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Recycled High Density Polyethylene/Ethylene Vinyl Acetate Blends: The Effect of Caprolactam-Maleic Anhydride and Polyethylene-grafted-Maleic Anhydride on Tensile Properties and Swelling Behavior

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Abstract – *In this study, compatibilizing effects of caprolactam-maleic anhydride (CL-MAH) and polyethylene-grafted-maleic anhydride (PE-g-MAH) with different blend compositions on polymer blends of recycled high density polyethylene (RHDPE) and ethylene vinyl acetate (EVA) blends were investigated by tensile properties and swelling behavior. The use of CL-MAH improved the compatibility of RHDPE80/EVA20 blend composition, and tensile properties of the resultant blend compared well with those of RHDPE/EVA blends. Addition of PE-g-MAH improved the tensile properties of RHDPE/EVA/PE-g-MAH blends than RHDPE/EVA blends without compatibilizer. The scanning electron microscopy (SEM) morphologies of fracture surface for RHDPE/EVA blends with presence of compatibilizers showing better incorporation of two different phases of polymer matrix.*

Keywords: Polymer Blends, Recycled High Density Polyethylene, Ethylene Vinyl Acetate, Compatibilizers, Caprolactam-Maleic Anhydride, Polyethylene-grafted-Maleic Anhydride

1.0 INTRODUCTION

Polymer blending has become a matter of increasingly importance way to achieve materials with targeted properties especially for specific applications, at a comparatively low in cost than development of a new kind of polymer [1]. Polymer blending has been acknowledged as the most versatile and cost-effective method to formed new multi-phase polymeric materials which able to satisfy the complex demands of performance [2]. Polymer blends are usually categorized as homogenous blends, in which the components are miscible; and heterogenous blends, in which the components are immiscible [3]. Immiscible polymer blends contain various multiphase morphologies depending on the composition and on process parameters. These multiphase morphologies need to be controlled to obtain reproducibility properties [4]. Coarse phase morphologies which present in immiscible polymer blends can cause poor ultimate properties of the blends. The interfacial tension between segregated phases is high and acts as a blockade to an effective stress transfer between the phases. Improvement of polymer blends can be done by addition of compatibilizer which is the easiest ways to achieve compatibilization [5]. Compatibilizer plays a vital role in surpressing coalescence of the dispersed domains achieved through stabilizing the interphase. Compatibilized blends are considered by the presence of finely dispersed phase, sturdy resistance to phase coalescence, better adhesion between blend phases, and obtain desirable properties [6-8].

2.0 METHODOLOGY

2.1 Materials

RHDPE with melt flow index of 0.7g/10min (190°C) and density of 939.9kg/m^3 was used. EVA contains 18.1wt% VA, with melt index of 2.5g/10min (80°C, 2.16kg) and density of 0.93g/cm^3 was supplied from A.R. Alatan Sdn. Bhd., Kedah Darul Aman, Malaysia. Maleic anhydride with molecular weight of 98.06g/mol was supplied by Zarm Scientific & Supplier Sdn. Bhd., Penang, Malaysia. Caprolactam with molecular weight of 113.16g/mol, polyethylene-grafted-maleic anhydride containing 0.85wt% of maleic anhydride and dibenzoyl peroxide (DBP) with 75% of water were also obtained from A.R. Alatan Sdn. Bhd., Kedah Darul Aman, Malaysia.

2.2 Sample 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 160°C with speed 50 rpm for 2 min, followed by addition of EVA and mixed until homogenous. The compatibilizer, CL-MAH and PE-g-MAH; and dibenzoyl peroxide (DBP) were added to the mixer for the remaining minutes. Each of the compounding was compression molded into sheets of 2mm thickness using a hydraulic press at 160°C for 6 min and cooled under pressure for 4 min. Table 1 shows the formulation used in this study.

Table 1: Formulation of RHDPE/EVA, RHDPE/EVA/CL-MAH, and RHDPE/EVA/PE-g-MAH blends at different blend compositions.

Composites	**RHDPE/EVA (phr)**	**CL-MAH (phr)**	**PE-g-MAH (phr)**
RHDPE/EVA	80/20, 60/40, 40/60, 20/80	-	-
RHDPE/EVA/CL-MAH	80/20, 60/40, 40/60, 20/80	6	-
RHDPE/EVA/PE-g-MAH	80/20, 60/40, 40/60, 20/80	-	6

2.3 Characterization and Measurements

Mechanical properties of the composites were measured by tensile testing using Universal Testing Machine Instron 5582 with crosshead speed of 30 mm/min. Dumbbell shaped specimens were conditioned at ambient temperature before testing. Surface morphologies of RHDPE/EVA blends with and without compatibilizers were studied by using Scanning Electron Microscopy machine. Surfaces of the samples were coated with a thin platinum layer about 12µm thickness using the Auto Fine Coater to avoid electrostatic charged during examination.

3.0 RESULTS AND DISCUSSION

3.1 Tensile Properties

Figure 1 shows the effect of compatibilizer on tensile strength of RHDPE/EVA, RHDPE/EVA/PE-g-MAH, and RHDPE/EVA/CL-MAH blends. It can be seen that all blends significantly decreased in tensile strength with increasing the content of EVA. This was due to the presence of EVA which influenced the crystalline structure of RHDPE due to EVA amorphous characteristic. Increasing the content of EVA obstructs the ordered arrangement of RHDPE while reducing their crystallinity which causes reduction in tensile strength value [9].

At the same blend composition, RHDPE/EVA/PE-g-MAH blends revealed higher tensile strength than RHDPE/EVA blends in all different blend compositions. This can be explained by the improvement of interfacial adhesion as PE-g-MAH was added due to reaction of carbonyl group in PE-g-MAH to ester group of EVA, which would form covalent bonds, enhance the improvement and efficiency of stress transfer from RHDPE to EVA matrix. This is similar to the findings of several researches which agreed that with accumulation of compatibilizer to the blends will help to increase the tensile strength of the blends [10-12].

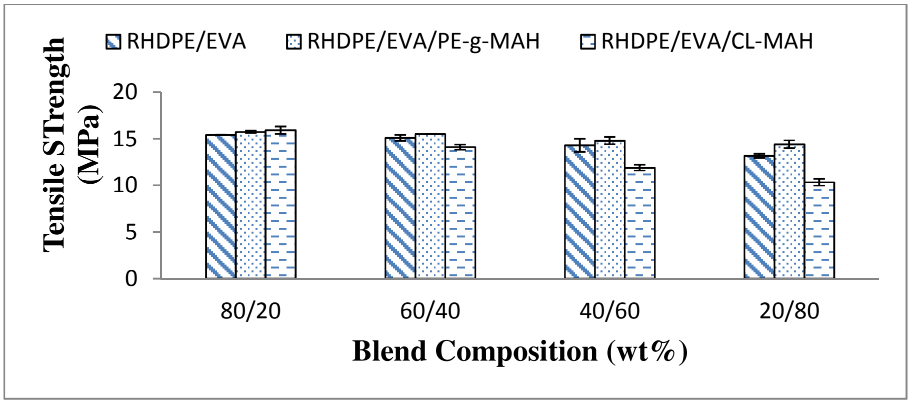


Figure 1: Effect of blend composition and compatibilizer on tensile strength of RHDPE/EVA, RHDPE/EVA/PE-g-MAH, and RHDPE/EVA/CL-MAH blends

Figure 1 also showed that the presence of CL-MAH in blend composition of RHDPE80/EVA20/CL-MAH displayed the highest in tensile strength, but showing decreased trend in other blend compositions of RHDPE and EVA. Hybridization of Caprolactam with Maleic Anhydride will create hydrogen bonding and dipole-dipole interaction between the two compatibilizers, thus created reactive compatibilizer which can upgrade the improvement of interfacial adhesion of the blends. The reduction of interfacial adhesion size improved the miscibility of the two components as dispersed phase EVA blended well into RHDPE matrix. However, as the content of EVA increased, the tensile strength of RHDPE/EVA/CL-MAH blends significantly decreased due to reactive compatibilizers of Caprolactam and Maleic anhydride not compatible with increasing the content of EVA.

Figure 2 presents the effect of different blend compositions and compatibilizers on elongation at break of RHDPE/EVA, RHDPE/EVA/PE-g-MAH, and RHDPE/EVA/CL-MAH blends. The elongation at break increased gradually with the increased in EVA loading because of increased in the toughness of RHDPE/EVA blends due to higher elastic properties of EVA. Elasticity of EVA will somehow reduce the crystallinity properties of RHDPE thus increased elongation at break of the blends. Ismail et al. stated that by increasing the NBR content in PVCw/NBR blend will reduced the stiffness and brittleness of the blends gradually while increased the elongation at break [13].

At similar blend composition, the elongation at break of RHDPE/EVA with both compatibilizers, PE-g-MAH and CL-MAH was lower than the blends without compatibilizers. The addition of compatibilizers was an obvious indication of improved adhesion at the interface of the matrixes, increased blend stiffness, but usually causes a notable reduction in the elongation at break.

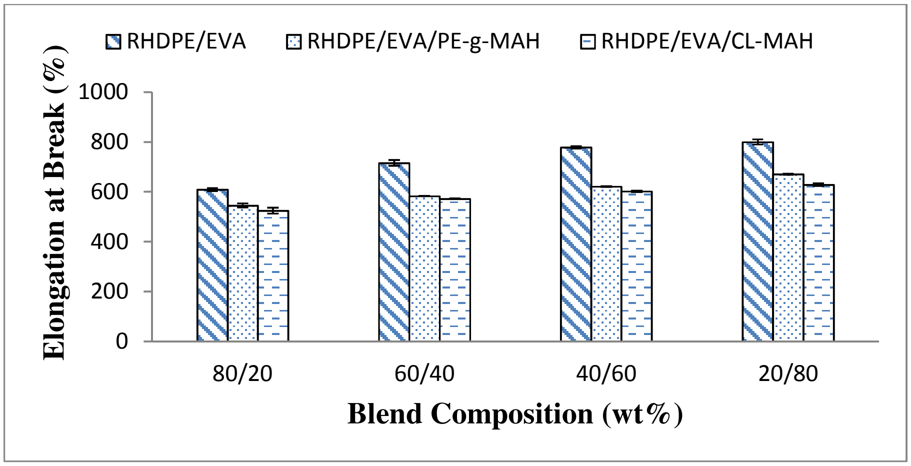


Figure 2: Effect of blend composition and compatibilizer on elongation at break of RHDPE/EVA, RHDPE/EVA/PE-g-MAH, and RHDPE/EVA/CL-MAH blends.

The effect of different blend composition and compatibilizer on modulus of elasticity of RHDPE/EVA, RHDPE/EVA/PE-g-MAH, and RHDPE/EVA/CL-MAH blends was shown in Fig. 3. All of the sets presented a decreasing trend as the composition of EVA increased. The decreased of modulus of elasticity for all the composites by increasing EVA content might due to the elastic characteristic of EVA which having rubber-like properties, thus increased ductility of the composites. Moreover, decreased in stiffness and toughness of composites resulted in decreasing tensile modulus of the composites. Faker et al. had studied the effect of mechanical behavior of PE/EVA blends. The result concluded that from the results of particle size distribution measurements showed that PE-rich blends had better tensile properties than EVA-rich blends. This was due to small and well distributed particles lead to improvement in compatibility and interfacial interaction [14].

The modulus of elasticity for compatibilized blends with PE-g-MAH had better modulus of elasticity than RHDPE/EVA blends without presence of compatibilizer. The addition of PE-g-MAH to the RHDPE/EVA blends altered the surface of the blends and became coarse. The association of PE-g-MAH at the interface of two different matrixes resulted in lowering interfacial tension of the blends, therefore increased the modulus values [5].

However from Fig. 3, the addition of CL-MAH increased modulus of elasticity than RHDPE/EVA and RHDPE/EVA/PE-g-MAH blends but just on RHDPE80/EVA20 but decreased the tensile modulus with increasing EVA content. The reasoning behind this could be that the compatibilizing effect was restricted merely to the amorphous part of EVA that will reduce the modulus value accordingly. These results also validated to the incompatibility of the blends with addition of CL-MAH corresponding to increase of EVA which reduced the tensile properties of the blends.

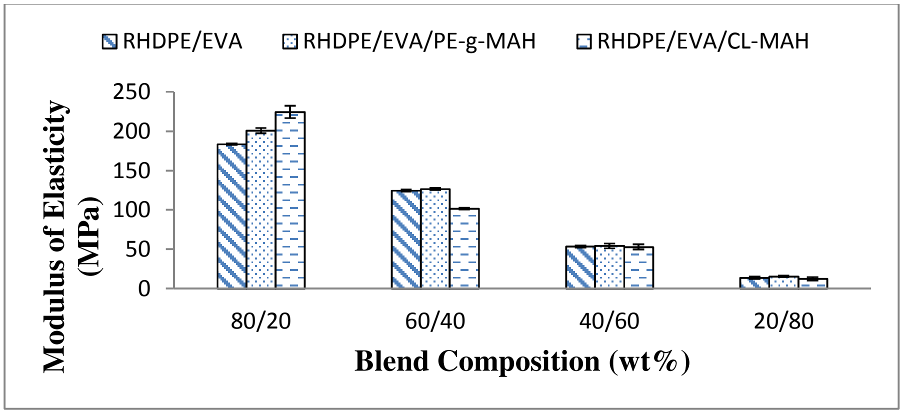


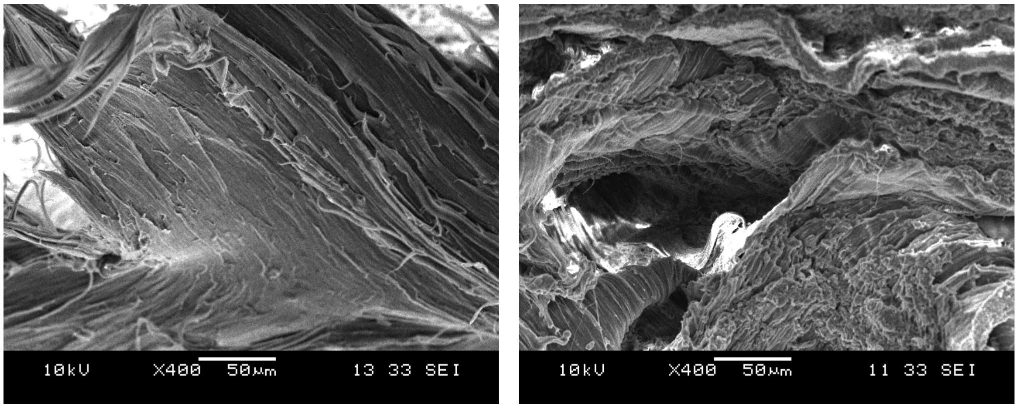
Figure 3: Effect of blend composition and compatibilizer on modulus of elasticity of RHDPE/EVA, RHDPE/EVA/PE-g-MAH, and RHDPE/EVA/CL-MAH blends.

3.2 Morphology Analysis

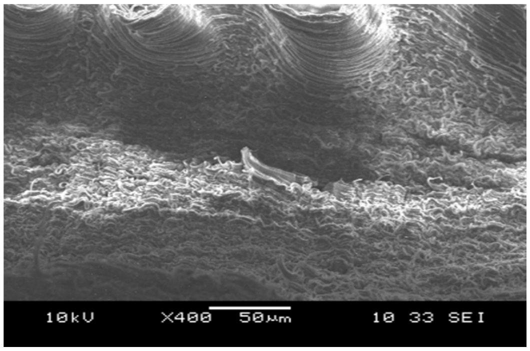
SEM micrographs of the tensile fracture surfaced of RHDPE/EVA, RHDPE/EVA/PE-g-MAH, and RHDPE/EVA are shown in Fig. 4. From Fig. 4 (a-c), the morphology of blends without addition of compatibilizer showed poor interfacial adhesion with clearly seen many gaps and distinct cavities within the RHDPE phase and EVA phase. It can be seen in all these figures that as the EVA composition increases, the nature of failure surface changes from the rough failure surfaces into smooth failure surfaces.

At same composition, the addition of PE-g-MAH in RHDPE/EVA/PE-g-MAH blends is shown in Fig. 5 (a-c), the compatibilization of blends were improved with the interface of two phases became more indistinct and less noticeable. Nevertheless, the presence of many tear lines indicated that strong interfacial adhesion between the polymers, thus higher strength needed to break the blends as proved with higher value of tensile strength in Fig. 1. Akhlaghi et al. had reported that by addition of PE-g-MAH as a compatibilizer in HDPE/EVA matrix decreased the interfacial tension between the major and minor phases, hence providing finer dispersion of minor phase in the matrix [15].

Figure 5 (d-f) displays the SEM micrographs of tensile fracture surfaces of RHDPE/EVA/CL-MAH blends at different blend compositions. Fig. 5 (d) shows the EVA was finely and uniformly dispersed in the continuous RHDPE matrix with existence of CL-MAH. This suggesting a relatively higher efficiency of compatibilization of the blend using CL-MAH consequently enhanced tensile properties. However, Fig. 5 (e-f) clearly revealed the smoothness in fracture surfaces with uneven distribution of the dispersed EVA phase and unstable particle structure may cause by the reactive compatibilizer which not applicable to the blend composition of RHDPE60/EVA40 and RHDPE20/EVA80.

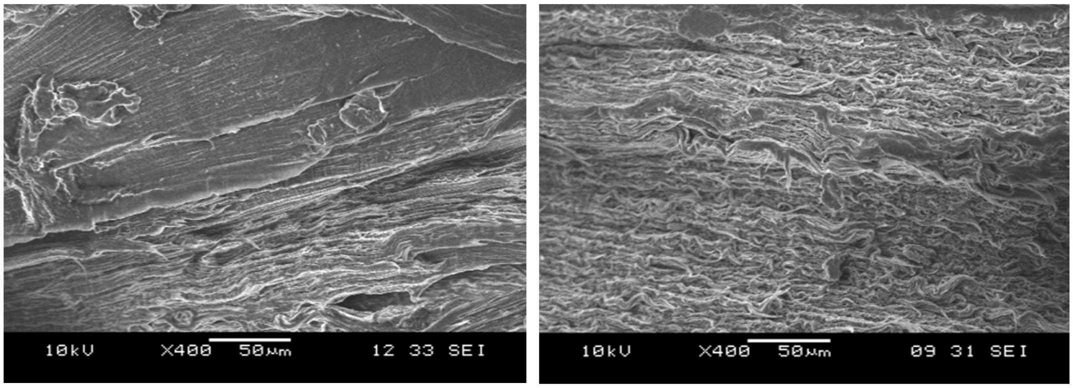


(a) RHDPE80/EVA20 (b) RHDPE60/EVA40

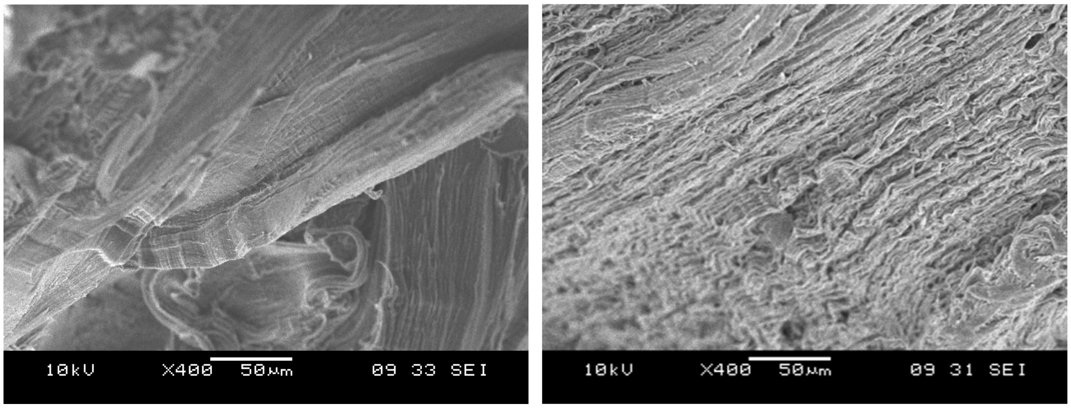


(c) RHDPE20/EVA80

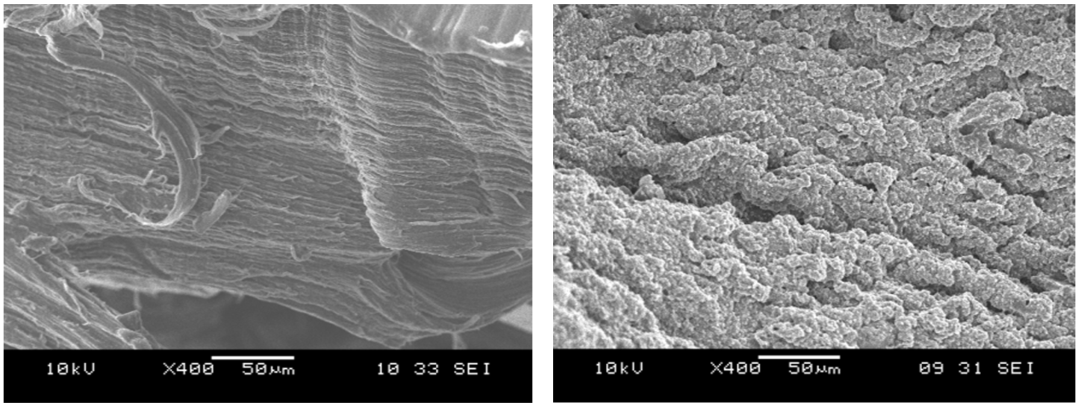
Figure 4: Scanning Electron Microscopy micrographs of tensile fractured surfaces of RHDPE/EVA blends.



(a) RHDPE80/EVA20/PE-g-MAH (d) RHDPE80/EVA20/CL-MAH



(b) RHDPE60/EVA40/PE-g-MAH (e) RHDPE60/EVA40/CL-MAH



(c) RHDPE20/EVA80/PE-g-MAH (f) RHDPE20/EVA80/CL-MAH

Figure 5: Scanning Electron Microscopy micrographs of tensile fractured surfaces of RHDPE/EVA/PE-g-MAH and RHDPE/EVA/CL-MAH blends.

4.0 CONCLUSION

The addition of CL-MAH and PE-g-MAH as compatibilizers into RHDPE/EVA blends improved tensile strength and modulus of elasticity while reducing elongation at break. Microscopic fractured morphology of the blends showed better interfacial interaction between two phases of RHDPE and EVA with the existence of compatibilizers. The introduction of hybrid compatibilizer, which is combination of caprolactam and maleic anhydride help to improve the properties of RHDPE/EVA blends better than properties of RHDPE/EVA blends with addition of PE-g-MAH.

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