minutes. Table 1 shows the formulation used in this study.

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**Characterization and Measurements.** Tensile properties of five samples from each composition were determined according to ASTM D638 by using the Instron 5569 with a cross-head speed of 50 mm/min. Morphology of the tensile fracture surface of rHDPE/EVA/ESP blends with and without BU were carried out using a scanning electron microscope (SEM), model JEOL JSM 6460LA. Water absorption test of five samples of every composition was carried out according to ASTM D750-95 standard. The water absorption was determined by weighing the samples at regular interval. A Mettler balance type AJ150 was used with precision of ±1 mg. The percentage of water absorption (Mt) was calculated using equation in [9].

**Results and Discussion**

**Tensile Properties.** Fig. 1(a) shows that the tensile strength of the composites decrease with the increasing content of ESP. This is due to the polymer system itself which consist of non-polar and polar components of rHDPE and EVA, respectively [10]. Besides, the ESP was unable to support the stress transfer from rHDPE/EVA matrix phase. A similar observation was reported by Supri et al. [11]. The addition of BU has improved the tensile strength which indicated that BU had facilitated the stress transfer between the phases due to the increased interfacial adhesion between the rHDPE/EVA matrixes and the ESP filler [11]. Fig. 1(b) shows that the elongation at break of the composites decreases with increasing filler loading. This is because the fillers has hardened the composites. The addition of BU to the blends has further reduced its ductility. At the same composition, rHDPE/EVA/ESP_BU composites had lower elongation at break then rHDPE/EVA/ESP composites due to the restriction in chain mobility of the matrix, and to the filler particles acting as defect points [8]. Fig. 1(c) shows that the Young’s modulus has increased when the amount of ESP in the matrix increased. At higher filler loading the composites will be able to withstand more loads. The Young’s modulus for compatibilized composites is higher than uncompatibilized composites. Again, this is due to better interfacial
adhesion between rHDPE/EVA matrix and ESP with the presence of BU as a compatibilizer. Similar observation was reported by Ali Dadfar et al [13].

![Graphs showing the effect of filler loading on tensile strength, elongation at break, and Young's modulus.](image)

Fig. 1. Effect of filler loading on (a) tensile strength, (b) elongation at break, and (c) Young’s modulus of rHDPE/EVA/ESP composites and rHDPE/EVA/ESP-BU composites

**Morphology Properties.** SEM micrograph in Fig. 2 shows as the filler loading increases, the micrographs show the detachment of filler from the matrix. The lower filler loading shows better dispersion and less pull out of filler from matrix compared to the higher filler loading. Fig. 2(b), 2(d), and 2(f) indicated rough surfaces and good dispersion of ESP filler in the rHDPE/EVA/ESP composites with benzyl urea. This results show that BU has improves the compatibility by more finely dispersing the filler in the polymer matrix compared to the uncompatibilized composites. Accordingly, the interfacial strength is improved.

![SEM micrographs of tensile fracture surface of rHDPE/EVA/ESP composites.](image)

Fig. 2. SEM micrograph of tensile fracture surface of rHDPE/EVA/ESP composites.
**Water Absorption Analysis.** Fig. 3 shows the percentage of equilibrium water absorption versus filler loading for rHDPE/EVA/ESP composites. It shows that the percentage of water absorption for RHDPE/EVA/ESP$_{BU}$ composites are lower than uncompatibilized rHDPE/EVA/ESP composites. The BU has builds up chemical bonds, which reduce the moisture-caused rHDPE/EVA debonding. This in turn reduces the extent of water absorption. Similar observation was reported by Atul Rajan et al [4].

![Figure 3. Percentage of equilibrium water absorption for rHDPE/EVA/ESP composites.](image)

**Conclusion**

The addition of BU as a compatibilizer in the mechanical properties, morphology, and water absorption in rHDPE/EVA/ESP composites were evaluated. BU enhanced the interfacial adhesion between rHDPE/EVA matrix and ESP which improved the tensile properties, water absorption resistance, and equilibrium water absorption of the rHDPE/EVA/ESP composites. Morphological studies on tensile fracture surfaces indicated that BU modified rHDPE/EVA/ESP composites have higher strength and better interaction than the uncompatibilized composites.

**References**