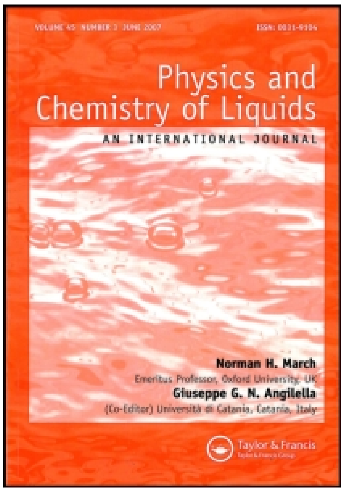

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Solution properties of chlorinated polypropylene and maleic anhydride grafted chlorinated polypropylene

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SOLUTION PROPERTIES OF CHLORINATED POLYPROPYLENE AND MALEIC ANHYDRIDE GRAFTED CHLORINATED POLYPROPYLENE

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The solution behaviors of the chlorinated polypropylene (CPP) and its grafted polymers (CPP-g-MAH) were systematically studied to characterize their polar change with grafting maleic anhydride (MAH) onto the chain of CPP. The molecular weights of the polymers were determined with light scattering measurements, and the Mark–Houwink equation of CPP in toluene was also obtained. The result showed that the Mark–Houwink equation of CPP was suitable for estimating the molecular weight of the polydisperse samples of CPP and not suitable to CPP-g-MAH because the molecular polarity of the graft polymers had changed with grafting MAH onto CPP. The solubility result of CPP and CPP-g-MAH in various solvents indicated that the polarity of CPP gradually increased with grafting MAH onto its chain, which would cause the solubility of poorly hydrogen bonded solvents for CPP-g-MAH to gradually become poor, whereas that of moderately hydrogen bonded solvents for the polymers becomes better with an increase of the MAH graft content. This is consistent with the results of their dilution ratio and solubility parameter. Stabilities of the $344^{\#}$ resin–CPP-g-MAH–toluene solutions showed that the miscibility of CPP-g-MAH and $344^{\#}$ resin was improved with increase of the MAH grafted content.

Keywords: Chlorinated polypropylene; Maleic anhydride; Molecular polarity

INTRODUCTION

In the polymer industry, grafting the polar and reactive maleic anhydride (MAH) onto polyolefins and their chlorinated polymers is often adopted to increase their molecular polarity, which will improve miscibility of the polyolefine and the other resin blends and influence their service performance [1–5]. These previous works had as their main focus the synthesis of MAH grafted polyolefins, the characterization of their chemical structure, and the development of chemical models. For example, Li *et al.* [2] studied solution grafting of maleic anhydride onto linear low-density polyethylene,

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and obtained the reaction condition and the dosage of anticrosslinking agents. The study of Heinen *et al.* [3] suggested that MAH structures grafted on PE, EPM, and PP in the melt and solution were saturated, monomeric MAH molecules, which was consistent with the result reported by Huang *et al.* [4]. Funasaka *et al.* [5] compared with the solvent solubility of propylene–butene copolymers (PPB) and its grafts modified with maleic anhydride in various solvents at the time of researching their adhesive ability, and found that nonpolar solvents such as toluene and cyclohexane could solve PPB and at even higher temperatures the solubility range could not be much wider. When the degree of grafting was 4.1%, the solubility range of this polymer expanded, that is, ten kinds of solvents could dissolve the grafts at room temperature. Up to now, however, no systematic literature data are available for the regular change of their molecular polarity with the MAH graft content and the effect of the MAH grafted content on their solution properties. Therefore, it is important to systematically investigate the quantitative change relationships between molecular polarity of the graft polymer and MAH grafted content for the purpose of foundational research and application. In the present work, chlorinated polypropylene grafted maleic anhydride (CPP-g-MAH) is selected as an example, and the solution properties of chlorinated polypropylene (CPP) and its graft polymers (CPP-g-MAH), such as relationship between molecular weight and intrinsic viscosity, dilution ratio, solubility parameter and solubility, and stability of the $344^{\#}$ resin–CPP-g-MAH–toluene solution are systematically determined to investigate the polar change of graft polymers with grafting MAH onto the chain of CPP. The results obtained will supply a group of foundational data for the graft reaction of MAH onto polyolefins, which is of important value for the molecular design of the MAH graft reaction.

In our previous work, the miscibility of the alkyd resin ($344^{\#}$ resin)–CPP blends was also investigated by the dilute solution properties of polymer blends [6]. The present work will be developed on the basis of the previous study.

EXPERIMENTAL

Materials

Chlorinated polypropylene (CPP), which contains 30% chlorine by weight, is a commercial product purchased from Jin Zhujiang Chemical Factory in Guangdong. Alkyd resin (commercial name is $344^{\#}$ resin) is an industrial product purchased from Zhengzhou Paint Family, which consists of 50% resin and 50% toluene by weight. Maleic anhydride, reaction initiator of benzoperoxide (BPO) and solvents, e.g., xylene, acetone, toluene, tetrahydrofuran, and other reagents, are of A.R. or C.P. grades.

Synthesis of Graft Polymer

Maleic anhydride grafted chlorinated polypropylene (CPP-g-MAH) was synthesized by MAH and CPP in xylene solvent in the presence of benzoperoxide (BPO) as an initiator at 353 to 393 K. Then the graft polymer was separated from the reaction mixture by dropwise addition of a large batch of acetone, and next the precipitated polymer

TABLE I the data of MAH grafted content and chlorine content of CPP-g-MAH samples

No.	*CPP*	*1#*	*2#*	*3#*	*4#*
MAH content (wt%)	0.00	2.20%	2.87%	3.94%	5.83%
Chlorine content (wt%)	30%	28.58%	28.37%	27.94%	27.6%
MAH (mol)/100 g PP	0	0.00032	0.00043	0.0006	0.00089
Cl (mol)/100 g PP	0.01207	0.01163	0.01162	0.01155	0.01167
MAH (mol) + Cl (mol)/100 g PP	0.01207	0.01195	0.01205	0.01215	0.01256

was dissolved in xylene again. The procedure was repeated three times and the crude product of CPP-g-MAH was obtained. Finally the crude product was refluxed with acetone for 24 h in an extractor to remove the poly(maleic anhydride) and remaining monomer of MAH in small quantities and the pure graft polymer was obtained by this procedure. IR spectrum proved that maleic anhydride was effectively grafted onto CPP. The MAH graft content was expressed by weight percent of MAH in the graft polymer and determined by refluxing the graft polymer in the KOH–ethanol solution and then back titration with HCl–isopropanol solution. The chlorine content was determined by a combustion method.

According to the above procedure, four CPP-g-MAH products of different MAH graft content were prepared and their grafting MAH content and chlorine content are listed in Table I.

If mole number of MAH and chlorine is used to substitute for their weight content based on 100 g polypropylene (PP), it can be shown that the sum of mole number of MAH and chlorine are roughly equal to that of the chlorine in CPP before grafting. That is to say, grafting a MAH unit onto a CPP chain will substitute a chlorine atom in CPP chain and MAH will exist in the CPP chain as a single molecular unit, which is related to the fact that the homopolymerization of maleic anhydrides is difficult. This is also consistent with the result reported by Huang [4], who pointed out that most maleic anhydride are grafted onto squalane in the form of a single unit. So CPP-g-MAH molecular structure would be similar to that of CPP.

Viscosity Measurement

Solution viscosities were measured at 298.15 K with a suspended level Ubbelahda Viscometer in four concentrations according to the method of the Ref. [6]. The intrinsic viscosity was obtained by means of Huggins (η_{sp}/C *versus* C) and Kraemer ($\ln \eta_r/C$ *versus* C) extrapolations, where η_{sp} ($= \eta/\eta_0 - 1$) was the specific viscosity and η_r ($= \eta/\eta_0$) the relative viscosity, respectively.

Measurement of Molecular Weight

A DAWN® DSP multi-angle laser photometer from Wyatt Technology Corp. was used as the mass and size detector and a differential refractive index detector, Optilab DSP, was used as the concentration detector. A Model P100 pump (Thermp separation Products, San Jose, USA) and a TSK-GEL G6000HHR column (7.8 mm × 300 mm) completed the equipment. Tetrahydrofuran (THF) freshly distilled from sodium and benzophenone, filtered through a membrane and degassed, was used as eluent at a flow rate of 1.0 mL/min.

The refractive index increment, d*n*/d*c*, in THF solution was measured with a RM-102 differential refractometer at National Research Center for Certified Reference Materials in Beijing. A value of d*n*/d*c* of CPP, 0.0977 mL/g was obtained at 298.15 K.

Dilution Ratio and Solubility Parameter

It is known that toluene is a good solvent and hexane or methanol are nonsolvents for the samples of CPP and CPP-g-MAH. Hexane or methanol was dripped into the toluene solution to turbidity, respectively. In order to accurately determine turbidity of the solutions, the laser-turbidity method [7] was adopted in the experiments, where the toluene solution of 30 mL was exactly taken and its concentration was 8–10 mg/mL. The dilution ratio (nonsolvent volume/toluene volume) and the volume fraction ϕ_i (toluene volume or nonsolvent volume/sum volume after titration) were calculated. The solubility parameter δ_{sm} of mixed solution could be calculated by means of the following formula, $\delta_{\mathrm{sm}} = \phi_1\delta_1 + \phi_2\delta_2$, where ϕ_1, δ_1, ϕ_2, and δ_2 were the volume fraction and solubility parameter of toluene and nonsolvent, respectively. According to Refs. [7,8], the solubility parameter of mixed solvent of toluene and hexane is the lower limit of the sample. The solubility parameter of mixed solvent of toluene and methanol is the upper limit of the sample. The mean value of the lower limit and the upper limit is the point estimation value of the sample.

Measurement of Relative Solubility

The CPP or CPP-g-MAH was put in a special solvent at 298.15 K for three days. The sample was shaken for a certain time and then the solution was filtrated, and the polymer content in the clear solution was determined by weighing method. The polymer solubility was obtained by calculating dissolving polymer quantity for every 10 mL of solvent.

Because of the polymer molecular structure and its complex dissolving process in solvent, it is difficult to exactly determine the polymer solubility. Therefore, the solubility determined under the same conditions will be termed the relative solubility.

Stability of Polymer Solutions

CPP or CPP-g-MAH, which was dissolved in toluene, and $344^{\#}$ paint were blended in various proportions. The mixtures, which contained 70% (wt%) toluene in the solution, were stirred uniformly, and remained stewing about 24 h, after which observation of the stability of the solution was carried out.

RESULTS AND DISCUSSION

Molecular Weight Distributions and Mark–Houwink Equation

The CPP was separated into five fractions using fractional precipitations by solvent/nonsolvent technique. Five fractions, unfractionated CPP and CPP-g-MAH were selected to determine their molecular weights with on-line light scattering measurements

TABLE II The molecular weights and distributions of molecular weights of CPP and its grafted polymers measured by light scattering measurement

No.	M_n	M_w	M_z	Mw/Mn	Mz/Mn	R_n (nm)	R_w (nm)	R_z (nm)	$[\eta]$ (mL/g)
CPP1	299 400	326 900	352 200	1.092	1.176	30.7	32.9	34.9	116.21
CPP2	212 500	254 600	289 300	1.198	1.362	24.6	28.1	30.9	91.32
CPP3	149 100	164 200	176 900	1.101	1.187	19.3	20.9	22.2	71.90
CPP4	119 900	128 900	137 000	1.075	1.143	17.7	18.4	19.1	60.95
CPP5	86 730	91 500	96 000	1.055	1.107	18.1	18.3	18.6	46.32
CPP	93 010	183 400	260 400	1.972	2.799	16.6	24.9	30.7	79.52
$2^{\#}$	105 000	218 700	345 500	2.083	3.291	16.8	24.2	30.5	65.68
$3^{\#}$	108 400	239 400	374 900	2.209	3.46	16.7	25.1	31.5	34.85
$4^{\#}$	112 000	260 400	442 500	2.326	3.952	15.9	24.6	32.5	38.84

and intrinsic viscosities. The results determined are shown in Table II. At the same time, the indexes of molecular weight distributions and the root mean square radiuses of gyration, which were expressed with number-average, weight-average, *Z*-average molecular weight respectively, obtained in the on-line measurements are also given in Table II.

Table II indicated that the fraction of CPP was successful because the molecular weight distributions of its fractions were narrow, where the maximum of the indexes of molecular weight distributions was 1.198 and the others were all less than 1.101. At the same time, the figure of elution volume of CPP in THF *versus* strength of light scattering showed that its peak width of elution in THF was quite narrow, which also proved that the fraction of CPP was successful.

As a norm, the relationship between polymer molecular weights and its intrinsic viscosity $[\eta]$ can be fitted with the Mark–Houwink equation, $[\eta] = KM^{\alpha}$, where K and α are constants specific to the solvent and temperature used in the measurements. In our work, the Mark–Houwink relation for the fractions shown in Fig. 1, of which the linearly dependent coefficient is 0.9964, is expressed as:

$$[\eta] = KM^{\alpha} = 0.0174M^{0.6919} \quad (\mathrm{mL\,g^{-1}}) \tag{1}$$

Generally, the value of α is between 0.5 and 0.8 for a flexible polymer in the special solution, and between 1.8 and 2.0 for a rigid polymer [9]. For the CPP in toluene, the chlorine content of which is 30% (wt%), the fitting value (α) of Mark–Houwink equation is 0.6919, which suggests that the molecules of CPP in toluene exist as a random coil.

If the molecular weight and the intrinsic viscosity of the unfractionated sample of CPP were plotted in Fig. 1, the experimental point was almost located on the fitting line of the Mark–Houwink relation. The result shows that the Mark–Houwink equation is suitable to the polydisperse samples of CPP when chlorine content is about 30% (wt%). Thus, Eq. (1) is of a value for application in the coating industry.

But when the molecular weights and the intrinsic viscosities of the grafted polymers of CPP-g-MAH were also plotted in Fig. 1, it was found that the experimental points all departed from the fitting line of Mark–Houwink relation, and the departing degree of experimental points became greater with the increase of the maleic anhydride content. This shows that the intrinsic viscosity of CPP-g-MAH is less than that of CPP when their molecular weights are the same. This result indicates that the extent

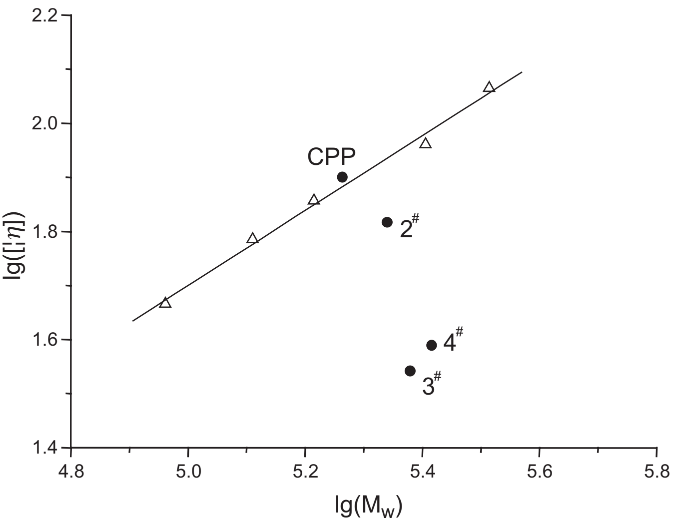

FIGURE 1 The relation of weight average molecular (M_w) and intrinsic viscosity ([η]) of CPP and its graft polymer CPP-g-MAH.

of expansion of CPP-g-MAH molecules dissolved in toluene is less than that of CPP molecules. That is because polarity of grafting polymer molecules increases with grafting MAH onto the chain of CPP, which will result in changes in their existing status in toluene and a dissolving power of toluene which gradually becomes poor.

Dilution Ratio and Solubility Parameter

Samples were dissolved in toluene, and the solutions were respectively titrated to turbidity with hexane and methanol. The experimental results of the dilution ratio and solubility parameters were listed in Table III.

From Table III, it was found that the dilution ratio of hexane to toluene gradually decreased with the increase of the MAH graft content, but the dilution ratio of methanol to toluene gradually increased. Because hexane is an apolar solvent and methanol is a strongly polar solvent, it is clear that molecular polarity also increases with the increase of graft content of MAH onto CPP.

Point estimated values of solubility parameters of the polymers (CPP and CPP-g-MAH) were calculated according to the method reported by the Ref. [7]. The data listed in Table III showed that the solubility parameters of graft polymers increase a little with the increase of MAH graft yield.

Relative Solubility

The experimental results showed that relative solubility of CPP and its graft polymers in the following solvents, such as *n*-hexane, methyl acetate, alcohol, acetic acid, dimethyl sulfoxide, and so on, were less than 0.1 g/10 mL, which indicated that the solvents were poor for them. The results of solubility obtained in the good solvents and the borderline solvents are listed in Table IV.

TABLE III The results of the dilution ratio and solubility parameters ($(\mathrm{MJ/m^3})^{1/2}$) of polymers at 298.15 K (solution volume of toluene: 30 mL)

Resin	*CPP*	$1^{\#}$ *CPP-g-MAH*	$2^{\#}$ *CPP-g-MAH*	$3^{\#}$ *CPP-g-MAH*	$4^{\#}$ *CPP-g-MAH*	$344^{\#}$ *resin*
Hexane/mL	73.3	65.2	58.4	24.2	21.9	31.7
Dilution ratio	2.44	2.17	1.95	0.81	0.73	1.05
Methanol/mL	7.3	7.9	8.3	10.6	10.84	49.3
Dilution ratio	0.24	0.26	0.28	0.35	0.36	1.64
δ_{low}	15.89	15.97	16.05	16.75	16.83	16.53
δ_{high}	20.37	20.50	20.60	21.08	21.13	25.05
$\delta_{\mathrm{average}}$	18.13	18.23	18.33	18.91	18.98	20.79

TABLE IV Solubility of CPP and CPP-g-MAH in different solvents at 298.15 K (g/10 mL solvent)

Solvent	*Solubility parameter*	*CPP*	$2^{\#}$ *CPP-g-MAH*	$4^{\#}$ *CPP-g-MAH*
Poorly hydrogen bonded				
Cyclohexane	16.78	0.53	0.19	0.02
Carbon tetrachloride	17.7	2.40	2.47	0.6
Toluene	18.21	3.23	2.35	1.98
Chloroform	19.85	3.40	2.66	2.59
Tetrachloroethane	20.10	2.69	1.66	0.38
Moderately hydrogen bonded				
Ether	15.57	0.19	0.44	0.68
Ethyl acetate	18.62	0.11	0.21	0.59
Butanone	19.03	0.39	0.50	0.73
Tetrahydrofuran	19.48	2.31	2.53	2.71
Pyridine	21.69	0.87	1.92	2.55
Methylpyrrolidone	22.86	0.17	0.18	0.24

In the coating industry, solvents are often divided into three classes, *viz.* poorly, moderately, and strongly hydrogen bonded, for the sake of convenience to research the dissolving power for resin in various solvents. From Table IV, it was found that the solubility of CPP and CPP-g-MAH in the poorly hydrogen bonded solvents decreased with the increase of the MAH grafted content. Their solubility in the moderately hydrogen bonded solvents increased with the increase of the MAH graft content. Because CPP-g-MAH was insoluble in acetic acid, alcohol etc., this indicated that CPP-g-MAH did not still form strongly hydrogen bonded power in the strongly hydrogen bonded solvents which was consistent with the result by Funasaka *et al.* [5]. So the solubility of CPP and its graft polymers in solvents, which change with the graft content of MAH onto CPP, is also used as a means to characterize their molecular polarity and hydrogen bonded power.

Compared with Table III, it is found that the solubility data of poorly hydrogen bonded solvent are consistent with the range of solubility parameters obtained from mixed solvents. But the solubility in moderately hydrogen bonded solvents is relatively complex in which both acetone (solubility parameter equals 20.25) and methyl acetate (solubility parameter equals 19.64) are insoluble. These results show that using the method of mixed solvents to determine solubility parameter is only a simplified method.

Miscibility of the 344# Resin–CPP-g-MAH Blends

Fan *et al.* [6] reported that the blend of CPP and 344# resin was immiscible if the solution was layered when the two polymers were dissolved in toluene, and that the blend was miscible if the solution was not laminated. In order to investigate the effect of MAH graft content on the miscibility of CPP and 344# resin, a lamination test was carried out. The results obtained are listed in Table V. In Table V, the data of CPP and 344# resin blends came from the Ref. [6], the resin in 344#/resin represents CPP or CPP-g-MAH, and $x_{344\#}$ indicates weight fraction of 344# alkyd resin in the blend of 344# resin and CPP or CPP-g-MAH.

From the results of Table V, it is clear that the solutions are homogeneous when x_{344} is less than 0.5, and the solutions are laminated when $x_{344\#}$ is greater than and equal to 0.5. The solutions of 2# CPP-g-MAH and 344# resin mixture are laminated when $x_{344\#}$ is greater than and equal to 0.667. Furthermore, the solutions of 4# CPP-g-MAH/344# resin are not laminated in the total solution range. The result indicates that the molecular polarity of CPP gradually becomes large with the increase of the graft content of MAH onto CPP, which improves the miscibility of CPP-g-MAH and 344# resin. Therefore, the solution of CPP-g-MAH/344# resin in toluene would be homogeneous in the total range when MAH grafted content is 5.83% and their blends are likely to be miscible.

From the above data listed in Table III, it was observed that the solubility parameters of the polymers (CPP-g-MAH) increased and became gradually close to that of 344# resin with increase of the MAH graft content. Their difference of solubility parameters between ungrafted CPP and 344# resin was maximum and up to $2.84\,(\mathrm{MJ/m^3})^{1/2}$, which led to immiscibility of the two polymers in some mixing ranges and miscibility in other mixing ranges. However, when the MAH grafted content was 5.83% (that is 4# CPP-g-MAH), the difference of solubility parameters between 4# CPP-g-MAH and 344# resin was minimum in the polymer samples and up to $1.81\,(\mathrm{MJ/m^3})^{1/2}$, which indicated that two polymers in toluene would be homogeneous and their blends were likely to be miscible. This result is of significant value in preparing polyolefine coating.

CONCLUSION

Some conclusions may be drawn by systematically determining the solution behavior of CPP and its grafted polymers (CPP-g-MAH). First, the molecular weights of the polymers were determined with on-line light scattering measurements, and the Mark–Houwink equation of CPP in toluene at 298.15 K was also obtained. The result shows that the equation is suitable to the polydisperse samples of CPP which has a chlorine content of about 30% (wt%) but not suitable to predict the molecular weight of CPP-g-MAH.

Second, the relative solubility of CPP and CPP-g-MAH in various solvents shows that the solubility of CPP and CPP-g-MAH in the poorly hydrogen bonded solvents decreases with the increase of the MAH grafted content and their solubility in the moderately hydrogen bonded solvents increases with the increase of the MAH graft content, which is consistent with the result of their dilution ratio. So their solubility in various solvents, which changes with the graft content of MAH onto CPP, is also a quantitative means to characterize their molecular polarity.

TABLE V Stability of 344$^{\#}$ resin–CPP-g-MAH–toluene solutions (toluene content in solution 70% (wt%))

344$^{\#}$/resin	*0/1*	*1/10*	*1/5*	*1/2*	*1/1*	*2/1*	*5/1*	*1/0*
$x_{344\#}$	0	0.091	0.167	0.333	0.5	0.667	0.833	1
CPP	Not lamination	Not lamination	Not lamination	Not lamination	Lamination	Lamination	Lamination	Not lamination
2$^{\#}$ CPP-g-MAH	Not lamination	Not lamination	Not lamination	Not lamination	Not lamination	Lamination	Lamination	Not lamination
4$^{\#}$ CPP-g-MAH	Not lamination	–	–	–	Not lamination	Not lamination	Not lamination	Not lamination

Third, stabilities of the $344^{\#}$ resin–CPP-g-MAH–toluene solutions indicate that the miscibility of CPP-g-MAH and $344^{\#}$ resin was improved with increase of the MAH grafted content. When the MAH grafted content is greater than 5.83%, the blends of CPP-g-MAH and $344^{\#}$ resin become miscible in the total mixing range, which is because the molecular polarity of CPP gradually becomes large with the increase of the graft content of MAH onto CPP.

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References

[1] V.D. Martin (2003). *Macromolecular Symposia*, **202**, 1.

[2] S.X. Li, T.J. Ge, Z.J. Bai *et al.* (1996). *Hecheng Shuzhi Ji Suliao/China Synthetic Resin and Plastics*, **13**(4), 16.

[3] W. Heinen, M. Van Duin, C.H. Rosenmoller *et al.* (1997). Annual Technical Conference – ANTEC, Conference Proceedings. *Materials*, **2**, 2017–2021.

[4] H.L. Huang, Z.H. Yao and J.G. Zhang (2000). *Yingyong Huaxue/Chinese J. of Applied Chemistry*, **17**(3), 256.

[5] T. Funasaka, T. Ashihara, S. Maekawa *et al.* (1999). *International Journal of Adhesion & Adhesives*, **19**, 367.

[6] Z.L. Fan, D.Z. Liu and J.J. Wang (2003). *Physics and Chemistry of Liquids*, **41**(4), 391.

[7] Z.L. Fan, Y. Liu and D.Z. Liu (2003). *Suliao Gongye/China Plastics Industry*, **31**(4), 44.

[8] Polymer Group of Chemistry Department in Beijing University. *Polymer Experimental Science (Chinese)*, pp. 555–558 (Beijing University, 1990).

[9] D.W. Van Krevelen (1990). *Properties of Polymers*, 3rd completely revised edition, pp. 243–282. Elsevier Science Publishers B.V.