

Compatibilization of Polypropylene/Poly(acrylonitrile-butadiene-styrene) Blends by Polypropylene-Graft-Cardanol

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ABSTRACT: The polypropylene-graft-cardanol (PP-g-cardanol) was prepared by reactive extrusion with polypropylene (PP) and natural renewable cardanol which could increase the interfacial energy of PP and inhibit the degradation of PP during the process of reactive extrusion and usage. In this article, PP-g-cardanol and polypropylene-graft-maleic anhydride (PP-g-MAH) were used as compatibilizers of the polypropylene (PP)/poly(acrylonitrile-butadiene-styrene) (ABS) blends. PP/ABS (70/30, wt %) blends with PP-g-cardanol and PP-g-MAH were prepared by a corotating twin-screw extruder. From the results of morphological studies, the droplet size of ABS was minimized to 1.93 and 2.01 μm when the content of PP-g-cardanol and PP-g-MAH up to 5 and 7 phr, respectively. The results of mechanical testing showed that the tensile strength, impact strength and flexural strength of PP/ABS (70/30) blends increase with the increasing of PP-g-cardanol content up to 5 phr. The complex viscosity of PP/ABS (70/30) blends with 5 phr PP-g-cardanol showed the highest value. Moreover, the change of impact strength and tensile strength of PP/ABS (70/30) blends were investigated by accelerated degradation testing. After 4 accelerated degradation cycles, the impact strength of the PP/ABS (70/30) blends with 5 phr PP-g-cardanol decrease less than 6%, but PP/ABS (70/30) blends with 5 phr PP-g-MAH and without compatibilizer decrease as much as 12% and 32%, respectively. The tensile strength of PP/ABS (70/30) blends has a similar tendency to that of impact strength. The above results indicated that PP-g-cardanol could be used as an impact modifier and a good compatibilizer, which also exhibited better stability performance during accelerated degradation testing. © 2014 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2015**, *132*, 41315.

KEYWORDS: blends; compatibilization; degradation; mechanical properties; morphology

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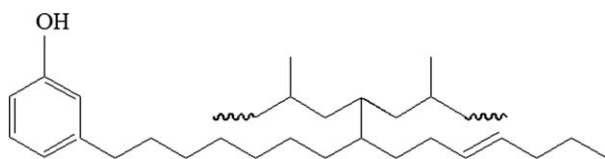
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INTRODUCTION

Blending modification is an effective method for obtaining the polymeric materials with excellent comprehensive performance. With the development of science and technology, the field of blends has rapidly expanded with a diversity of properties. The properties of blends are determined by the properties of every component, phase compatibility and adhesion at the interface and the distribution of the dispersed phase.^{1–8} Unfortunately, most polymer components appeared immiscibility with each other due to the distinction in surface adhesion, intermolecular interaction and viscoelastic index. To improve the compatibility and the properties of blends, a third component is added as a compatibilizer.^{9–12} Many researchers devote themselves to this field and obtain many significant results.^{13–16} Polyamide 6 (PA6)/poly(2,6-dimethyl-1,4-phenylene oxide) (PPO) blends compatibilized by poly(styrene-co-maleic anhydride) (SMA) were studied by Guo and Shen,¹⁵ they reported that the addition of SMA improved the effect of compatibilization resulted in improvement of the morphology and impact strength of

PA6/PPO blends. You et al.¹⁶ investigated polypropylene-graft-reduced graphene oxide (PP-g-rGO) as a novel compatibilizer for PP/polystyrene (PP/PS) immiscible polymer blends. They found that the average diameter for the dispersed PS phase was reduced and the tensile strength was enhanced by the addition of PP-g-rGo into PP/PS blends.

PP has many excellent chemical and mechanical properties, such as good chemical resistance, high softening points, good processibility and economic advantages. However, PP has an obvious deficiency of low impact strength. On the other hand, ABS with a good impact resistance is one of the most popular toughening modifier. PP toughened with ABS in the presence of compatibilizer has been investigated in recent years.^{17–21} The studies of PP/ABS blends with multiwall carbon nanotubes (MWNT) which was reported by Khare,¹⁹ have displayed that dispersed type or morphology structure refined by MWNT. From the studies by Patel et al.,²⁰ the use of PP-g-2-hydroxyethyl methacrylate (2-HEMA) improved the mechanical properties of PP/ABS blends. Hyung Gon Lee et al.²¹ had examined



Scheme 1. Chemical structure of PP-g-cardanol.

mechanical, morphological and rheological properties of PP/ABS with PP-g-MAH. They reported that the compatibility of PP/ABS blends increased with the addition of PP-g-MAH.

On the other hand, the degradation of PP is significant due to the tertiary carbon atom on PP chains. Our former study on PP-g-cardanol (Scheme 1)²² has showed that cardanol grafted onto PP could inhibit the light and heat degradation because the natural product of cardanol had the dual effect of electron donating and electron withdrawing due to the $p-\pi$ conjugated system of cardanol. Herein, using PP-g-cardanol as a compatibilizer on PP/ABS blends has studied in this article and in the reference of the PP-g-MAH. We expected that its performance stands comparison with PP-g-MAH. The morphology of brittle fractured surface and the properties of PP/ABS blends compatibilized by PP-g-cardanol are investigated by SEM, universal testing machine and rheometer equipped. Moreover, accelerated degradation is a common method under experimental conditions to estimating the normal service life of material.^{23–25} The mechanical properties by accelerating degradation of PP/ABS blends with PP-g-cardanol and PP-g-MAH have also studied in this article. This research provides useful theoretic guidance for the practical application of natural cardanol grafted onto PP.

EXPERIMENTAL

Materials

Isotactic polypropylene homopolymer powder (PPH-XD-045, Melt Flow Index (MFI) = 2.1–6.0 g/10 min) was purchased from Shandong Kairi Chemical Industry, China. Cardanol was purchased from a local market without further purification. The initiator dicumyl peroxide (DCP) was supplied by Sinopharm Chemical Reagent, China. It was dried in a dark room for a week before use. Polypropylene (PP-T30S, MFI around 3.0 g/10 min) was purchased from Fujian Union Petroleum Chemical, China. Poly(acrylonitrile-butadiene-styrene) (ABS-PA747, MFI around 12.0 g/10 min) was purchased from Chi Mei (Taiwan, China). Maleic anhydride-grafted polypropylene (PP-g-MAH, WJ-A02, MFI around 19.0 g/10 min, 1.1 wt % of maleic anhydride) was purchased from local market. Methyl ethyl ketone (MEK, CP grade) was purchased from Hengyang Kaixin.

Preparation of PP-g-Cardanol

The PP-g-cardanol was prepared by a corotating twin-screw extruder (Polylab Rheomix PTW24/28, Thermo Scientific, Germany). Precalculated weights of PP (XD-045), cardanol and DCP were premixed in a high-speed disintegrating machine (JFW-A, China), at 2600 rpm. The barrel temperature of extruder was 180°C and the screw speed of extruder was 50 rpm. The extrudate was cooled with water, followed by granulation. The grafting degree of products was measured by a UV-vis spectrometer (Lambda850, PerkinElmer).²⁶ The grafting

degree of PP-g-cardanol is 3.88%. The melt flow index (MFI) test (XRL-400 A, Chengde Precise Instrument Company, China) was performed at 190°C under a pressure of 2.16 kg. The MFI of PP-g-cardanol is around 5.0 g/10 min.

Preparation of PP/ABS Blends

PP (T30S), ABS and PP-g-cardanol were predried in a vacuum oven at 80°C for 24 h. Then, PP/ABS blends in a ratio 70/30 by weight were extruded by melt-mixing in a corotating twin-screw extruder (HAAKE, Polylab Rheomix PTW24/28, Germany). The compatibilizer of PP-g-cardanol at a ratio 0, 1, 3, 5, 7, and 9 parts per hundred parts resin by weight (phr) was added to the PP/ABS (70/30) blends. The barrel temperature ranging from 190 to 210°C along the feeding port to the die, and the screw speed of extruder was 50 rpm. Through water cooling and pelletizing, then gained blends. By change PP-g-cardanol to PP-g-MAH, and repeating the processes, the PP/ABS (70/30) blends with PP-g-MAH were prepared as the contrast group.

Morphology of Brittle Fractured Surface

The blends samples were fractured under cryogenic conditions and etched with MEK for 5 h for ABS extraction. Then, the latter was Pt-coated. The morphology of brittle fractured surface was examined by a JSM-7500F (Japan) scanning electron microscopy at 5 kV accelerating voltage.

Mechanical Properties

Samples were compression modeled by injection molding machine (JN55E, China) at 210°C for 5 min. Before molding, all samples were dried in a vacuum oven at 80°C for 24 h. The tensile strength and the flexural strength of PP/ABS (70/30) blends were measured by the LR5K universal testing machine of LLOYD British Company, according to the standards of GB/T1040-1992 and GB/T9341-2008, respectively. The crosshead speed was 20 mm min⁻¹. The notched impact strength of PP/ABS (70/30) blends was tested with a JJ-20 (China) impact-testing machine according to GB/T1043-1993. For each measurement, five samples were tested and averaged at room temperature.

Rheology Properties

The rheological properties of the PP/ABS blends were evaluated on an AR-2000 Rheometer in a parallel plate mode. The samples with a thickness of 2.0 mm and diameter of 25 mm were prepared by compression molding. The frequency sweeps ranged from 0.1 to 100 rad s⁻¹ at 210°C. Constant strain amplitude (1% strain) was used to ensure that all measurements were carried out within the linear viscoelastic range.

Accelerated Degradation of PP/ABS Blends

According to time-temperature equivalence principle, the degradation of samples at 90°C for 72 h (1 cycle) is equal to 1 year at room temperature.²⁷ The accelerated degradation of PP/ABS (70/30) blends had completed in an oven, and then measured the mechanical properties after 1 to 4 cycles. The test methods reference to the Mechanical Properties.

RESULTS AND DISCUSSION

Morphology

Figure 1 showed the SEM morphology of brittle fractured surfaces of PP/ABS blends with the PP-g-cardanol content of 0, 1, 3,

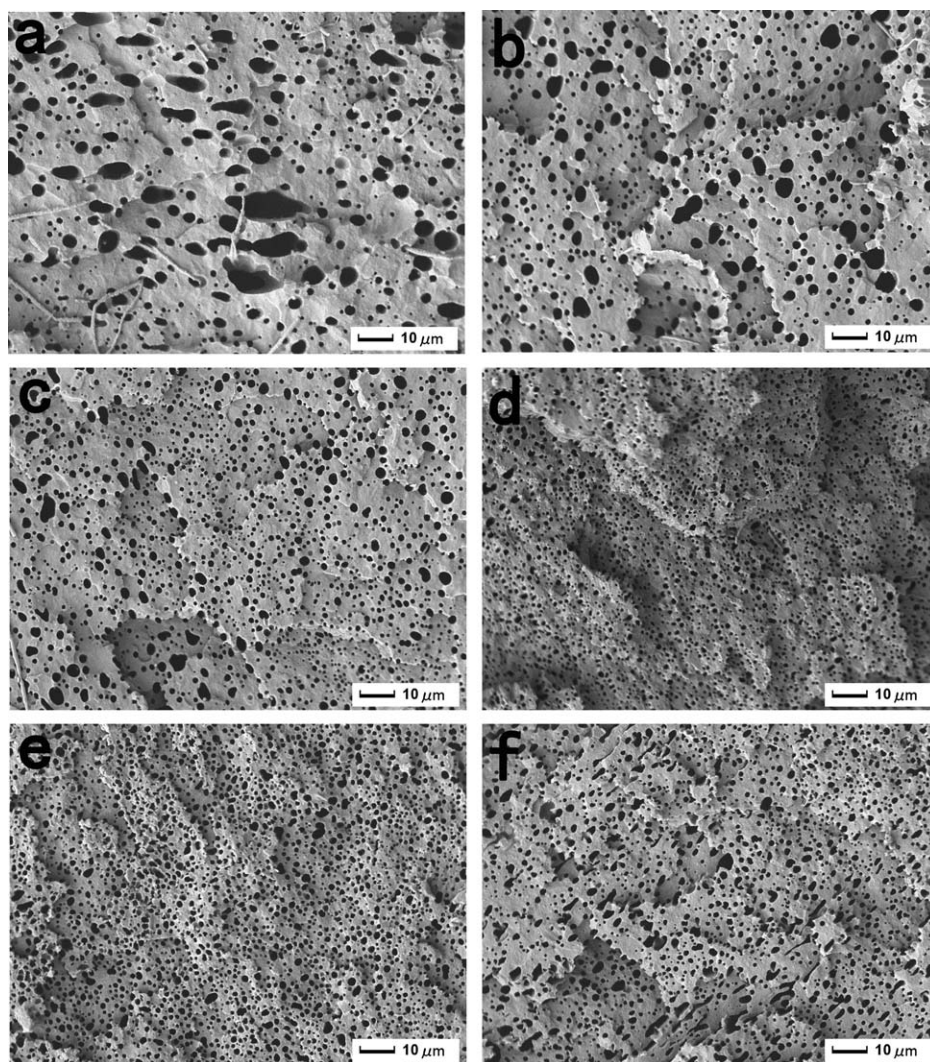


Figure 1. SEM micrographs of the PP/ABS (70/30) blends with PP-g-cardanol content: (a) 0 phr; (b) 1 phr; (c) 3 phr; (d) 5 phr; (e) 7 phr; (f) 9 phr.

5, 7, and 9 phr. The ABS particles had been removed by MEK perfectly in order to observe the morphological feature. From Figure 1, PP-rich component as the continuous phase and ABS as the dispersed phase of PP/ABS (70/30) blends formed droplet morphology, larger cavities also developed due to the erosion of ABS from the PP/ABS (70/30) blends. The fractured surface of PP/ABS (70/30) blend without compatibilizer is smooth and a brittle surface is observed, suggesting that the interfacial adhesion between PP and ABS phases is poor [Figure 1(a)]. However, a marked difference is observed in the morphology of PP/ABS (70/30) with the addition of PP-g-cardanol [Figure 1(b–f)]. It is observed that the droplet size of ABS of PP/ABS (70/30) blends decrease from 4.09 to 3.43, 2.83, 1.93, and 2.48 μm when the PP-g-cardanol is added in the content of 0, 1, 3, 5, and 7 phr, respectively (shown in Table I). Especially, the droplet size of the ABS shows minimum value (1.93 μm) when the addition of PP-g-cardanol content up to 5 phr. It may be result of compatibilization efficiency of increased the interfacial layer thickness and improved the interfacial adhesion between the PP and ABS phases related to the phenyl, phenolic hydroxyl and

long alkyl chains of PP-g-cardanol similar to the molecular structure of ABS and PP. A further increase of PP-g-cardanol content to 7 and 9 phr resulted in an increase in the droplet size of ABS to 2.48 and 2.79 μm , respectively. This phenomenon was similar to the reports by Patelet al.¹⁷ and Lee et al.²¹ Here, especially, a non-spherical of dispersed droplets is observed when the PP-g-cardanol content up to 9 phr [Figure 1(f)]. It is suggest that compatibilizer efficiency is related with compatibilizer content. At a low level of compatibilizer content, efficient compatibilizer reduces interfacial tension between the phases of PP/ABS blends and prevents dispersed droplets coalescence.^{21,28} As a result, the compatibility between PP and ABS was improved and the dispersed droplet size was reduced. With the increases of PP-g-cardanol content, the number of dispersed droplets per volume unit increases and the distance between dispersed droplets decreases at a constant volume fraction [Figure 1(b–d)]. However, when the compatibilizer content become over saturated, the increase of collision probability of dispersed droplet by the shear flow resulted in an increase of flow-driven coalescence,²⁹ which leading to weak compatibilizer efficiency

Table I. Mechanical Properties and Average Diameter of Droplet Size of the PP/ABS (70/30 wt %) Blends with the Compatibilizer of PP-g-Cardanol or PP-g-MAH

Compatibilizer	Tensile strength (MPa)	Impact strength (kJ m^{-2})	Flexural strength (MPa)	Average diameter (μm)
0	36.51	3.20	43.32	4.09
1 phr PP-g-cardanol	37.63	3.28	44.19	3.43
3 phr PP-g-cardanol	40.89	3.32	44.91	2.83
5 phr PP-g-cardanol	42.54	3.43	45.25	1.93
7 phr PP-g-cardanol	41.95	3.37	45.18	2.48
9 phr PP-g-cardanol	41.16	3.36	44.94	2.79
1 phr PP-g-MAH	38.83	3.33	44.69	3.40
3 phr PP-g-MAH	40.52	3.43	45.20	3.02
5 phr PP-g-MAH	42.26	3.45	45.96	2.32
7 phr PP-g-MAH	42.66	3.48	46.01	2.01
9 phr PP-g-MAH	39.63	3.42	45.87	2.64

and the increases of dispersed droplet size. This could explain the increase of dispersed droplet size and the change in morphology of PP/ABS (70/30) blends when the PP-g-cardanol content up to 7 and 9 phr. From the results of Figure 1, it is suggested that PP-g-cardanol is an effective compatibilizer of the PP/ABS (70/30) blends, especially when the PP-g-cardanol is added in the content of 5 phr.

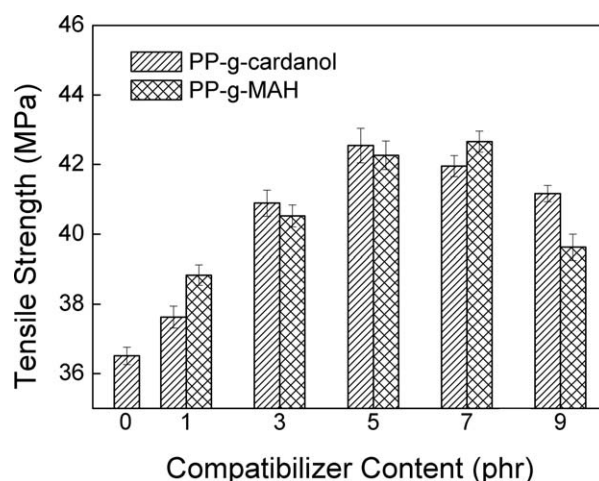
From Table I, compared to the PP/ABS (70/30) with PP-g-cardanol, a similar tendency of the droplet size of ABS of PP/ABS (70/30) blends with PP-g-MAH was observed. The droplet size of ABS is decrease from 4.09 to 2.01 μm when the addition of PP-g-MAH up to 7 phr. However, the PP-g-MAH is added in the content of 9 phr, the droplet size of ABS is increase to 2.64 μm . Hyung Gon Lee et al.²¹ had concluded that PP-g-MAH is an effective compatibilizer of PP/ABS blends due to the dipolar interactions between maleic anhydride group and polar nitrile group in the ABS. From Figure 1 and Table I, it is suggest that PP-g-cardanol is a compatibilizer as effective as PP-g-MAH of the PP/ABS (70/30) blends.

Mechanical Properties

Figure 2 showed the tensile strength of PP/ABS (70/30) blends with 0, 1, 3, 5, 7, and 9 phr PP-g-cardanol or PP-g-MAH. It was showed that the tensile strength of PP/ABS (70/30) blends increases with the increasing PP-g-cardanol or PP-g-MAH content. When the PP/ABS (70/30) blend with PP-g-cardanol content up to 5 phr, the tensile strength increases from 36.51 to 42.54 MPa, while the blend with PP-g-MAH increases to 42.66 MPa when the content of PP-g-MAH up to 7 phr. It may be due to an increase in the interfacial adhesion between PP and ABS resulted of compatibilization efficiency by the addition of PP-g-cardanol or PP-g-MAH, and the tensile strength of PP/ABS (70/30) blends with PP-g-cardanol or PP-g-MAH was observed to be higher compared to the PP/ABS (70/30) blends without any compatibilizer. However, a further increasing of PP-g-cardanol or PP-g-MAH content, the tensile strength of PP/ABS (70/30) blends decrease apparently may be due to the solubilizing effect²¹ of the comptibilizer in the PP/ABS blends.

Figures 3 and 4 illustrated the impact strength and the flexural strength of the PP/ABS (70/30) blends with 0, 1, 3, 5, 7, and 9 phr PP-g-cardanol or PP-g-MAH, respectively. Similar to the tensile strength, the impact strength and the flexural strength of the PP/ABS (70/30) blends increase with increasing PP-g-cardanol or PP-g-MAH content. When the adding PP-g-cardanol content up to 5 phr, the impact strength and flexural strength of PP/ABS (70/30) blends show maximum values of 3.43 kJ m^{-2} or 45.25 MPa, respectively, while the PP/ABS (70/30) blends with the PP-g-MAH content up to 7 phr show maximum values of 3.48 kJ m^{-2} or 46.01 MPa, respectively. The flexural strength of the PP/ABS (70/30) blends with PP-g-cardanol is slight lower than that with PP-g-MAH may be due to the MFI of PP-g-cardanol (around 5.0 g/10 min) much lower than that of PP-g-MAH (around 19.0 g/10 min).

From Figures 2–4, it was observed that the tensile strength, impact strength and flexural strength of PP/ABS (70/30) blends increased with the increasing PP-g-cardanol or PP-g-MAH

**Figure 2.** Tensile strength of the PP/ABS (70/30) blends with the compatibilizer of PP-g-cardanol or PP-g-MAH.

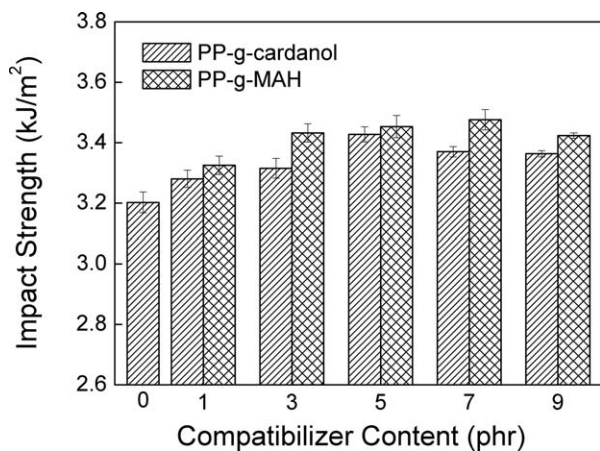


Figure 3. Impact strength of the PP/ABS (70/30) blends with the compatibilizer of PP-g-cardanol or PP-g-MAH.

content up to 5 or 7 phr, respectively. The increase of mechanical properties of PP/ABS blends may be due to the toughening effect^{20,21} of ABS in the PP/ABS blends result of the increase of compatibility by addition of the PP-g-cardanol or PP-g-MAH. The values of mechanical properties and average droplet size of the PP/ABS (70/30) blends with the PP-g-cardanol or PP-g-MAH are shown in Table I. From Table I, it is showed that minimum values of the droplet size are observed when the tensile strength, impact strength, and flexural strength of PP/ABS (70/30) blends show maximum values that the PP-g-cardanol or PP-g-MAH was added in the content of 5 or 7 phr, respectively. Above results suggest that PP-g-cardanol improved compatibility between PP and ABS phases, and play the role as a good impact modifier and compatibilizer as effective as PP-g-MAH.

Rheology Properties

Figure 5 illustrated the complex viscosity of PP/ABS (70/30) blends with the PP-g-cardanol in the content of 0, 3, 5, and 7 phr, respectively. From Figure 5, it can be seen that the complex viscosity of the PP/ABS (70/30) blends increase at first and then decrease with increasing the PP-g-cardanol content. Especially,

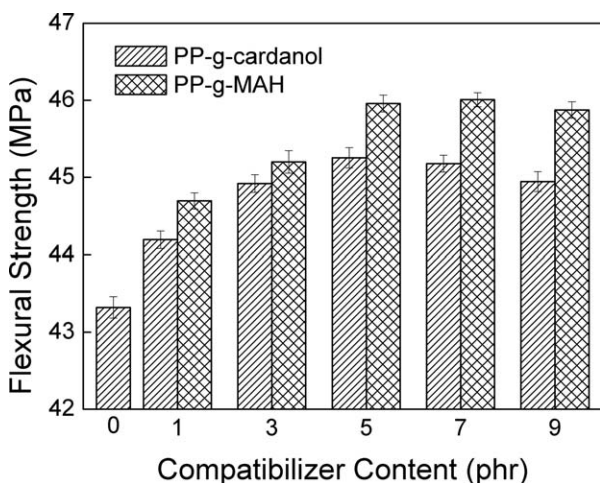


Figure 4. Flexural strength of the PP/ABS (70/30) blends with the compatibilizer of PP-g-cardanol or PP-g-MAH.

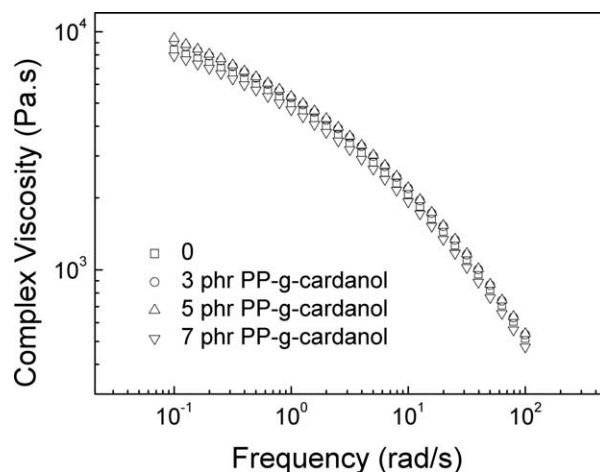


Figure 5. Complex viscosity of PP/ABS (70/30) blends with the compatibilizer of PP-g-cardanol.

the complex viscosity of the PP/ABS (70/30) blends shows the highest value when the adding PP-g-cardanol content is up to 5 phr. It is due to the improved compatibility of the PP/ABS (70/30) blends by adding PP-g-cardanol. It also observed that the complex viscosity of the PP/ABS (70/30) blends changes more significant at the low-frequency region. The complex viscosity at low-frequency region is related to the interfacial properties of the polymer blends.^{18,30,31} In this study, it is may be due to the interfacial adhesion of PP/ABS blends was improved by adding PP-g-cardanol leading to a longer time relaxation. However, the complex viscosity of the PP/ABS (70/30) blend decreases when the PP-g-cardanol content up to 7 phr, it is may be due to the dispersed particles coalescence result of the excessive PP-g-cardanol.

Accelerated Degradation

Figures 6 and 7 showed the impact strength and the tensile strength of PP/ABS (70/30) blends after accelerated degradation. It reveals that degradation occurred after accelerated degradation cycles. From Figure 6, it observed that the impact strength of PP/ABS (70/30) blends without compatibilizer decrease

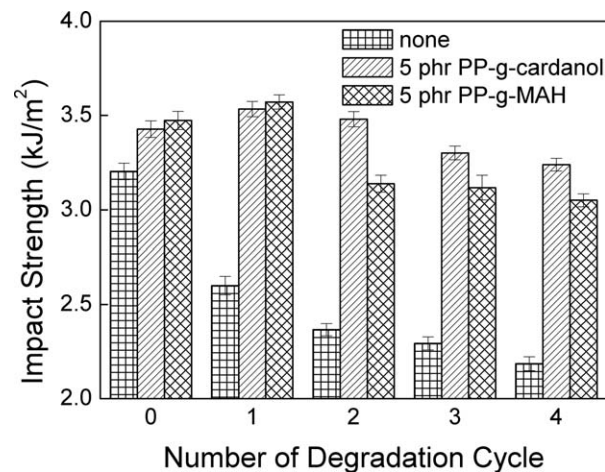


Figure 6. Impact strength of the PP/ABS (70/30) blends with the compatibilizer of PP-g-cardanol (5 phr) or PP-g-MAH (5 phr) after accelerated degradation.

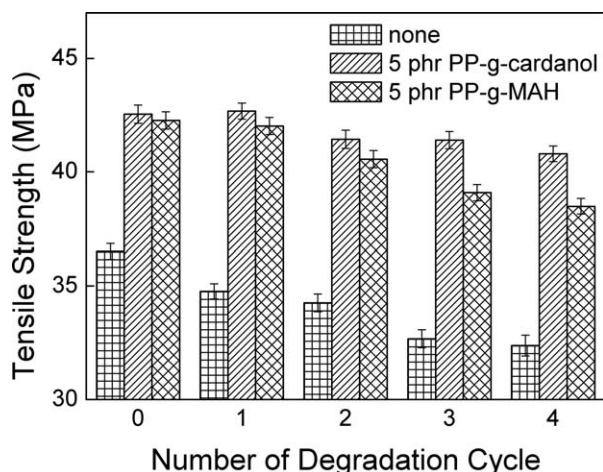


Figure 7. Tensile strength of the PP/ABS (70/30) blends with the compatibilizer of PP-g-cardanol (5 phr) or PP-g-MAH (5 phr) after accelerated degradation.

dramatically only after experiencing 1 accelerated degradation cycle and decrease as much as 32% after four cycles. Because of the unsaturated bonds of polybutadiene chain segment in ABS, it became the weakest component in ABS. During the process of accelerated degradation, the broken bond of unsaturated polybutadiene chain segments³² and the broken of PP main chain may result in a brittle surface layer, which may lead to reduce the adhesion at the phase surface. As a result, the stress was concentrated at the phase surface of weak adhesion results in mechanical properties decreased dramatically. Although the impact strength of PP/ABS (70/30) blends with the compatibilizer of PP-g-cardanol or PP-g-MAH (5 phr) decrease with an increase of degradation cycles, it's decrease not significantly even experiencing 4 cycles compared to PP/ABS (70/30) blends without compatibilizer. This may be due to the adding compatibilizer (PP-g-cardanol or PP-g-MAH) improves the compatibility of the PP/ABS (70/30) blends and slows degradation.²³ The tensile strength of PP/ABS (70/30) blends has a tendency similar to that of the impact strength (Figure 7).

From Figures 6 and 7, it is also observed that the PP/ABS (70/30) blends with PP-g-cardanol content of 5 phr has a good mechanical properties than that of PP-g-MAH (5 phr) after accelerated degradation. Especially, the impact strength of PP/ABS (70/30) blends with PP-g-cardanol (5 phr) decrease <6%, but with PP-g-MAH (5 phr) decrease as much as 12% after 4 accelerated degradation cycles. The results suggest that PP/ABS (70/30) blends with PP-g-cardanol (5 phr) possess a better resistance to degradation than do that PP-g-MAH (5 phr). A possible explanation for the impact strength and the tensile strength differences between PP/ABS (70/30) blends with PP-g-cardanol and PP-g-MAH after accelerated degradation may be related to the dual effect of electron donating and electron withdrawing of PP-g-cardanol due to the p- π conjugated system of cardanol.²²

CONCLUSIONS

The compatibilization of PP-g-cardanol on PP/ABS blend is apparent from the measurements of SEM, mechanical properties

and rheology properties. From the morphological studies of the PP/ABS (70/30) blends, the average diameter of droplet size of ABS showed minimum value of 1.93 μm with the addition of PP-g-cardanol content 5 phr. Second, the improvement of the tensile strength, the impact strength and the flexural strength of PP/ABS (70/30) blends related to the toughening effect of ABS due to the increase of compatibility between PP and ABS by adding the PP-g-cardanol. Third, the complex viscosity of the PP/ABS (70/30) blends with 5 phr PP-g-cardanol shows the highest values due to the increase of interfacial adhesion between PP and ABS phases by adding PP-g-cardanol. Fourth, the compatibilizers of PP-g-cardanol or PP-g-MAH are also beneficial for the stability performance of the PP/ABS (70/30) blends during accelerated degradation testing. Meanwhile, owing to the stabilization of phenolic groups on the radical groups, PP-g-cardanol shows more excellent stability performance than PP-g-MAH. This kind of natural product grafted on synthesis polymer is expected to increase the interfacial adhesion and stability performance.

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