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Study on styrene-assisted melt free-radical grafting of maleic anhydride onto polypropylene

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Abstract

Free-radical melt grafting of the multi-monomer system of maleic anhydride (MAH)/styrene (St) onto polypropylene (PP) was studied using a single-screw extruder. The effects of St and initiator concentrations on the grafting reaction were investigated. It was shown that the addition of St to the melt-grafting system as a comonomer could significantly enhance MAH graft degree onto PP. The maximum MAH graft degree was obtained when the molar ratio of MAH to St was approximately 1:1. However, the melt flow rate (MFR) value of the grafted PP was the highest at this ratio. This implies that the interaction and reaction between MAH and St monomers plays an important role in the grafting reaction. St improves the grafting reactivity of MAH and also reacts with MAH to form the St–MAH copolymer (SMA) before the two monomers graft onto PP. Grafting of SMA onto PP greatly enhanced the graft degree of MAH. When the molar ratio of MAH to St is 1:1, the main grafting reaction in the system is the grafting of SMA to PP; therefore, the graft degree of the grafted PP is the highest. When the concentration of MAH is higher than that of St, some MAH monomer reacts with St to form SMA, but others can directly graft onto PP macroradicals. When the amount of St added is higher than that of MAH, part of the St monomer may preferentially react with PP macroradicals to form more stable styryl macroradicals, while others copolymerize with MAH to yield SMA. The MFR value of the grafted PP is lower in this case.

Keywords: Polypropylene; Melt grafting; Mutli-monomer

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1. Introduction

Polypropylene (PP) is one of the most important general-purpose polymers widely used in various fields, but the non-polarity of the material restricts its application. In recent years, modification of PP in an extruder via free-radical grafting has become a widely used post-polymerization method to functionalize the material [1–3]. Various monomers have been used for melt-grafting of PP. Maleic anhydride (MAH) has received intense attention primarily due to the difficulty in homopolymerization of this monomer during free-radical grafting [4]. The actual presence of poly(maleic anhydride) segments (grafted or free) under standard processing conditions has been an important subject of debate in the literature for some time [2,3,5–7]. Analysis of MAH-modified PP has shown that MAH is usually grafted onto the PP chain ends [2] and that the grafted MAH is present as single MAH or succinic anhydride [6]. Glycidyl methacrylate (GMA) is also a very interesting monomer because of its bifunctional character: a free-radically reactive double bond together with an epoxy group that can react with a number of other functional groups such as carboxyl, hydroxyl, anhydride and amine [8–11]. Usually, GMA can be grafted onto PP without destroying the epoxy groups that would be essential to subsequent reactive blending with other polymers.

However, the graft degree of those monomers are usually low [12]. It has been found that the radical grafting of a monomer onto PP in the presence of a peroxide initiator is often accompanied by severe degradation of the PP backbone. It has been well established that a free-radical grafting process starts with the formation of macroradicals along the PP chains by a so-called hydrogen abstraction mechanism. These macroradicals may subsequently follow

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two competing pathways. They can either initiate grafting of the monomers or undergo chain scission, which is the main side reaction during the process. Recently, it was shown that the addition of styrene (St) as a second monomer in the melt-grafting system assisted in reducing the PP chain scission and increasing the graft degree of GMA on PP [10–15]. The idea of using St as a comonomer originated from a detailed analysis of the mechanism of free-radical grafting. To obtain high graft degree with reduced degradation of PP, it is essential that the PP macroradicals react with the grafting monomers before they undergo chain scission. Because GMA is not sufficiently reactive towards PP macroradicals, it would be helpful to use a second monomer that can react with them much faster than GMA, and the resulting free radicals can then copolymerize readily with GMA [12,13]. In this way, instead of grafting GMA directly onto the PP chains, the comonomer (St) serves as a medium to bridge the gap between the PP macroradicals and the GMA monomer. It has been proposed that, in the above system, St preferentially reacts with the PP macroradicals to form more stable styryl macroradicals, which then copolymerize with GMA to form branches [12–14]. By this mechanism, the addition of St may not only improve the grafting level of GMA but also reduce PP degradation. Both benefits were enhanced when an increasing amount of St was used.

It was also shown that the addition of St can improve the graft degree of some other monomers such as hydroxy ethyl methacrylate (HEMA) and methyl methacrylate (MMA), but not the graft degree of vinyl acetate (VAc) and ricinoloxazoline maleinate (OXA). This is due to the fact that St can copolymerize easily with HEMA, MMA and GMA but not with VAc and OXA [16,17].

As for the free-radical grafting of MAH onto PP, the addition of St was shown to be effective in improving the graft degree while minimizing the degradation of PP [18]. Generally, a limited graft degree and severe degradation of PP during the melt grafting of MAH onto PP are caused by the low reactivity of MAH towards free radicals and its low solubility in the PP melt. The low free-radical reactivity of MAH is inherently due to its structural symmetry and deficiency of electron density around the double bond [19]. It is conceivable that the presence of a monomer capable of donating electrons could activate MAH by rendering its structure unsymmetrical and its π bond of radical-anion character. For example, St and α-methylstyrene are good electro-donating monomers and they can interact with MAH through charge transfer complexes and copolymerize with MAH alternatively in certain conditions. No MAH oligomers were found when PP was grafted with MAH and St [18] or with MAH and α-methylstyrene [20].

The melt-grafting system with comonomers is actually a multi-monomer grafting system. From the above discussion, we know that a special interaction occurs between MAH and St but not between GMA and St. Therefore, we could expect that the reaction in the MAH–St multi-monomer grafting system is different from that in the GMA–St grafting system. Our previous work has proved that the interaction between MAH and St in the melt-grafting process of MAH–St onto PP played an important role in the system [21,22]. The primary aim of the present study was to provide a detailed investigation on melt free-radical grafting of MAH onto PP with St as a comonomer and to discuss the mechanism of the multi-monomer grafting reaction.

2. Experimental

2.1. Materials

The PP resin (PP2401) used in this study was a homopolymer, having a melt flow rate (MFR) of 4.5 g/10 min, made by Yanshan Petrochemical Co. The MAH monomer and St monomer used were commercially available (analytical grade). Dicumyl peroxide (DCP) used as an initiator was obtained from Fushun 3rd Chemical Factory.

2.2. Melt grafting

The melt-grafting reactions were carried out in a single-screw extruder ($\Phi = 30$ mm, $L/D = 25$). PP pellets, monomer(s) (1.5–7 wt%), and initiator (0.05–0.5 wt%) were dry-blended before being charged into the extruder. The temperature of the cylinder was set at 170–210°C. The screw speed was fixed at 30 rpm.

2.3. Purification and characterization of grafted PP

The grafted PP samples were dissolved in refluxing xylene at a concentration of 1%(wt/vol.), and excess acetone was then added to precipitate them. Acetone could precipitate only the grafted and ungrafted PP [i.e. PP, PP-*g*-MAH, PP-*g*-St, and PP-*g*-(MAH–St)]. By this procedure any homo- and copolymer of MAH and St was separated, and the actual graft degree could be obtained. The precipitated samples were filtered, washed and dried under vacuum at 80°C for 24 h.

The sample thus obtained was then hot-pressed under 190°C into a piece of thin film and analyzed by FTIR. FTIR spectra were recorded on a Nicolet FTIR-8201 PC.

In some special cases, the filtrate left after the purification process was also analyzed. Solvents such as acetone and xylene were evaporated from the filtrate left until a white powder was obtained, which was then vacuum heated at 85°C for 3 h. FTIR spectra were recorded from the thin flake compressed with mixtures of the thus-obtained powder and analytical-grade KBr powder. Differential scanning calorimetry (DSC) measurements were carried out using a TA 2910. The samples were heated from room temperature up to 250°C at a constant rate of 10°C min^{-1}. In the meantime, the scan curves were obtained.

MFR measurements of the purified PP samples were carried out using XRZ-400 type MFR equipment at 230°C with a load of 2.16 kg weight according to the ASTM D1238-86T standard.

3. Results and discussion

3.1. FTIR analysis

The FTIR spectra of the pure PP, PP-*g*-MAH, and PP-*g*-(MAH–St) are shown in Fig. 1. In the cases of PP-*g*-MAH and PP-*g*-(MAH–St), new absorption bands at 1782 and 1857 cm^{-1} were observed, which can be assigned to the absorption of the carbonyl groups(–C=O) of cyclic anhydride. It can be assumed that most of the grafted MAH in the samples was in a cyclic form, because no peak was found at 1715 cm^{-1}, which should be assigned to the carboxylic acid. The absorptions at 2723 and 704 cm^{-1} can be assigned to the characteristic absorption of the PP skeleton and the grafted St, respectively. The absorption band at 2723 cm^{-1} was chosen as an internal reference in this case.

The spectra showed that the addition of St could significantly increase the graft degree of MAH. The intensity of the carbonyl absorption band at 1782 cm^{-1} of PP-*g*-(MAH–St) obtained from the PP/MAH/St/DCP (100/3/3/0.3) melt-grafting system was much stronger than that of PP-*g*-MAH obtained from the PP/MAH/DCP (100/3/0.3) system. The absorbance ratio (R_a) of the areas of the bands at 1782 and 2723 cm^{-1} shows the relative graft degree of MAH. Similarly, the absorbance ratio of 704 and 2723 cm^{-1} shows the relative graft degree of St. The results of these calculations will be discussed in detail later.

It should be noted that the partially enlarged spectrum of PP-*g*-MAH shown in Fig. 2 demonstrates two peaks at 1782 and 1790 cm^{-1}, both of which can be assigned to symmetric stretching of the carbonyl groups in the grafted anhydride. The one at 1790 cm^{-1} was attributed to a single succinic anhydride attached to the chain end of PP [2,4]. The band at 1782 cm^{-1} is assigned to poly(maleic anhydride) or other forms of MAH [2].

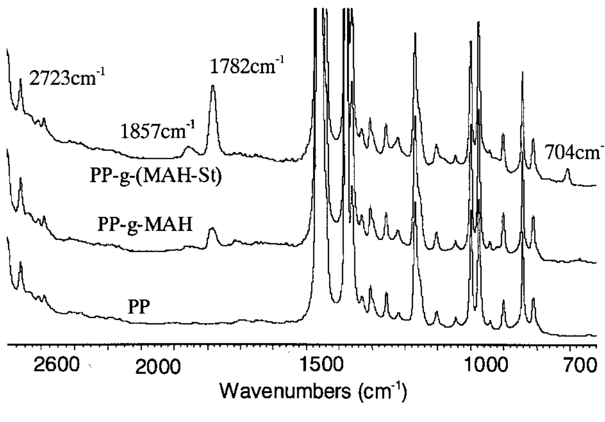

Fig. 1. FTIR spectra of PP, PP-*g*-MAH obtained from PP/M AH/DCP(100/3/0.3) system and PP-*g*-(MAH–St) obtained from the PP/MAH/St/DCP(100/3/3/0.3) system.

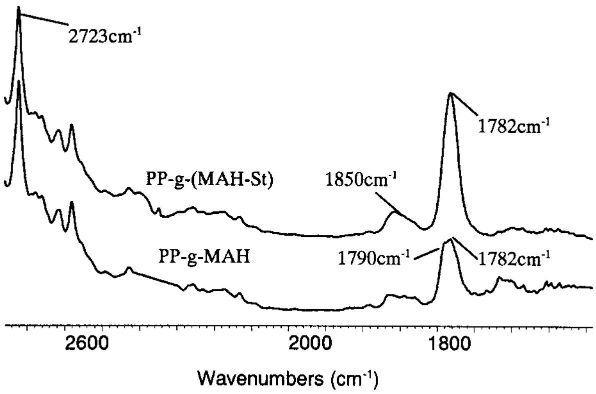

Fig. 2. Comparison of FTIR absorption bands of carbonyl groups of PP-*g*-(MAH–St) and PP-*g*-MAH.

The FTIR spectrum of the St–MAH copolymer (SMA) obtained by radical copolymerization is shown in Fig. 3. The band at 1782 cm^{-1} in the spectrum of SMA was the same as in the spectrum of PP-*g*-(MAH–St), in both of which the band at 1790 cm^{-1} was not found.

The disappearance of the band at 1790 cm^{-1} in the spectra of PP-*g*-(MAH–St) means that fewer single MAH moieties were attached to PP chain ends when St was added into the melt-grafting system. It is obvious that there are different mechanisms for the melt-grafting systems of MAH onto PP in the presence or in absence of St [22].

3.2. Effect of St concentration on graft degree and MFR of grafted PP

Fig. 4a shows the effect of St concentration on the graft degree of MAH and St of the grafted PP when the MAH and DCP concentrations were fixed at 3 and 0.3% based on PP, respectively. The graft degree of St increased with increasing St concentration. The MAH graft degree of the resulting polymer was drastically higher in the presence of St than in the absence of St. It reached a maximum when the molar ratio of MAH and St was approximately 1:1 and then

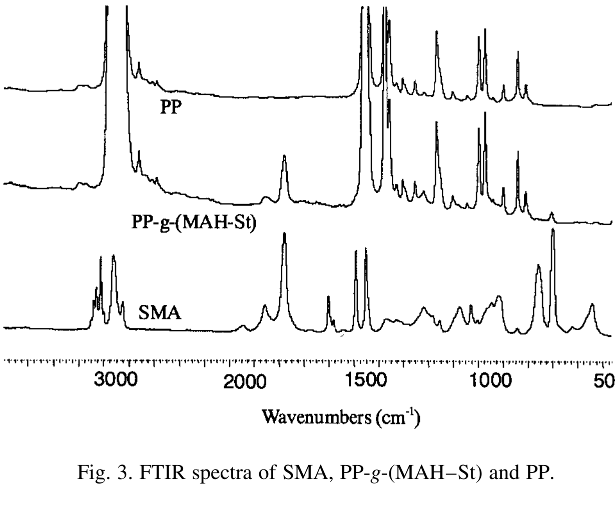

Fig. 3. FTIR spectra of SMA, PP-*g*-(MAH–St) and PP.

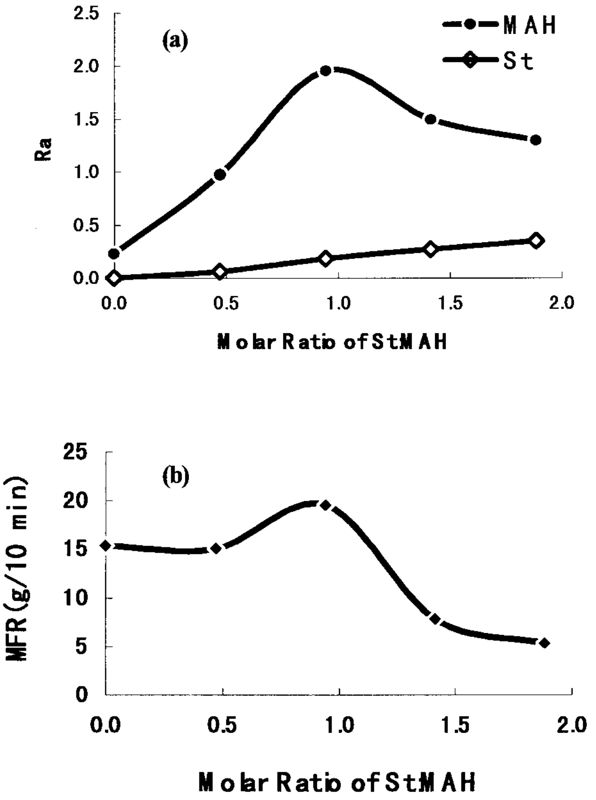

Fig. 4. Effect of St concentration on: (a) graft degree of MAH and St; and (b) MFR value of the grafted PP. MAH = 3 phr, DCP = 0.3 phr.

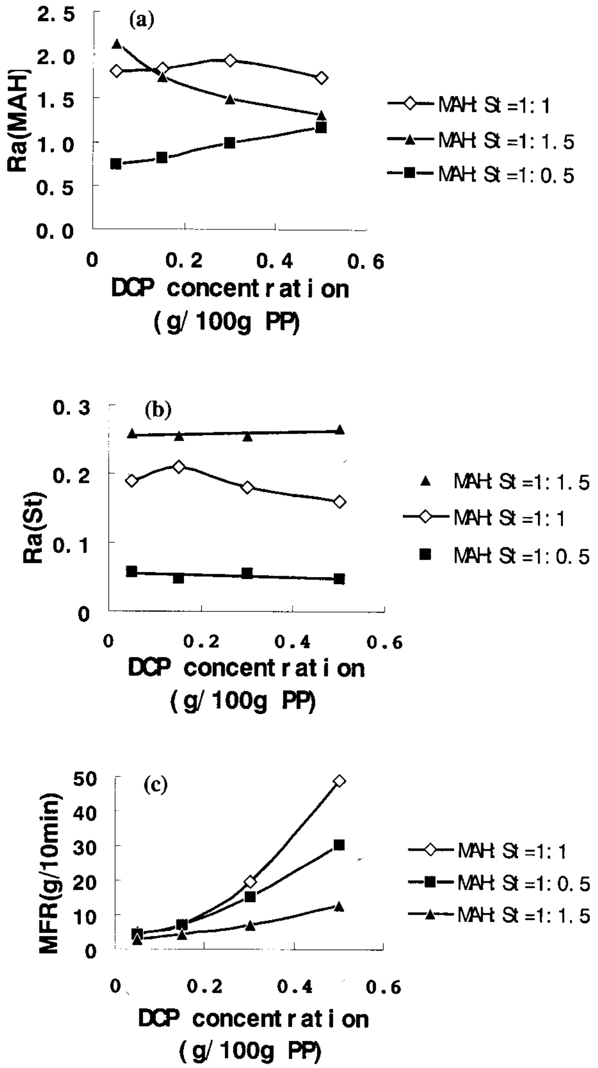

Fig. 5. Effect of DCP concentration on: (a) MAH absorbance ratio; (b) St absorbance ratio; and (c) MFR value of the grafted PP. MAH = 3 phr.

decreased when the concentration of St was higher than that of MAH. Fig. 4b shows the effect of St concentration on MFR values of the grafted PP obtained under the same conditions as in Fig. 4a. The MFR value of the grafted PP decreased with increasing St concentration, which means that the addition of St effectively reduces the degradation of PP. However, when the St concentration equaled the MAH concentration, the MFR of the resulting PP reached its highest value, even higher than the MFR value of the grafted PP obtained in the absence of the St monomer.

3.3. Effect of DCP concentration on graft degree and MFR of the grafted PP

The concentration ratio of MAH and St was shown to have great influence on graft degree and the MFR of the grafted PP. Therefore, three series with different MAH to St ratios (i.e. MAH/St = 1:0.5, 1:1, 1:1.5, respectively, with the MAH concentration fixed at 3 phr) were selected for investigation of the influence of DCP concentration on graft degree and MFR values of the grafted PP. The effects of DCP concentration on the graft degree of MAH and St, and MFR values of the grafted PP are shown in Figs. 5 a–c, respectively.

The MAH graft degree of the 1:1 series was the greatest among the three series (Fig. 5a). The same result is shown in Fig. 4a. The MAH graft degree of this series first increased then decreased tardily along with increasing DCP concentration, although the overall fluctuation was relatively small. The highest MAH graft degree was obtained when the DCP concentration was 0.3 wt % based on PP. It should be noted that the MFR values of the grafted PP for the same series went up sharply when the DCP concentration was greater than 0.3 wt% as shown in Fig. 5c, i.e. PP underwent severe chain scission when DCP concentration was greater than 0.3 phr. This means that when the concentration ratio of MAH and St is 1:1, increasing initiator concentration within some limit is effective for improving the graft degree of MAH. However, when the initiator was used excessively, the graft degree decreased because of severe chain degradation of the PP backbone.

For the series of MAH/St at 1:0.5, the MAH graft degree of the grafted PP increased steadily with increasing amount of the peroxide (Fig. 5a), while the St graft degree remained

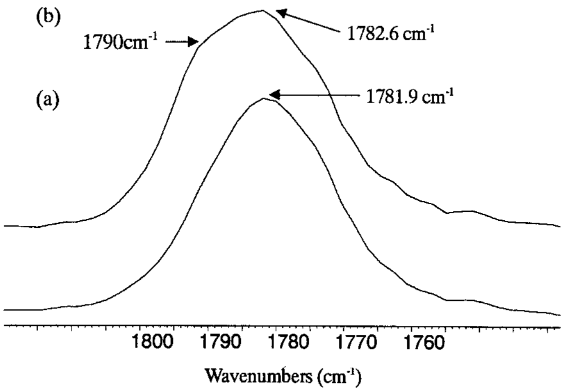

Fig. 6. FTIR spectra of PP-*g*-(MAH–St) obtained from different DCP concentration systems: (a) DCP = 0.05 phr; and (b) DCP = 0.5 phr. MAH = St = 3 phr.

almost unchanged as shown in Fig. 5b. This implies that when the amount of peroxide increased, more PP macroradicals were produced by a hydrogen abstraction reaction and more chain ends of PP macroradicals resulted from chain scission. More MAH monomers could react with these macroradicals and form single succinic anhydride moieties at PP chain ends. As a result, the graft degree of MAH went up. This was proved by the FTIR spectra shown in Fig. 6. When the DCP concentration was increased from 0.05 to 0.5 phr, the position of carbonyl absorption moved from 1781.9 to 1782.6 cm^{-1}. Meanwhile, a shoulder at 1790 cm^{-1} was formed. This means that more single succinic anhydride moieties attached to PP chain ends. The MFR of the grafted PP for the MAH/St = 1:0.5 series increased with the increasing DCP concentration. It can be seen that all the MFR values for this series were lower than those of the series of MAH/St = 1:1. This seems rather strange because less monomer was added for the series of MAH/St = 1:0.5 than for the series of MAH/St = 1:1. As will be discussed later in this article, the MFR values of the grafted PP are not determined by the starting concentration of the monomers added to the grafting system.

For the series of MAH/St = 1:1.5 (wt/wt), MAH graft degree of the grafted PP decreased gradually along with increasing DCP concentration (Fig. 5a), while the St absorbance ratio, which was the highest among the three series, remained unchanged (Fig. 5b). When the initiator concentration was increased, more PP macroradicals were produced. More St monomer could then react with these macroradicals and form styryl macroradicals because the St concentration was greater than the MAH concentration. The above reaction stabilized the PP macroradicals, thus depressing PP degradation. As a result, the MFR values of this series shown in Fig. 5c were the lowest among the three series.

3.4. Mechanism of styrene-assisted free-radical grafting of MAH onto PP

The melt free-radical grafting process starts with the formation of macroradicals along the PP chain by a hydrogen abstraction reaction as stated in the introduction. These PP chain macroradicals (shown as radical I in Fig. 7) and PP chain end macroradicals (shown as radical II in Fig. 7) produced by β-scission can both take part in grafting reaction.

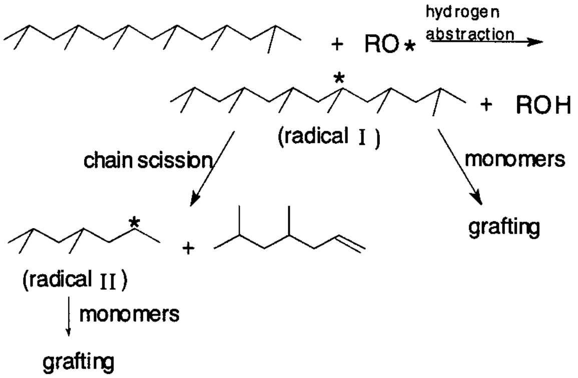

Fig. 7. Scheme of reactions in the free-radical grafting system of PP.

For the melt free-radical grafting system of MAH with PP in the absence of St, the graft degree of MAH is very low because of the low reactivity of MAH toward radical reactions. Much of the MAH monomers added are grafted onto PP chain ends after PP undergoes chain scission [2], showing absorption bands at 1790 cm^{-1} of MAH in the FTIR spectra. When St is added, the two monomers, St and MAH, can interact with each other one-to-one to form a charge transfer complex (CTC), the existence of which has been known for a long time and used to explain the formation of alternating copolymers of MAH and St in bulk or solution copolymerization. The formation of CTC increases electric asymmetry on the double bond of MAH, thus enhancing the monomer's reactivity [16]. The two monomers can also copolymerize with each other under initiation of peroxide to form a chain of MAH–St copolymer (SMA), which may then react with PP macroradicals producing branches by termination between radicals. Therefore, the graft degree of MAH can be significantly improved in the presence of St.

For the grafting system into which equimolar amounts of MAH and St were added, the two monomers can easily interact and copolymerize with each other. The grafting of SMA may yield relatively long branch; therefore, the MAH graft degree of MAH/St = 1:1 series were the highest. Meanwhile, the formation of SMA also greatly lessens the concentration of free monomers, namely the total number of grafting reactive species, particularly the activated reactive species. Thus the degradation of the PP backbone could not be depressed, consequently making the MFR values of the grafted PP the greatest, even greater than those of the grafted PP in the absence of St.

When the St concentration was less than the MAH concentration (e.g. for the MAH/St = 1:0.5 series), the amount of the resulting SMA copolymer was less, so the graft degree of MAH was also less than that with the MAH/St = 1:1 series. Besides, there were still some free MAH monomers that could be grafted onto PP chains, which means that the amount of reactive species to graft and to reduce PP degradation is more than that of the MAH/St = 1:1 series; therefore, the MFR of the modified PP was lower. The unchanging St graft degree along with increasing DCP concentration (as shown in Fig. 5b, MAH/St = 1:0.5) may be due to the fact that the amount of SMA copolymer to be produced and to be grafted was relatively fixed by the initial concentration of the two monomers.

When more St than MAH was added to the grafting system, part of the St monomer reacted with MAH to form the SMA copolymer. Other St monomers could preferentially react with PP chain macroradicals to produce relatively stable styryl macroradicals, thus partially depressing PP degradation. Because of the stability of the latter macroradicals, their further reaction with SMA copolymer would be difficult. As a result, the MAH graft degree of the resulting material was lower than that of the MAH/St = 1:1 series. The unchanging St graft degree with increasing DCP concentration (as shown in Fig. 5b, MAH/St = 1:1.5) meant that the amount of grafted St was not influenced by how the St monomers grafted onto the PP chain, either by reacting with PP macroradicals first to form styryl macroradicals or by reacting with MAH first to form SMA copolymer which was then grafted onto the PP chain. The different way of grafting mainly affects the graft degree of MAH.

The above discussion introduced the concept of the copolymerization and grafting of SMA. If the proposed mechanism is true for the system of MAH–St melt-grafting onto PP, there should be some ungrafted SMA copolymer remaining in the extruded PP. The SMA copolymer can be dissolved by acetone during the purification process mentioned in the experimental section; therefore, it should stay in the filtrate. We analyzed the filtrate left from the purification process of the modified PP (for MAH/St = 1:1 system) to determine whether there was any SMA copolymer. After solvents were evaporated from the filtrate, some white powder was obtained, which was then vacuum-heated to remove any unreacted MAH monomer. Fig. 8 shows the FTIR spectrum of the thus obtained powder as well as that of SMA, which has also been shown in Fig. 3. Both spectra shown in Fig. 8 showed the strong characteristic absorption of the benzene ring and the carbonyl groups of cyclic anhydride. The difference in the two spectra was due to the different MAH contents of the two substances. Polystyrene and poly(maleic anhydride) could also be present in the above mentioned filtrate and the white powder and give rise to the absorption bands of the benzene ring and the carbonyl groups, respectively. For that reason, we cannot only use the FTIR spectra as conclusive proof of the presence of SMA copolymer. The DSC curve of the white powder showed a thermal transition at about 160°C, which corresponds to the glass transition of SMA as reported in the literature [23]. No glass transition for polystyrene and poly(maleic anhydride) was detected. Thus we attain the conclusion that SMA copolymer is formed during the melt-grafting of MAH–St onto PP, although the grafting of SMA onto the PP chain still needs more research and characterization.

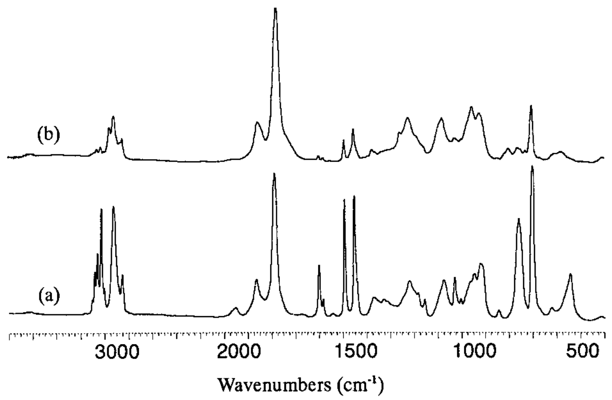

Fig. 8. FTIR spectra of: (a) SMA; and (b) white powder obtained from the filtrate of MAH/St = 1:1 series.

Due to the formation of SMA, the concentration of free monomers, namely the total concentration of the grafting reactive species in the system was changed. Based on the above discussion, it is conceivable that the presence and concentration of free monomers, St or MAH or both, have a great influence on the competition between the grafting reaction and the β-scission reaction of PP macroradicals. For example, when some free St monomer was present (as for the MAH/St = 1:1.5 series), the MFR values of the grafted PP were low as a result of depressed PP degradation or β-scission. Thus, it is considered that the type and concentration of the grafting reactive species in the grafting system are among the crucial factors that determine the MFR values of the grafted PP.

4. Conclusions

The addition of St as comonomer to the melt-grafting system of MAH onto PP could greatly improve the graft degree of MAH. It has been shown that the maximum MAH graft degree is obtained when the molar ratio of MAH to St is approximately 1:1 for the same concentration of MAH. St reacts with MAH to form SMA copolymer during the melt-grafting process and the grafting of SMA leads to improving the graft degree of MAH. When the molar ratio of MAH to St is 1:1, the main grafting reaction in the system is the grafting of SMA to PP; therefore, the graft degree of the grafted PP is the highest. When the concentration of MAH is higher than that of St, some MAH monomer reacts with St to form SMA; others can directly graft onto PP macroradicals. When the amount of St added is higher than that of MAH, part of the St monomer may preferentially react with PP macroradicals to form more stable styryl macroradicals, while others copolymerize with MAH yielding SMA. The MFR of the grafted PP is lower in this case.

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