Grafting Maleic Anhydride onto EVA and Effect on the Physical and Rheological Properties of PETG/EVA-*g*-MAH Blends

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ABSTRACT: Maleic anhydride (MAH) was grafted onto ethylene vinyl acetate copolymer (EVA) in the internal mixer in the presence of dicumyl peroxide (DCP), and a prepared reactive compatibilizer, MAH-*g*-EVA, was blended with Poly (ethylene glycol-*co*-cyclohexane-1,4-dimethanol terephthalate) (PETG). The gel content determination and element analysis (EA) was performed to confirm the grafting reaction. It was found that grafting reaction of MAH on to EVA could compete with crosslinking reaction of DCP during the modification process. In addition, the introduction of small amount of MAH showed a great effect on reducing gel content by decreasing crosslink reaction. As MAH content increased, grafted MAH concentration increased, whereas the grafting yield decreased. It was also confirmed that MAH-*g*-EVA acted as a reactive compatibilizer in the blend with PETG, and enhanced compatibility by reacting with the hydroxyl end groups, OH, of PETG. Addition of EVA in the blend leads a plastic deformation of PETG, and MAH had a great effect on enhancing interfacial adhesion resulting in significant increasing of % strain; however, improved compatibility could not be changed much in low strain tensile strength and Young's modulus. © 2012 Wiley Periodicals, Inc. J Appl Polym Sci 125: 2732–2739, 2012

Key words: PETG; EVA; MAH; blends; compatibilization; crosslinking

INTRODUCTION

Polymer blends have attracted considerable concerns for many years since blends of two polymers give rise to improved physical properties. Most of polymer blends are immisicible system and shows a poor physical property because it greatly depends on the compatibility of two polymers, and their interfacial tension hinders the formation of coalescence during melt mixing resulting in poor interfacial adhesion.[1] Addition of block or graft copolymer on the blend have been common way to enhance the compatibility and interfacial adhesion of two polymers. However, molecular weight of polymer applied to blends is also important factor should be considered, and there are cost problem induced by preparation of copolymer for special application.[2–4]

The reactive compatibilization is the method to improve compatibility by a chemical reaction of two polymers during their blending. It is well known that the polymers with —OH, —COOH, and/or —NH$_2$ groups in their chain ends could chemically coupled with reactive polymers such as maleic anhydride,[5–7] acrylic acid,[8] oxazoline,[9,10] epoxide,[11–15] and vinyl silane.[16] The compatibilizer formed by chemical reaction between polymer chains that is mutually reactive is called *In situ* formed compatibilizer. The maleic anhydride (MAH) is one of the most widely used reactive compatibilizer due to its good chemical reactivity. Many researchers have reported that MAH could be easily grafted onto polyolefin in the presence of an initiator during melt processing.[17–23] Gaylord et al.[17–19] reported the formation of single and oligomeric grafts by grafting of MAH onto polyolefins and proposed that bridge could be formed between grafted MAH and polymer chains.

Poly (ethylene glycol-*co*-cyclohexane-1,4-dimethanol terephthalate) (PETG) is amorphous polyester with about 80°C of glass transition temperature (T_g). This polymer offers a broader processing parameters than normal crystallizable polymers and is very useful for obtaining high-clarity amorphous molding.[24] Many studies dealing with physical, mechanical, and rheological properties of PETG and its blend

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with other polymers were investigated for many years.[25–32] Yu et al.[26] found that MAH-grafted thermoplastic elastomer (TPE) improved the compatibility between the PETG and TPE resulting in good dispersion of the TPE in the PETG matrix. Saheb and Jog[28] reported that PETG blends with poly(butylene terephthalate) (PBT) showed a single composition-dependent T_g, melting point depression, and reduced crystallization rate. Papadopoulou et al.[29,30] found that the PETG/poly (ethylene terephthalate) (PET) blends were miscible when the PETG content was more than 50 vol %, and the properties were enhanced due to interaction of two polymers. Hwang et al.[32] investigated thermal and mechanical properties of the blend of PETG and liquid crystalline copolyester (LCP). They found that the interfacial adhesion of PETG and LCP was poor indicating that the blend was immiscible but LCP acted as a reinforcing agent in the blends improving tensile strength and modulus.

In this study, MAH was grafted to ethylene vinyl acetate copolymer (EVA) in the presence of dicumyl peroxide (DCP) as an initiator, and MAH-g-EVA was blended with Poly (ethylene glycol-co-cyclohexane-1,4-dimethanol terephthalate) (PETG). The dependence of MAH grafting reaction onto EVA on the concentration of DCP and MAH was investigated. PETG/MAH-g-EVA blend was prepared, and the morphology and mechanical properties such as tensile strength, Young's modulus, and % strain at break were assessed to determine the effect of MAH-g-EVA on the blend as reactive compatibilizer.

EXPERIMENTAL

Materials

Poly (ethylene glycol-co-cyclohexane-1,4-dimethanol terephthalate) (PETG, S2008) was obtained from SK Chemicals (Seoul, South Korea). PETG is amorphous polymer with 80°C of the glass transition temperature (T_g) and the density was 1.27 g/cm³. Ethylene vinyl acetate copolymer (EVA, VS410) containing 26 wt % of vinyl acetate (VA) with −65 and 73°C of T_g and melt temperature (T_m) was provided by Hyundai Petrochemical Co. (Seoul, South Korea). Melt Flow Index (MFI) and density were 4.0 g/10 min (ASTM D1238) and 0.950 g/cm³, respectively. Dicumyl peroxide (DCP, 98%) as a radical initiator was purchased from the Aldrich Chemical Co. (St. Louis, MO). Maleic anhydride (MAH) for the graft modification was obtained from Shinyo Pure Chemical Co. (Osaka, Japan) and was used as received. EVA was vacuum dried at room temperature for 24 h before use.

TABLE I
Composition of DCP and MAH for EVA Modification

DCP contents (phr)	MAH contents (phr)				
	0	0.5	1.0	2.0	3.0
0.1	D1M00	D1M05	D1M10	D1M20	D1M30
0.2	D2M00	D2M05	D2M10	D2M20	D2M30

Preparation and characterization of MAH-g-EVA

The composition for modification used in this study is presented in Table I. The designation of D and M mean DCP and MAH, respectively, and the numerical values present content (phr) of the DCP and MAH in the samples. MAH-g-EVA was prepared with a Brabender plasti-coder® (PLE331) (Duisburg, Germany). EVA and MAH (0.5, 1.0, 2.0, and 3.0 phr, based on DCP concentration) was first mixed for 5 min at 175°C, and then DCP (0.1 and 0.2 phr) was introduced, and mixing was continued for 10 min. The torque variation was recorded during the mixing.

The gel content was measured to investigate the crosslinking reaction of EVA that may be occurred during grafting reaction by DCP. MAH-g-EVA (0.3 g) was sealed in a standard sieve pouch (120 meshes) and weighted, and then extracted by immersing in boiling xylene for 12 h. The antioxidant, Irganox1010 (Ciba Specialty Chemicals), was added to protect further thermal crosslinking reaction. All extracted samples were dried in a vacuum oven at 50°C for 24 h, and the gel content was calculated using eq. (1).

$$\text{Gel contents}(\%) = \frac{W_2 - W_3}{W_1 - W_3} \times 100 \qquad (1)$$

where W_1, the weight of specimen and the pouch before extraction; W_2, the weight of specimen and the pouch after extraction; W_3, the weight of the pouch.

Element analysis (EA) was performed to determine the MAH content grafted to EVA. The MAH-g-EVA was dissolved with xylene for 5 h, and then it was precipitated with acetone to remove an unreacted MAH. The precipitated MAH-g-EVA was further washed with acetone and dried in vacuum oven until the sample weight approaches to steady state. The EA was determined with the Elemental Analyzer (EA 1108) (Fisions Instrument Spa, San Carlos, CA), and MAH content and grafting yield were calculated using eqs. (2)–(4).

$$[O]_{total} - [O]_{EVA} = [O]_{grafted \ MAH} \qquad (2)$$

$$[O]_{\text{grafted MAH}} \times [(C + H)/O]_{\text{MAH}} = [C + H]_{\text{grafted MAH}}$$
$$(3)$$

$$[O]_{\text{grafted MAH}} + [C + H]_{\text{grafted MAH}} = [MAH]_{\text{EVA−g−MAH}}$$
$$(4)$$

Preparation and characterization of PETG/MAH-g-EVA blend

The prepared MAH-g-EVA polymer (20 phr) and PETG were blended with a Brabender plasti-coder® with a rotor speed of 50 rpm at 210°C for 10 min. The blend samples were cooled at room temperature, and then the film with 0.3 mm of thickness was prepared by compression molding process with two 12 × 12 cm platens by a hydraulic laboratory press (Model C) (Carver, Wabash, IN) with a pressure of 14.7 MPa for 3 min at 210°C. The Molau solution test was carried out to investigate compatibility between functional groups of PETG and MAH-g-EVA.[26,32–35] A total of 0.3 g of both PETG/unmodified EVA (uEVA) (20 phr) and PETG/D1M20 (modified EVA, mEVA) (20 phr) was mixed with 10 mL of tetrachloroethane/phenol (4 : 6 weight ratio) solution, and then left for 60 days at room temperature. The melt flow index (MFI) of the blends was measured by a melt flow indexer (Davenport MFI-10) (Lloyd instruments, Hants, UK). The three different temperatures (180, 200, and 220°C) were used, and then the MFI changes were recorded. The load and cutting time were 2.16 kg and 30 s, respectively. A Rheometer (UDS-200) (Paar Physica, Ashland, VA) was used to determine the rheological properties. Frequency sweeps within the range of 1–150 rad/s were conducted at 180, 200, and 220°C, and 0.3% of the strain amplitude was fixed to obtain reasonable signal intensities. The mechanical properties such as tensile strength and elongation at break were characterized by a universal testing machine (Instron M4465) (Instron, Norwood, MA) according to ASTM D638. The test was performed with a crosshead speed of 300 mm/min. The morphology of the blends was investigated by using the field emission scanning electron microscope (FE-SEM, Hitachi S-4300) (Japan). The specimens were fractured after freezing in liquid nitrogen, and fracture surface was sputter coated with gold and observed.

RESULTS AND DISCUSSION

Grafting of MAH to EVA

A mixing torque is commonly used to monitor chemical reaction during the mixing. Figure 1 shows a torque variation of MAH-g-EVA polymer mixed with various content of MAH as a function of time while 0.1 phr of DCP content was fixed. EVA poly-
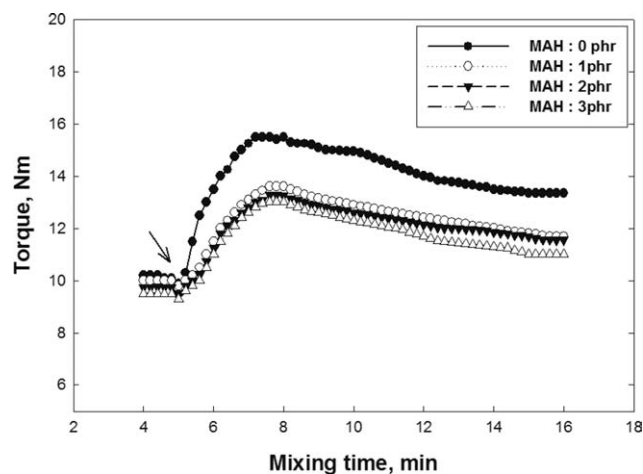
Figure 1 Variation of mixing torque during EVA modification reaction with 0.1 phr of DCP at various MAH contents.

mer with higher concentration of MAH shows lower mixing torque. This can be explained that low molecule MAH presents as a molten state without reactivity. Introduction of DCP as marked by the arrow causes a significant increase in torque value, and this may be attributed to crosslinking reaction of EVA with DCP. In addition, the maximum torque decreased significantly as MAH content increases. This decrease may be due to grafting reaction of MAH implying that MAH grafting reaction may compete with crosslinking reaction by DCP during the modification. DCP was added after EVA and MAH were mixed to have more grafting reaction of MAH than crosslinking reaction of DCP.

Kim et al.[7] reported that when molten EVA is presented with DCP and MAH at 175°C at a plasticorder, following reaction would occur.
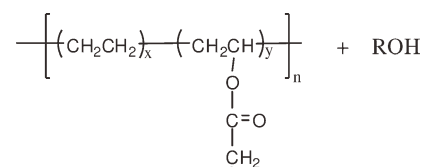
(a) Formation of the initiator radical

$$R-O-O-R \qquad 2RO\cdot$$
(Peroxide)   (Primary radical)

(b) Radical transfer reactions occurring between the primary radical and EVA used
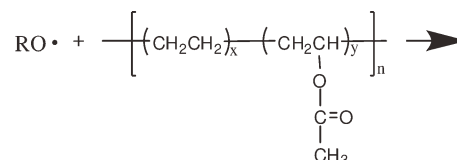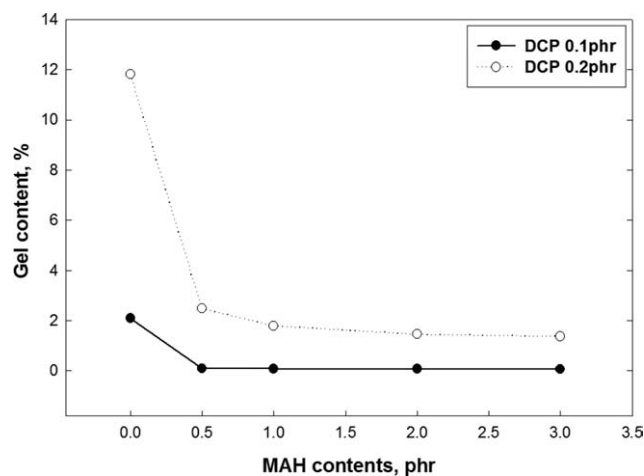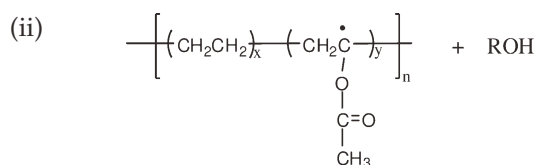

(i)

Figure 2 Gel contents of EVA-*g*-MAH as a function of MAH contents and DCP concentration.

(ii)

(c) Addition of MAH to primary radical or the EVA macroradical

(d) Crosslinking by mutual termination of the grafted propagating chains or EVA macroradicals

(i)

(ii)

The effects of the content of DCP and MAH on the gel contents were given in Figure 2. MAH-*g*-EVA with higher DCP content shows the higher gel content due to more crosslinking reaction of EVA induced by DCP. It was also found that 0.5 phr of MAH significantly reduced gel content and approaches to steady state, and this confirmed that a small amount of MAH has great effect on decreasing the crosslinking reaction.

Figure 3 shows the grafting yield and grafted MAH content as function of MAH content. As DCP content increases, both grated MAH content and grafting yield were improved. This may be due to
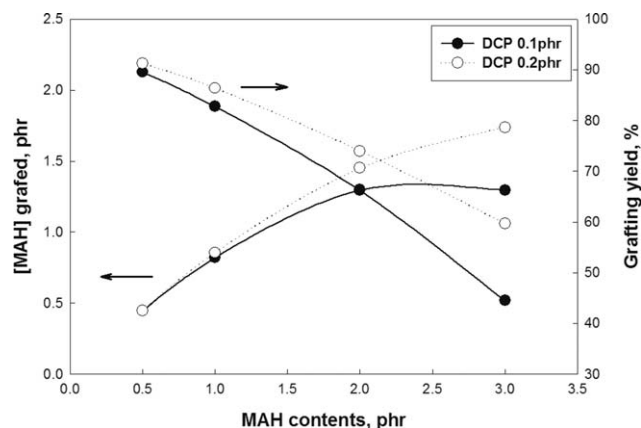

Figure 3 Grafting yield and grafted MAH concentration as a function of MAH contents.

the increase radicals promoting more reaction with EVA. It was observed that increase of MAH content leads an increase of grafted MAH content, but a decrease in the grafting yield. The grafting yield and gel content with various contents of DCP and MAH were given in Table II. D1M20 shows an optimum grafted MAH and yield, and then was used for further study of PETG/MAH-*g*-EVA blend as mEVA that is differentiated from uEVA, unmodified EVA.

PETG/EVA-*g*-MAH blend

The torque variation during the blending of PETG/MAH-*g*-EVA is shown in Figure 4. The numerical values denote the weight percentage (wt %) of MAH and DCP in PETG/EVA blends. EVA301 indicates 3 wt % of MAH with 0.1 wt % of DCP in blends. It was observed that torque value of the blend with mEVA is higher than uEVA and increases as MAH content increases, whereas DCP content was kept constant (0.1 phr). This may be due to increase of

TABLE II
Summary of Equipment and Condition for Modification

Exp no.	$[DCP]_i$ (phr)	$[MAH]_i^a$ (phr)	$[MAH]_g^b$ (phr)	Grafting yield^c(%)	Gel contents (%)
D1M00	0.1	0	–	–	2.09
D1M05	0.1	0.5	0.45	89.56	1.08
D1M10	0.1	1	0.82	82.77	0.08
D1M20	0.1	2	1.30	66.20	0.07
D1M30	0.1	3	1.30	44.48	0.05
D2M00	0.2	0	–	–	11.81
D2M05	0.2	0.5	0.45	91.20	2.48
D2M10	0.2	1	0.85	86.41	1.80
D2M20	0.2	2	1.45	74.01	1.45
D2M30	0.2	3	1.73	59.70	1.37

^{a} MAH concentration introduced.
^{b} MAH concentration grafted.
^{c} The grafting yield is defined as the ratio of concentration of grafted MAH to initial MAH, $[MAH]_g/[MAH]_i$.
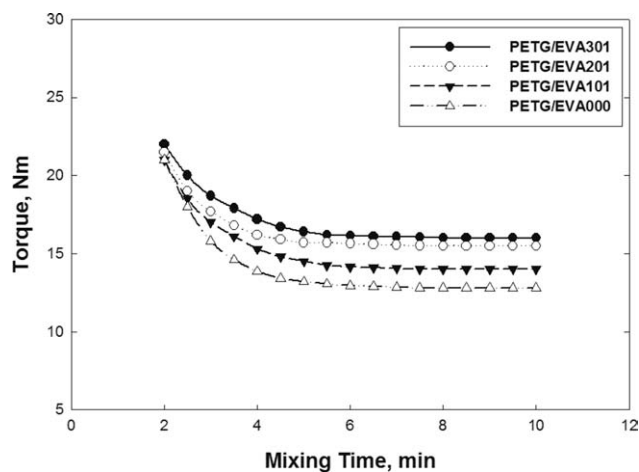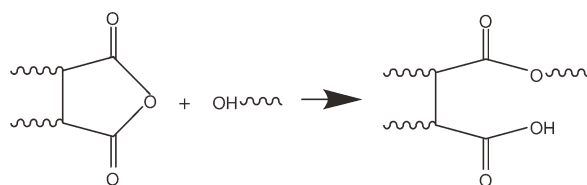
Figure 4 Variations of torque during mixing of PETG and mEVA (20 phr) with various MAH contents.

molecular weight resulted from reaction between functional group of PETG and a grafted MAH of MAH-g-EVA. It was reported that polymers with a reactive monomer such as MAH react with end group, —OH and —COOH, of PBT, PET, and Nylon as shown below.[7]

It can be assumed that EVA-g-MAH also could react with PETG due to its —OH and —COOH group.

The results of Molau solution test for PETG/uEVA (20 phr) and PETG/mEVA (20 phr) after 60 days were given in Figure 5. The phase separation was found in Figure 5(a), and EVA is presented in upper part with cloudy state, whereas PETG in bottom part is clear. Figure 5(b) also shows a similar behavior as (a), but EVA suspension is spread out through the whole tube. This may be due to chemical interaction between end groups of PETG and MAH of mEVA to keep from the phase separation. Yu et al.[26] reported that MAH grafting TPE (TPEg) would have a chemical linkage resulted from the reaction of MAH with the hydroxyl group of PETG during melt extrusion from PETG/TPEg blends study.

Figure 6 presents that the MFI results of PETG, EVA, and their blends. The mEVA shows significant lower value than uEVA, and this can be attributed to crosslinking reaction resulting in conversion of linear structure into a crosslink structure by the modification of EVA. While MFI of PETG/uEVA is

Figure 5 Molau test solutions consisting of 1,1,2,2-tetrachloroethane/phenol (4 : 6 by weight ratio) mixed solvents and the following blends: (a) PETG/uEVA (20 phr) and (b) PETG/mEVA (20 phr). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

close to that of PETG since EVA exists as dispersion state, PETG/mEVA has MFI close to mEVA. It is believed that there might have some interaction
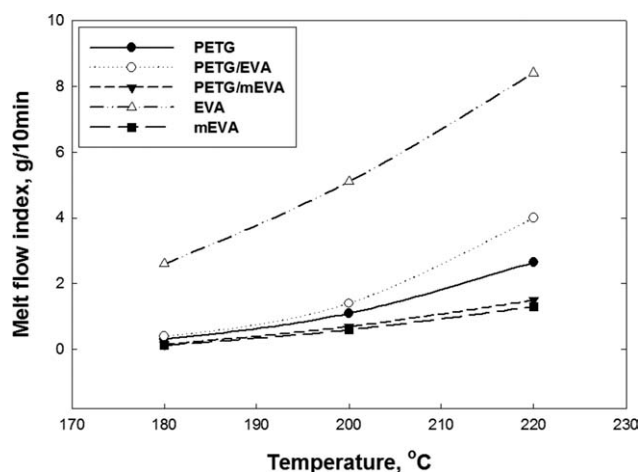

Figure 6 Melt flow index of PETG, EVA, and blends at different temperature.
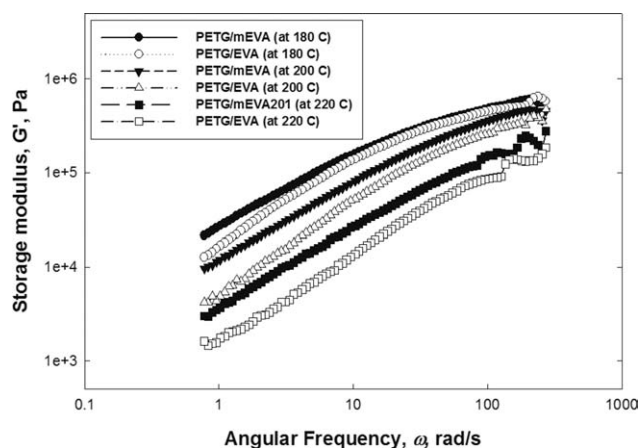
Figure 7 G' of PETG blends with uEVA and mEVA (20 phr) as function of angular frequency at different temperature.

between PETG and mEVA to increase of molecular weight. In case of PETG/EVA blends without MAH crosslinked by only DCP, the blend showed almost similar MFI to PETG/mEVA (Data not shown). This result supports that the mEVA is not the only factor that would affect to MFI of PETG/mEVA.

The rheological behavior at low frequency can be used to determine the effect of compatibilizer for polymer blends because polymer blends with the improved compatibility show higher dynamic storage modulus (G'). It is known that enhanced interfacial adhesion induced increase of the shear stress resistance in the blend.[36–40]

The G' of PETG blends with EVA at various temperatures with respect to angular frequency is given in Figure 7. At all temperature range, PETG/mEVA shows higher G' than PETG/uEVA at low frequencies. This can be attributed to increase of compatibility resulted from reaction of the grafted MAH with PETG lowering an interfacial tension of polymers.
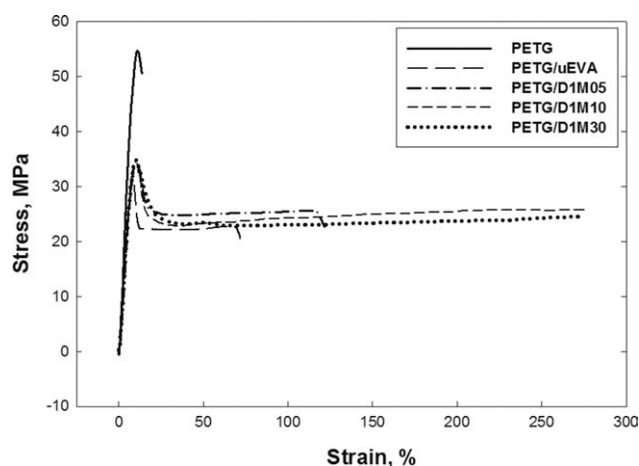

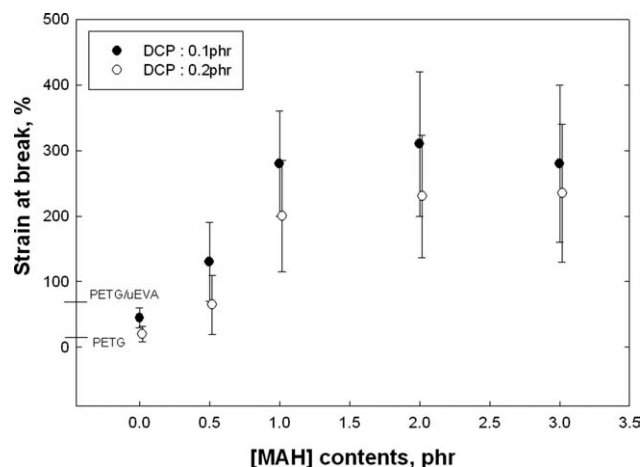**Figure 8** Stress–strain curve of PETG/EVA-g-MAH blends.

Figure 9 % strain of PETG blends with different content of EVA-g-MAH at break point.

The stress–strain curve for PETG, PETG/uEVA, and PETG/mEVA with various compositions was given in Figure 8. It was found that PETG itself showed a brittle nature without a plastic deformation, and yielding behavior was found after addition of uEVA and mEVA resulting in a plastic deformation. Over 250% strain was found in PETG blended with EVA having more than 1.0 phr of MAH. Consequently, the introduction of EVA in the blends led a plastic deformation of PETG, and the addition of MAH in EVA increased % strain. In addition, the % strain gradually did level off after 1.0 phr of MAH (Fig. 9). This could be attributed to increase of an interfacial adhesion force resulted from the reaction between end groups of PETG and MAH. It also was found that 0.2 phr of DCP decreases % strain compared with 0.1 phr in Figure 9, and this could be due to the crosslinking effect of EVA by DCP. As shown in Figure 10, the tensile strength and Young's modulus are not significantly changed overall regardless of MAH and DCP content. It can be seen that the increase of compatibility does not have a
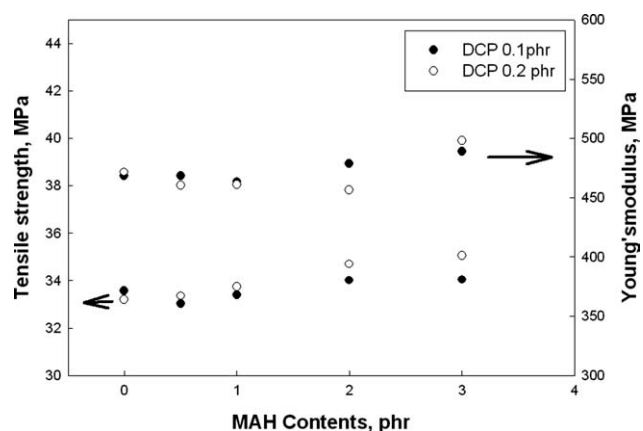

Figure 10 Tensile strength and Young's modulus of PETG blends with different content of EVA-g-MAH.
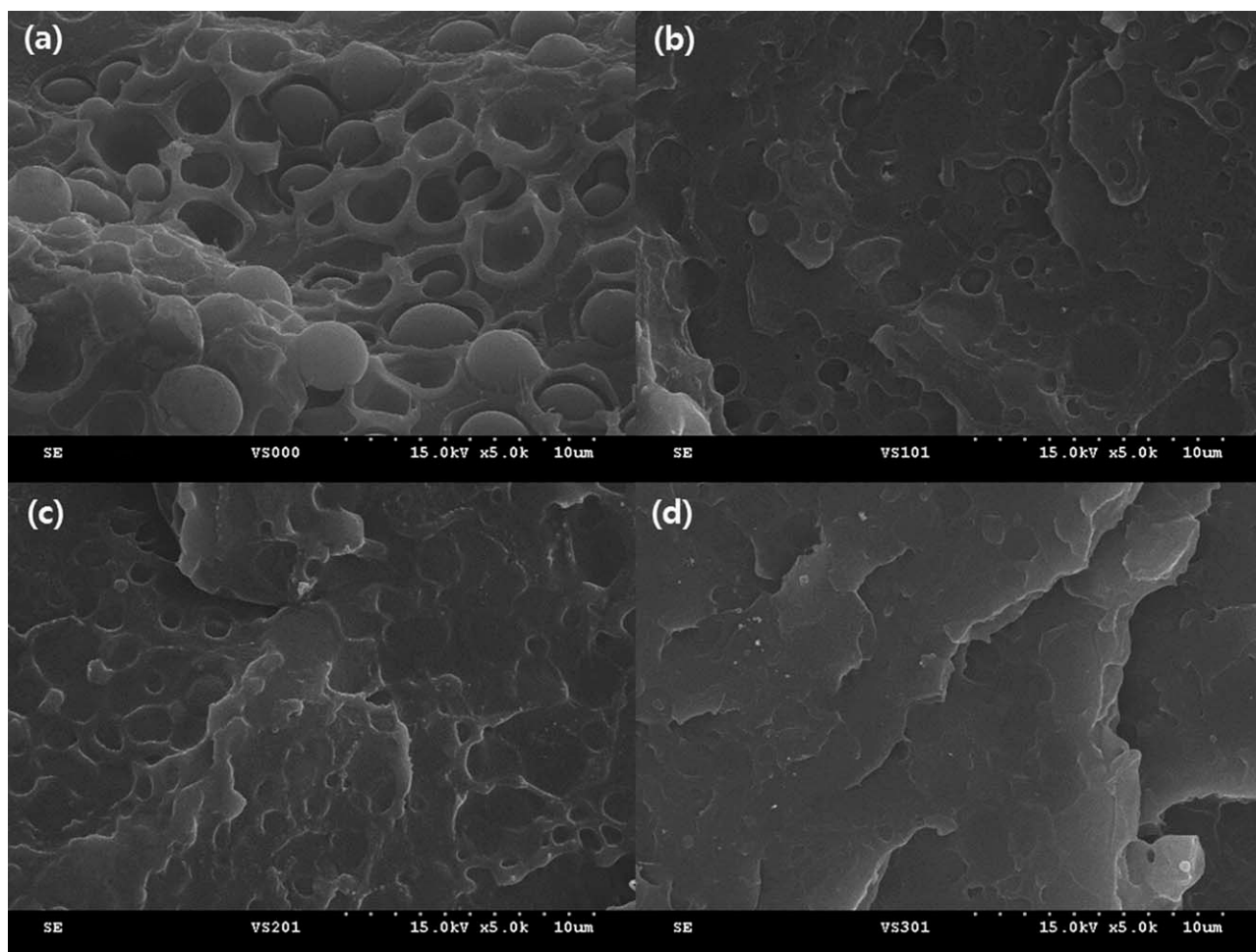
Figure 11 SEM micrograph of fractured surface of PETG blends with different content of EVA-*g*-MAH: (a) PETG/uEVA, (b) PETG/D1M10, (c) PETG/D1M20, and (d) PETG/D1M30.

considerable effect on the low strain tensile properties of the blends as in other previous studies.[41,42]

The mechanical properties of polymer blends significantly depend on the morphological structures of the blends. In Figure 11(a), the clear edges and cavities were observed between EVA and PETG matrix indicating poor interfacial adhesion. However, Figure 11(b,c) shows smaller cavity size, and Figure 11(d) presented better interfacial adhesion between the two phases. This could be attributed to reduced interfacial tension by reactive compatibilization resulting in enhancing the interfacial adhesion.

CONCLUSIONS

It was found that grafting reaction of MAH on to EVA could compete with crosslinking reaction of DCP during the modification through the mixing torque and gel content. Introduction of a small amount of MAH had great effect on reducing gel content by decreasing crosslink reaction. As MAH content increased, grafted MAH concentration increased, whereas the grafting yield decreased. It was confirmed that EVA-*g*-MAH acted as a reactive compatibilizer in the blend with PETG, and enhanced compatibility by reacting with the hydroxyl group, OH of PETG from the Molau solution test and rheological properties. Addition of EVA in the blend leads a plastic deformation of PETG, and MAH had great effect on enhance interfacial adhesion resulting in significant increasing of % strain; however, this increase of compatibility could not be changed significantly in low strain tensile strength and Young's modulus.

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