Surface Functionalization of Polypropylene by Entrapment of Polypropylene-*grafted*-Poly(ethylene glycol)

Hanjia Chen,[1] Xuhua Shi,[1] Yafei Zhu,[2] Yi Zhang,[3] Jiarui Xu[3]

[1]Department of Chemistry, School of Science, Shantou University, Shantou 515063, China
[2]Instrument and Testing Center, Sun Yat-sen University, Guangzhou 510275, China
[3]Key Laboratory for Polymeric Composite and Functionality Materials of Education, School of Chemistry and Chemical Engineering, Materials Science Institute, Sun Yat-sen University, Guangzhou 510275, China

ABSTRACT: A surface functionalization polypropylene was prepared by entrapment a copolymer of polypropylene-*grafted*-poly(ethylene glycol) into polypropylene. The effects of structure of copolymer, contact dies, and content of modifiers were studied. The results of attenuated total reflection infrared spectroscopy(ATR-FTIR) and contact angle measurements indicated that PP-*g*-PEG could preferably diffuse onto the surface and effectively increase the hydrophilicity of PP. PPw-*g*-PEG with lower PEG contents, lower molecular weight of PPw and PEG had better selective enrichment on the surface of PP blend film. By grafting of PEG-OH onto the MPP, PP macromolecular surface modifier with better solvent-resistance than that of PEG can be achieved. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 114: 2461–2468, 2009

Key words: polypropylene; macromolecular surface modifier; ATR-FTIR; polypropylene-*grafted*-poly(ethylene glycol)

INTRODUCTION

Blending with surface modifier is widely recognized as a potential technique to overcome the polypropylene's low surface energy and increase the properties of dye, adhere, coat, or compatibility with polar polymers of products of polypropylene(PP).[1–3] Macromolecular surface modifiers have more interaction between the surface modifier and host polymer to anchor to the host polymer and keep their lasting effectiveness. However, their structure should be designed carefully so as not to hinder the surface modifier to preferred-diffuse to the surface and lower the efficiency of surface modifiers.[4] As far as our knowledge, only little attention was paid to PP surface modification by blending with macromolecular surface modifiers, probably due to the complicated synthesis process of the modifiers.[5–9] For example, Bergbreiter[1] had used boron-promoted radical polymerization and a vinyl-terminated PP oligomer to synthesize block cooligomers of propylene and tert-butyl acrylate to functionalize the surface of PP, but the preparation dealt with unsaturated PP and its hydroboration or oxidation, which was not very suitable to large-scaled production.

Poly(ethylene glycol)(PEG) is a typical hydrophilic biopolymer, its chain was often introduced to polymers to increase their hydrophilic or biological-philic surface. In our prior research,[10] we have found that PEG can be enriched on the surface of PP and improve its hydrophilicity. However, due to the weak interaction between PEG and PP, PEG was found to be detracted easily from the surface when the modified material was contact with water and other polar chemical solvents. In this study, a series of PP-*g*-PEG with different structures were synthesized and blended with PP to attend the effect of structure and contact die on surface enrichment of additives by determining the composition on surface and in bulk by ATR-FTIR and contact angles, and the solvent-resistance of modified film was also investigated to filtrate the surface modifier with appropriate structure.

EXPERIMENTAL

Materials

Polypropylene was supplied by India Reliance, H030SG, MI = 3.5. Polypropylene-*grafted*-PEG was prepared by coupling polypropylene-maleic anhydride copolymer with hydroxyl-terminated PEG in xylene.[11] Other reagent-grade chemicals such as xylene and acetone were used as received without further purification.

General procedure for film casting

PP and macromolecular surface modifiers were blended in xylene by heating the mixture to 140°C for 30 min. After distilled off xylene in a vacuum oven, the films were obtained by melt-cast in a stainless steel mold at 190°C. The thickness of films were about 80–100 μm.

Determination of ATR-FTIR and IR

The surface composition of the blending film was investigated by ATR-FTIR using a Nicolet 210 FTIR spectrometer with a variable-angle multiple-reflection ATR accessory, which allowed the external angles of incidence to be continuously varied from 30 to 70°. The ATR internal reflection elements used was a Zn-Se crystal. Typical ATR spectra were obtained at a crystal with 45° over a range of 250–4000 cm^{-1}. IR spectra were also recorded using a Nicolet 210 FTIR spectrometer with a scan number of 32.

Contact angle measurements

Contact angles of the film surface were determined using a Dataphysics SCA-20 contact angle goniometer at ambient temperature. All measurements were preformed using the sessile drop method and made with drops of 1 μL distilled water after about 15 s. The reported values were the average of eight measurements at various places on the same film sample. The values of contact angle measurements varied by ±3°.

Scanning electron microscopy (SEM) observations

A JSM-6330F SEM was used to study the appearance of modified PP after the modifiers of film were eroded by a mixture of potassium dichromate and concentrated sulfuric acid.

Solvent-resistance experiment

The modified films were dipped in acetone for an enactment time at room temperature. The composition of modifier on the film surface was determined by ATR-FTIR after removing off the solvent.

RESULTS AND DISCUSSION

Thermal stability was an issue for polymer processing and product durability. In our prior research,[11] we have demonstrated that the initial degradation temperatures of PP-g-PEG all exceeded over 250°C, higher than the processing temperature for PP, indicating that the copolymers would be suitable for the surface modification of PP in terms of the thermal stability. We also revealed that the copolymers showed a melting peak similar to that reported for PP. It was important that the surface modifier could form similar crystalline structure with the host polymers so that the surface modifier can incorporate with the host polymer to avoid losing off easily from the surface.

To evaluate the surface enrichment, ATR-FTIR and FTIR measurements were used to obtain quantitative information about the composition on surface and in bulk of blends, respectively. The band area ratio of PP-g-PEG and PP is about 1103 cm^{-1}, used to express the concentration of PP-g-PEG, can be expressed as followed[11]:

$$R = \frac{7.8439 \times (0.069898 \times A_{972} - A_{1103})}{A_{1103}} \quad (1)$$

Effect of concentration on surface enrichment

Modifier enrichment on polymer surface was affected by many factors such as modifier structure, molding temperature, interface, and modifier concentration. In improper instance, modifier with outstanding surface enrichment performance may not exhibit any interface congregation. Therefore, before we discussed the effect of modifier structure on surface enrichment, which of conditions suitable the modifier to enrich on the surface of host polymer should be determined. In this study, four kinds of PP-g-PEG with different structures were selected to study the effect of modifiers' concentration on surface enrichments, their structures in detail were listed in Table I.

Based on the ATR-FTIR principle, a surface layer of about 3 μm would be involved.[12] The results were showed in Figures 1 and 2. It could be evident that the concentration of PP-g-PEG on the surface was higher than that in the bulk, especially for the

### TABLE I
The Structure of PPw-g-PEG

No.	PPw-g-PEG-1	PPw-g-PEG-2	PPw-g-PEG-3	PPw-g-PEG-4
Molecular weight of PPw	5000	5000	9000	15000
Graft ratio/%	25.1	35.8	9.73	5.34

Molecular weight of PEG side chain is 750.
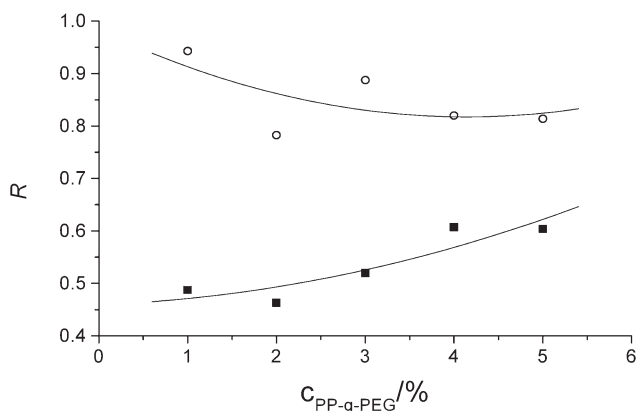
Figure 1 The peak area ratio of PP blends film at different contents of PPw-*g*-PEG-1. Footers: ○: surface; ■: bulk.

samples with lower loading of PP-*g*-PEG. At higher overall loadings, the PEG contents on the surface corresponded more closely to that expected for a normal distribution of PEG throughout the film, although the PEG content on the surface in our experiments always exceeded over that in bulk. However, this effect was most pronounced at weight percents of copolymer of less than 2 wt %. This indicated that PP-*g*-PEG can preferred diffuse to the surface of the blends and can be used as a surface modifier. The result was due to the induction of high-energy contact die. The graft copolymers of PP-*g*-PEG were amphiphilic, the PPw main chain was compatible with the host polymer, but the side chain of PEG can be separated from phases of PP. The graft copolymer was a high energy composition with a surface free energy of 44.0 dyn/cm, when compared with 29.0 dyn/cm of PP in the blends. Owed to the induction of high energy steel die whose surface free energy is 1200 dyn/cm, when the film was melt-cast in contact with steel, the copolymers would preferentially migrate to the interface so that the system can be in lowest energy state.
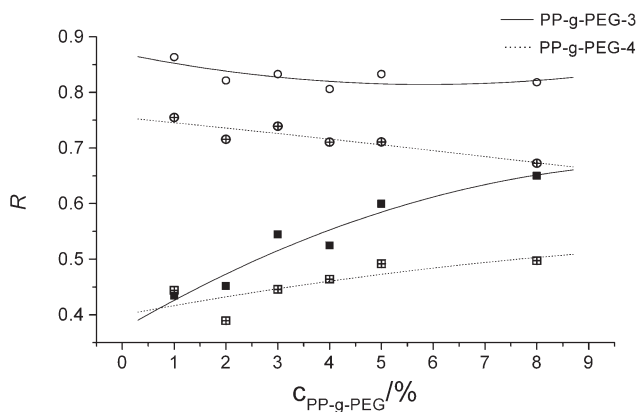
Furthermore, PP is a high crystalline polymer, when it was crystallizing, the copolymer was ejected from the crystal lattice of PP, which can further increase the surface selectivity of modifiers.

The composition of modifier on the surface of blend film and the efficiency of modifiers decreased as increasing the loading of graft copolymer. With increasing the concentration of copolymer, the interaction among molecules of PP-*g*-PEG increased, which enlarged the phase domains of PP-*g*-PEG and hindered the copolymer immigrate from inter of film. Therefore, the extent of segregation of these copolymers to the edges of the film decreased as the weight percent of the copolymers in the host PP increased.
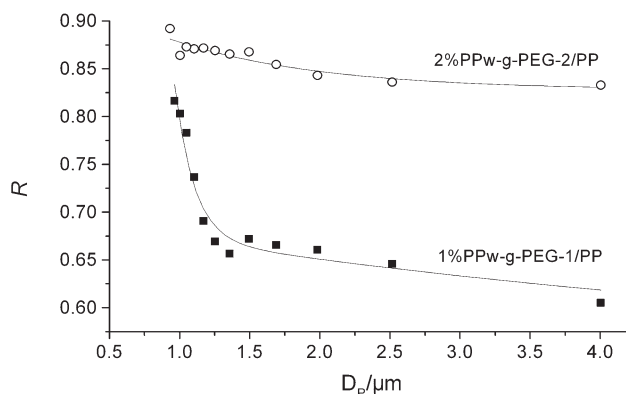
The selectivity of copolymer was also influenced by their structures. Although all the four kinds of PP-*g*-PEG had exhibited their strong surface enrichment, the degree of excess of copolymer and their tendency with percent were different. The result was due to the difference of structure and the phase segregation in the blends.

To further confirm that we had more PEG at low loadings on the surface, we performed depth studies on the blend films. The depth of penetration of the excursion wave in ATR spectra was given by eq. (2).[12]

$$d_p = \frac{\lambda}{2\pi n_1 \left[\sin^2 \theta - (n_2/n_1)^2\right]^{\frac{1}{2}}} \qquad (2)$$

where λ is the wavelength of the radiation in microns; d_p is the depth of penetration of the IR radiation; n_1 and n_2 are the refractive of the ATR crystal and polymer film, respectively; and θ is the angle of incidence of the IR beam on the ATR crystal.

On the basis of this equation, the depth of penetration for different angles and for the wavelengths of interest was calculated. The results were shown in Figure 3. The peak area ratio did increase as the depth of analysis decreased, especially for the film of 1 wt % PP-*g*-PEG-1/PP blend. Most of the

Figure 2 The peak area ratio of PP blends film at varying percent of PPw-*g*-PEG. Footers: ○: surface; ■: bulk.

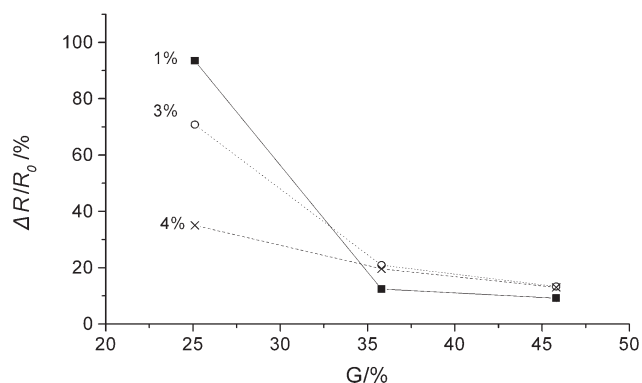**Figure 3** Surface concentration gradient of PPw-*g*-PEG in blends of PPw-*g*-PEG/PP.

copolymers were distributed in the outer of 1.5 μm. When compared with 1 wt % PP-g-PEG-1/PP blend film, although there was similar depth distribution for 2 wt % PP-g-PEG-2/PP blend, the slope of curve evidently decreased as a result of low selectivity of PP-g-PEG-2 at relatively high contents of graft copolymer in blends. This was consistent with the result of peak area ratios at different percent of PP-g-PEG in blends.

Effect of contact dies on surface enrichment

To investigate the effect of contact die on surface enrichment of PP-g-PEG, three different contact dies were chosen to prepare the blend films and determine their composition on surface and in bulk. The results were listed in Table II. The strong enrichment of PP-g-PEG at the high-energy steel or polyimide interface in melt-cast films from PP/PP-g-PEG is presumably driven by the decrease in the interfacial energy caused by the graft copolymer. The compositions of PP-g-PEG on surface exceeded those in bulk. In contrast to films formed in steel or polyimide, due to the induction of low surface free energy of interface of poly(tetrafluoroethylene), whose surface free energy is only 19.4 dyn/cm, PP was induced to immigrate to the interface of poly(tetrafluoroethylene), the content of PP-g-PEG on surface was lower than that in bulk.

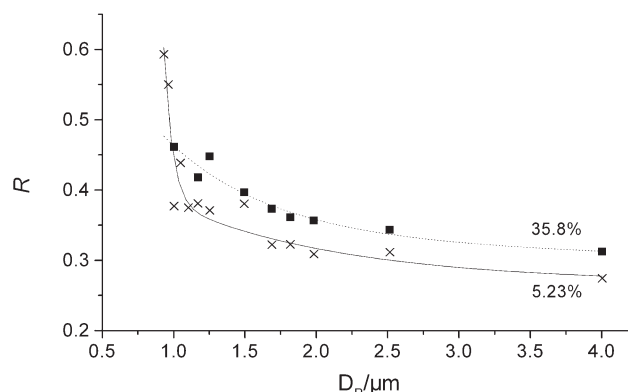### Effect of graft ratio of PP-g-PEG on surface enrichment

PPw-g-PEG is a comb-like amiphiphilic copolymer, the distribution and density of polar side chain inevitably influence the phase domains of copolymer in blend, further effect the modifier diffusion and enrichment on the surface. In this study, three kinds of PPw-g-PEG with similar molecular weight of PPw and PEG were chosen to compare their surface selectivity. The results shown in Figure 4 indicated that copolymer with lower graft ratio had higher surface enrichment, especially at the percent of 1 wt %. The surface selectivity of copolymer with 25.1% PEG content was about nine times of copolymer with

Figure 4 The influence of PEG content in PPw-g-PEG on surface excessive degree in PPw-g-PEG/PP blends film.

35.8% PEG content. The effect of PEG content in copolymer on its selectivity may be related to the ability of phase-separated. Higher density of side chain tended to form bigger phase domains and hindered the copolymer migrate to the interface similar to that in blends as compatilizer. Another result from Figure 4 was that the effect of PEG content in copolymer on surface enrichment at lower percent of PP-g-PEG seemed more profound than that at higher loadings. The surface selectivity of copolymer with 25.1% PEG content was only three times that of 45.8% at loadings of 4 wt % PP-g-PEG, while the value was about nine times at loadings of 1 wt % PP-g-PEG.

The higher surface selectivity of copolymer with lower PEG content was also confirmed by its depth distribution in Figure 5. The depth gradient was more obviously for PP-g-PEG with 5.23% PEG content than that with 35.8% PEG content. The copolymer was mainly distributed in the outer of 1.0 μm. This indicated that PP-g-PEG with lower PEG content had better enrichment by inducing of high-energy interface at lower loadings.

TABLE II
The Peak Area Ratio of PPw-g-PEG/PP Blends Under Different Contact Dies

M/R/dies	Fe	PI	Rv[a]	PTFE
600	0.5353	0.5724	0.5183	0.3452
750	0.5763	0.5074	0.4543	–
1000	0.71493	0.6319	0.5207	0.5512
2000	0.60223	0.4718	0.4438	0.5053

[a] In bulk.

Figure 5 Surface concentration gradient of PPw-g-PEG with different PEG contents in blends of PPw-g-PEG/PP formed under the contact die of PI.
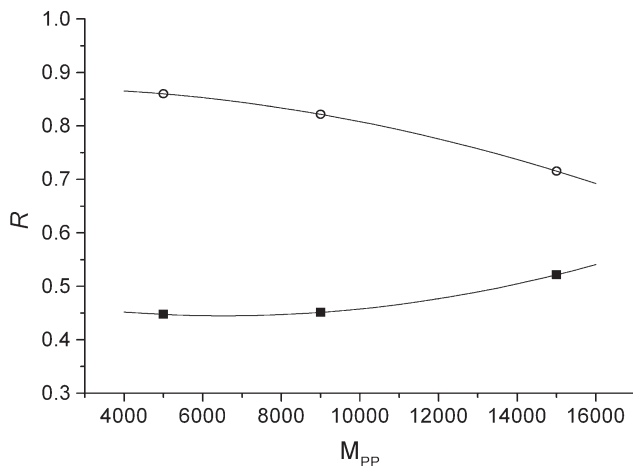
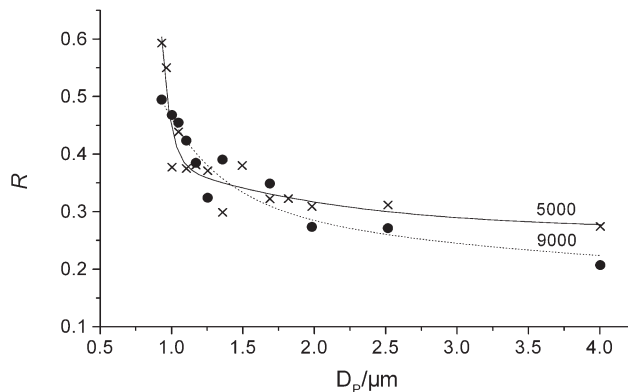## Effect of molecular weight of PPw in PPw-*g*-PEG on surface enrichment

The copolymers were amphiphilic as described before, the PPw main chain has strong interaction with host PP due to their similar structure, which make PP-*g*-PEG anchor into the host polymer and avoid the copolymer to be detracted from the product, meanwhile the interaction may also hinder the copolymer to immigrate to the surface. The results of effect of molecular weight of PPw on surface selectivity were shown in Figure 6. It was evident that the PP-*g*-PEG copolymers with smaller PPw main chain had greater surface selectivity than that with larger PPw. This indicated that the interaction between PPw main chain of copolymer and host polymer had hindered the modifiers to enrich on the surface.

The copolymer distribution in film in Figure 7 also shown this trend, concentration gradient in blend of copolymer with PPw molecular weight of 5000 was more obviously than that of 9000. The content of copolymer located in the site of 1.0 μm was two times that in 2.0 μm. Larger main chain and interaction between modifier and host polymer tended to leave the modifier in bulk. On the other hand, the relatively strong interaction may be in favor of the copolymer adhering to the surface to keep the lasting efficiency. Therefore, the molecular structure must be appropriate so that the surface selectivity of amphiphlic copolymer and their lasting efficiency can be attended at the same time.

SEM analysis was performed on melt-cast films of 3% PP-*g*-PEG/PP after the modifiers were eroded by a mixture of potassium dichromate and concentrated sulfuric acid. The results were shown in Figure 8. The graft copolymer in the domains seemed to have a phase-separated morphology as indicated in

Figure 7 Surface concentration gradients of PPw-*g*-PEG with different molecular weight of PPw segment in blends of PPw-*g*-PEG/PP under the contact die of PI.
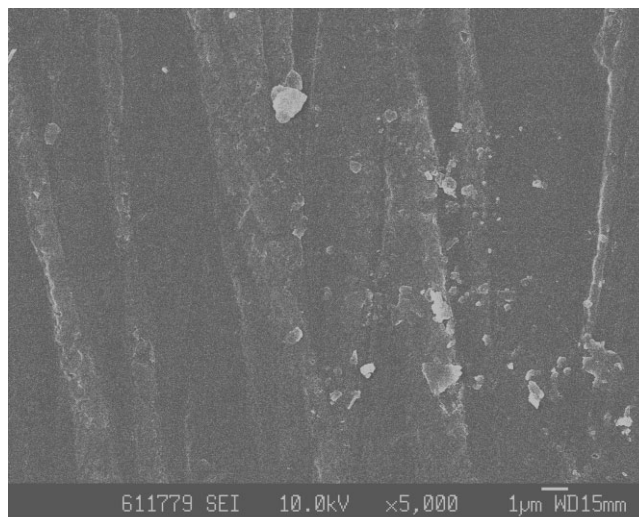
(a)

(b)

Figure 8 SEM micrographs of PPw-*g*-PEG in 3% PPw-*g*-PEG/PP blends film.Footers: Molecular weight of PPw (a): 5000, (b): 15,000.
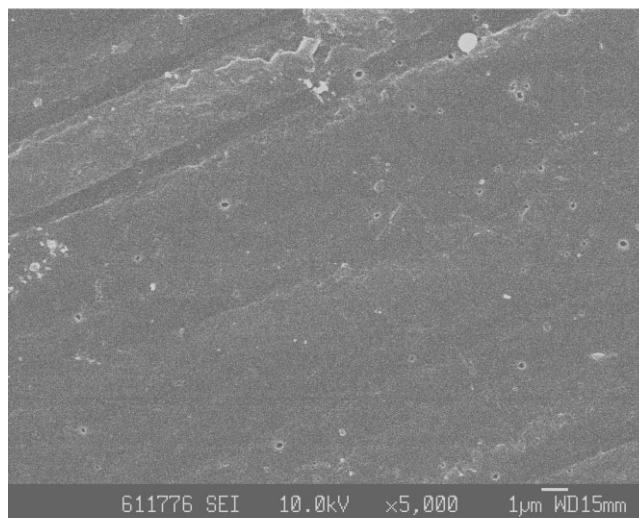

Figure 6 The influence of molecular weight of PP segment in PPw-*g*-PEG on surface peak area ratio of PPw-*g*-PEG/PP blends film.

TABLE III
Influence of Molecular Weight of PEG in PPw-g-PEG on Surface Peak Area Ratio

| Molecular weight of PEG	600	750[a]	1000	2000
R	0.4154	0.3903	0.4706	0.3665

[a] The end group is methoxyl, other is hydroxyl.

Figure 8. Because PP-g-PEG with larger PPw had better compatible with PP and stronger interaction between modifier and host polymer, when compared with PP-g-PEG having PPw molecular weight of 15,000, higher content and bigger phase domains of PP-g-PEG on surface were observed in modified film of PP-g-PEG with PPw molecular weight of 5000. This indicated that the strong interaction hindered the modifier to diffuse to the surface, although the smaller phase domains were usually in favor of the diffusion of PP-g-PEG to the surface. Another result from SEM figure was that PP-g-PEG with larger PPw can increase the compatibility, which rendered the modifier to distribute uniformly on the surface and increased the quality of modification.

Effect of molecular weight of PEG in PPw-g-PEG on surface enrichment

Although the general effect of copolymer selectivity at low loadings was common for all the copolymer, comparison of the data for the various molecular weight of PEG in Table IV showed that the surface selectivity of side chain PEG$_{1000}$ seemed higher than that of PEG$_{600}$ and PEG$_{2000}$. The variation probably reflected the extent to which phase separation occurred between PEG with different molecular weights. Copolymer with high molecular weight PEG was more phase-separated than that with lower molecular weight, and the bigger phase domains instead increased the resistance of copolymer transferring to the surface as increasing the molecular weight of PEG side chain.

In Bergbreiter work, an oligomer terminated with an ether-bound nitroxyl group verse an amide-bound nitroxyl group interacted with solvent to a smaller extent and was presumably located deeper in the entrapment functionalized polymer.[10] In this work, extensions of their studies were facilitated by preparation the hydroxyl- and methoxyl-terminated side chain graft copolymer and formation the blend films by melt-cast to avoid the effect of solvent. The results listed in Table III shown that the end group of copolymer had significant effect on surface selectivity. Copolymer with a less PP miscible end group had a significant surface selectivity than that of methoxyl-termination, which may be the result of
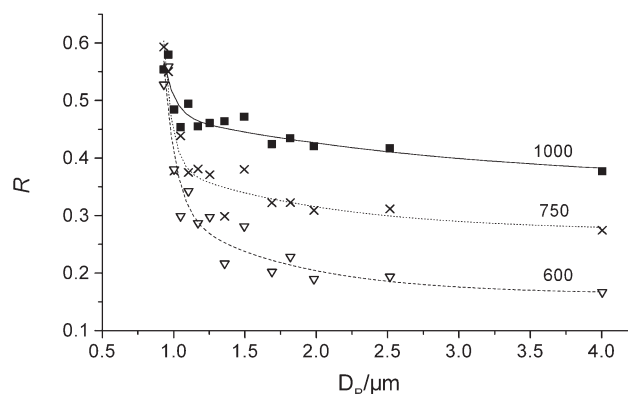
Figure 9 Surface concentration gradients of PPw-g-PEG with different molecular weight of PEG segment in blends of PPw-g-PEG/PP under the contact die of PI.

higher interaction between copolymer and host polymer.

The surface depth profiles of films with similar composition formed by melt-cast under polyimide were determined by variable ATR-FTIR. The results were shown in Figure 9. PP-g-PEG with smaller PEG side chain had more obvious depth gradient than that with larger PEG side chain. This meant that PP-g-PEG oligomers having larger PEG side chain had smaller surface selectivity than similar graft oligomers with smaller PEG group. The result was slightly different from that in ATR-FTIR at an angle of 45°. Commonly, the data from ATR-FTIR at 45° are the average values deal with the outmost about 3.0 μm, whereas the results in variable ATR-FTIR only express the property located the spot. Therefore, the result in variable ATR-FTIR is usually considered to be more precise than that from ATR-FTIR at 45°.

Effect of PPw-g-PEG on surface contact angles

Water contact angles have been shown to be very sensitive to the nature of the functional groups at a few angstroms of the upper layer on the surface for functionalized polymers. Measurement of water contact angles gives a good estimate of the polarity and the mobility of the polymer chains present in the outmost atomic layers of the surface. The functionalized PP in this work has essentially two types of functional groups, namely the high polar and hydrophilic portion contributed by the oly PEG units and

TABLE IV
Influence of Content of Modifier on Contact Angle of Modifier/PP Blends Film

Loadings of PPw-g-PEG, wt%	0	1%	2%	3%	5%	6%
Contact angle/°	119.8	70.2	73.9	76.6	80.0	87.2

TABLE V
Influence of PPw-*g*-PEG Structure on Contact Angle of 2% PPw-*g*-PEG/PP Film

$\overline{M_n}$ of PEG	600	750[a]	1000	2000
Steel die	82.5	89.8	88.8	100.0
PTFE die	105.1	–	101.6	99.5

[a] The end group is methoxyl, other is hydroxyl.

the nonpolar portion by the PP units. The contact angles for the surface of blending were uniformly much less than the contact angle found for the surface of pure PP (Table IV). This reflects a preferential migration of PEG blocks to the contact-angle interface when the blend film is formed in the steel. Steel is a high-energy substrate, so PEG is expected to be contact with steel and PP is transfer to the inside of film. The data in Table IV also showed that there was an effect of the weight percent of PP-*g*-PEG. Contact angle gradually increased as the weight percent of the PP-*g*-PEG decreased, this was consistent with the results of ATR-FTIR before, although the various-depth regimes were attended for contact angle and ATR-FTIR analysis techniques. The data in Table V also showed that the effect of molecular weight of PEG in PP-*g*-PEG on the contact angle of film prepared under the steel die. The contact angles increased with increasing the molecular weight of PEG in PP-*g*-PEG and PP-*g*-PEG with methoxyl-terminated PEG had bigger contact angle due to the good compatibility with PP. All of these results meant that PP-*g*-PEG with higher molecular weight and methoxyl-terminated PEG had lower enrichment on the surface. This was consistence with the result of distribution of PP-*g*-PEG on the surface determined by variable ATR-FTIR. To our surprise, although PTFE is a representative nonpolar interface, the contact angle of film formed under PTFE die was also lower than that of pure PP. This indicated that some parts of PEG blocks were ejected to the surface by the crystallization repulsion of PP.

Evaluation of solvent-resistance of modified PP

The difference between blend surface modification and chemical graft modification is that the modifier from the former has no any chemical bond linking to the host polymer, which may mean that the modifier is relatively easy to be detracted from the host polymer if the structure of modifier is designed irrelevantly. However, the defect can be meliorated by grafting the host chain into the modifier so that the modifier can be anchored to the host polymer firmly. In the front discussion, we have proved that PP-*g*-PEG can immigrate to the surface. Considering that the modified PP may be contacted with water

or organic solvents, herein we observed the solvent-resistance of modified materials by dipping the functionalized PP into acetone for enactment time and determining the composition by ATR-FTIR. The effect of dipping time on peak area ratio of surface was shown in Figure 10. As a compare, the solvent-resistance of PEG modified was also shown in the figure.

Because of the weak interaction between PEG and host PP and strong extraction of acetone, the PEG content on surface was dropped obviously within the first 2 min in PEG-modified film. With further increasing the dipped time, the losses of PEG on the surface were unchangeable at the losses of 60%. Unlike that of PEG film, the content of PP-*g*-PEG increased with increasing the dipping times in acetone, although the content also decreased at the first 2 min. The result was rationalized to the balance between diffusion of PP-*g*-PEG and extraction of acetone. Acetone is a representative polar organic solvent. When it contacts with modified film, two different effects can be observed. The first is extraction, which lowers the surface content of modifier if the modifier cannot cling to the host polymer firmly. The second is the induction, which can induce the modifier to migrate from the inside of film to the surface and increase the surface enrichment of modifier. In PEG/PP blend film, due to the weak interaction between PEG and PP, the extraction of acetone was dominant, so the decreasing of PEG on surface was mainly observed; In PP-*g*-PEG/PP modified film, the phenomena was also observed in the first 2 min, indicating that some proportion of PP-*g*-PEG did not cling to the surface firmly and relatively easy to be detracted. The induction was dominant when the dipping time was over 2 min in PP/PP-*g*-PEG film, therefore the content of PP-*g*-PEG increased with increasing the dipped times in acetone. This may result in high content of PP-*g*-PEG on the surface. The biggest losses of
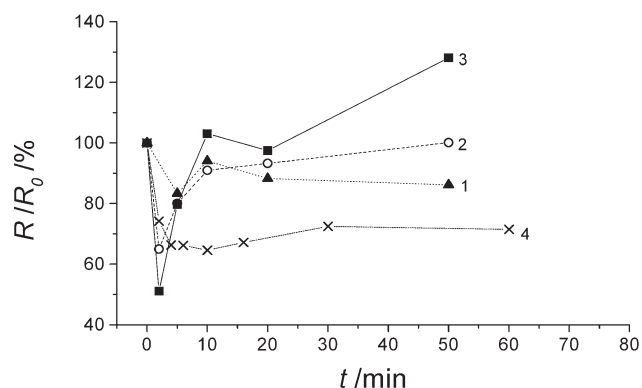
Figure 10 The change of modifiers in the surface vs. the immersion time in acetonefooters: 1, 2, 3: PPw-*g*-PEG with M_{PPw} of 5000, 9000, 15,000; 4: PEG.

PP-*g*-PEG on surface did not exceed over 10% after 50 min dipping, in some case, the content of PP-g-PEG even exceeded over that before dipping in acetone, especially in PP-g-PEG with higher molecular weight of PPw. All these indicated that there were relatively strong interaction existing between PP-*g*-PEG and PP and the modified materials had better solvent-resistance when compared with PEG/PP blend.

On the other hand, comparing the changes of PP-*g*-PEG with different molecular weight of PPw on surface with the immersion time in acetone, we can conclude that modifier with higher molecular weight of PPw had better solvent-resistance than that with lower molecular weight, which meant that there are stronger interactions between PP-*g*-PEG and PP in blend of PP-*g*-PEG with higher molecular weight of PPw. About 20% of the content of PP-*g*-PEG with PPw molecular weight of 15,000 on surface was surpassed over that of beginning after 50 min dipping. However, the molecular weight of PPw in the modifier should be designed carefully so that the strong interactions do not hinder its diffusion from the inside of film to the surface.

CONCLUSION

The PPw-*g*-PEG additive can preferred diffuse to the surface of blends and effectively increase the hydrophilicity of PP. The surface enrichment degree of PPw-*g*-PEG in PP blend film is obviously influenced by its structure, content, and contact interface. Lower content and higher surface energy die are in favor of the enrichment of PPw-*g*-PEG. PPw-*g*-PEG with lower PEG contents, lower molecular weight of PPw and PEG have better enrichment on the surface of PP blend film. By grafting of PEG-OH onto the MPP, PP macromolecular surface modifier with better solvent-resistance than that of PEG can be achieved.

References

1. Bergbreiter, D. E.; Brian, W.; Gray, H. N. Macromolecules 1998, 31, 3417.
2. Bamford, C. H.; Al-Lamee, K. G. Polymer 1994, 35, 2844.
3. Singh, R. P. Prog Polym Sci 1992, 17, 251.
4. Thomas, H. R.; O'malley, J. J. Macromolecules 1981, 14, 1316.
5. Lee, H.; Archer, L. A. Polymer 2002, 43, 2721.
6. Bergberiter, D. E.; Stinivas, B. Macromolecules 1992, 25, 636.
7. Lee, H.; Archer, L. A. Macromolecules 2001, 34, 4572.
8. Busscher, H. J.; Hoogsteen, W.; Dijkema, L.; Sawatsky, G. A.; van Pelt, A. W. J.; de Jong, H. P.; Challa, G.; Arends, J. Surf Interface Anal 1986, 9, 418.
9. Schmitt, J. J.; Gardella, J. A.; Salvati, L. Macromolecules 1989, 22, 4489.
10. Chen, H. J.; Zhu, Y. F.; Zhang, Y.; Xu, J. R. J Vinyl Additive Technol 2008, 14, 28.
11. Chen, H. J.; Zhu, Y. F.; Zhang, Y.; Xu, J. R. Front Chem Eng China 2008, 2, 102.
12. Bergbreiter, D. E. Prog Polym Sci 1994, 19, 529.