Structure and Properties of Impact Copolymer Polypropylene. II. Phase Structure and Crystalline Morphology

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ABSTRACT: In this work, an impact copolymer polypropylene (ICPP) was separated into 4 fractions, A, B, C, and D. The phase structure, thermal behavior, and crystalline morphology of the ICPP and its 4 fractions were studied thoroughly using scanning electron microscopy (SEM). Dynamic mechanical analysis (DMA), differential scanning calorimetry (DSC), and polarized light microscopy (PLM). Results of SEM and DMA show that ethylene–propylene rubber (EPR) and part of the ethylene–propylene segmented copolymer disperse as toughening particles in the ICPP. The size and size distribution of these particles are determined by chain structure of the fractions of ICPP. From fraction A to fraction D, the morphology changes from noncrystalline to semicrystalline gradually, as shown by DSC. DSC results also indicate that thermal behavior of the ICPP agrees greatly with its chain structure. PLM demonstrates that it is difficult for the ICPP to grow perfect spherulites, that is, partially, because the matrix of ICPP, fraction D, has defects in its macromolecular chain. Another cause is that there is a good compatible structure in the ICPP and so the noncrystalline component (including all fractions) hinders the growth of the spherulite. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 71: 103–113, 1999

Keywords: impact copolymer polypropylene; phase structure; crystalline morphology; chain structure; compatible structure

INTRODUCTION

Polypropylene (PP) is an important commercial plastic, but its insufficient low-temperature impact strength significantly hinders its even wider utilization. Blends of PP with elastomers such as an ethylene–propylene (EP) copolymer,[1–3] butyl rubber,[4] styrene butadiene-styrene (SBS) copolymer,[5] EP rubber (EPR),[6,7] and ethylene-pro-

pylene-diene (EPDM) copolymer[8–12] have been widely investigated to improve the impact and tensile properties of PP. Another commercial way to modify PP is by copolymerizing propylene with low content olefins to produce high-impact copolymer polypropylene. It proves to be an effective way and is widely used.[13–15]

High-impact PP, produced either by copolymerizing propylene with other α-olefins or by blending PP with various elastomers, shows biphasic or multiphase structure. The morphology of the product has a great effect on its properties.[1–8,16–23] As is well known, the effectiveness of

Table I   The Content of the Propylene, Ethylene Monomer, and Triad Sequence Distributions

Mark	P	E	PPP	PPE	EPE	PEP	PEE	EEE
CPPB	0.869	0.131	0.807	0.046	0.016	0.010	0.035	0.086
CPPJ	0.865	0.135	0.764	0.068	0.033	0.017	0.042	0.076
Fraction A CPPB	0.547	0.453	0.176	0.203	0.167	0.081	0.195	0.177
Fraction A CPPJ	0.635	0.365	0.300	0.208	0.127	0.142	0.122	0.101
Fraction B CPPB	0.658	0.342	0.470	0.125	0.063	0.033	0.090	0.220
Fraction B CPPJ	0.694	0.306	0.482	0.154	0.058	0.073	0.111	0.123
Fraction C CPPB	0.545	0.455	0.527	0.018	0	0	0.030	0.425
Fraction C CPPJ	0.831	0.169	0.716	0.094	0.025	0.011	0.018	0.140

toughening plastic by blending with rubber depends on the morphology of the blend, such as the shape of the dispersed phase, its size and distribution, the morphology of plastic matrix, and the degree and nature of adhesion between different phases.[16,17] The characteristics of the matrix and the rubber and blending process determine the morphology. The more compatible the blend of rubber and PP, the better the rubber disperses in PP.[1,8]

Impact copolymer polypropylene (ICPP) is a complicated copolymer and is widely used. Since phase structure, crystalline morphology, and thermal behavior affect the products properties greatly, it is necessary to study the phase structure, crystalline morphology, and thermal behavior thoroughly. Our previous work has shown that ICPP has a characteristic chain structure, that is, the composition and sequence structure change gradually between different fractions of ICPP.[24] Considering these, we studied the characteristic phase structure, crystalline morphology and thermal behavior of ICPP and its fractions in present work.

EXPERIMENTAL

Samples

Two commercial *in situ* prepared ICPPs are used in this work. They are purchased from Olefin Chemical Industries Co. Ltd. and Showa Denko K. K. in Japan (CPPJ), and Belgium Neste Chemicals N. V. (CPPB). First they were fractionated into 4 fractions according to the procedure suggested in our previous work.[24] We labeled the original impact copolymer PP as "Original," the precipitate after separating the fraction A from ICPP by xylene as "fraction A′ ", the others are the same as previous work.[24] The composition

and chain structure are listed in Tables I and II. Table III shows their mechanical properties.

Measurements

Polarized Light Microscope

Samples were prepared by fusing a scrap of the sample placed between 2 cover glasses. After melting at 230°C for 2 min, they were kept in quasi-isothermal state at 148°C for 48 h, 140°C for 24 h, and 132°C for 24 h. Fusing and crystallization was carried out in a thermostatic oven. Optical micrographs were taken using an XPT-7 polarized light microscope (Jiangnan Optical Instrument Factory, China).

Scanning Electron Microscope

Using a Hitachi X-650 electron microscope, impact fracture surfaces of notched Charpy specimen ($-20°C$) coated by gold were studied by scanning electron microscopy (SEM).

Dynamic Mechanical Analysis

E' and E'' moduli and their temperature dependence of ICPP were determined by a Tokyo Badwin-DDV-IV-EU dynamic mechanical analyzer. Recording of dynamic mechanical analysis (DMA) spectra followed the known techniques[23] under the following conditions.

Table II   The Content of the 4 Fractions in the Impact Copolymer Polypropylene

ICPP	Fraction A	Fraction B	Fraction C	Fraction D
CPPB	10.8	13.5	2.2	73.5
CPPJ	7.5	12.0	3.0	77.5

CPPJ   CPPB

Figure 1   SEM photographs of the fracture surface of the ICPP.

- Thickness of compression molded plate specimen was 2.5 mm.
- Heating rate was 5°C/min, with the following temperature range: −150 to +150°C.
- Frequency of test was 3.5 Hz.

E'', E', and their ratio, the mechanical loss tangent, were calculated by an on-line computer of the DMA instrument.

Differential Scanning Calorimetry

All the samples were investigated by a Perkin–Elmer DSC-2C scanning calorimeter in an atmosphere of high purity nitrogen. The scanning rate was 20°C/min. The scanning temperature ranges were −60 to 150°C for fraction A and fraction B and 20–180°C for others. The sample weight was 10–15 mg.

Fraction A and fraction B were annealed at 150°C for about 5 min and then quenched to −60°C. Isothermally crystallized samples were directly scanned from 20 to 180°C. Others were annealed at 180°C for about 5 min and then quenched to 20°C. The cooling curves of the original, fraction A′, and fraction D were investigated.

RESULTS AND DISCUSSIONS

Phase Structure of Impact Copolymer PP

Globular particles have been observed on fracture surfaces of ICPPs by SEM, as shown in Figure 1. They are in a toughening phase. SEM photographs demonstrate that the average particle size of CPPB (average diameter $\sim$ 1.6 μm; about 50 particles included) is bigger than that of CPPJ (average diameter $\sim$ 1.0 μm; about 50 particles included) and the distribution of particle size of CPPB is wider than that of CPPJ. It is also shown

Table III   The Mechanical Properties of the Impact Copolymer Polypropylene

ICPP	Melting Index (MI) g/10 min	Yielding Strength (10^5 Pa)	Notched Izod Impact Strength (J/m)	
			25°C	−20°C
CPPB	40	225	52	35
CPPJ	15	251	116	52

Figure 2 Dynamic mechanical analysis spectra of the ICPP: (a) CPPJ; (b) CPPB.

that the fracture surface of CPPJ is coarser than that of CPPB. Corresponding to these aspects, the impact strength of CPPJ is greater than that of CPPB, as shown in Table III. Works[1–8,25] on polymer alloys have shown that for multiphase polymer systems, the toughening effect is determined by 2 factors. First, to some extent, the smaller the particle and the narrower the distribution of particle size, the better the toughened product's impact properties. The small particle and narrow distribution of particle size partially come from the small surface tension between the matrix and the particle, which means good compatibility between its different phases in the multiphase system. Second, the stronger the adhesion between the particle and the matrix, the better the impact properties are. The strong interphacial adhesion also comes from good compatibility. The results of SEM and impact properties have proved that the compatibility between the matrix and the particle of CPPJ is better than that of CPPB. Our previous study has shown that ICPP contains EPR (fraction A, ~ 10 wt %), EP segmented copolymer (fraction B, ~ 10 wt %), EP block copolymer (fraction C, ~ 2 wt %), and fraction D.[24] Certainly, fraction D is the matrix. EPR and part of the EP

segmented copolymer should contribute to the particles detected by SEM. The EP block copolymer and part of the EP segmented copolymer should act as a compatibilizer. Three factors lead to the better compatibility in CPPJ. First, as the chain structure results indicate in our previous work (listed in Tables I and II), the ethylene content in fraction A and fraction B of CPPJ is lower than that of CPPB. Our previous work has also shown that ethylene unit content in fraction D of CPPJ is higher than that of CPPB.[24] So the ethylene units distribute more favorably in the fractions for CPPJ than for CPPB. Second, the component acting as compatibilizer is more in CPPJ than in CPPB. Third, the long propylene segments and ethylene segments (PPP and EEE in Table I) increase more gradually for CPPJ than for CPPB from fraction A to fraction D, which means that chain structures of the 4 fractions are more favorable for the good compatibility in CPPJ.

Dynamic mechanical spectra of impact copolymer PP are shown in Figure 2. One peak appears at about 20°C, which should be the glass transition of PP.[21–23,26–28] Another peak appears at about −40°C, which should be the glass transition of EPR (it may also comprise transition of EP segmented copolymer). The temperatures of the 2 transitions are shown in Table IV. These data again indicate that there are, mainly, 2 phases in ICPP. It is interesting that the peaks of the 2 main relaxations are closer to each other in CPPJ; that is, PP relaxation appears at a lower temperature, and the EPR relaxation peak moves to a higher temperature in CPPJ. EPR relaxation of CPPJ is flatter than that of CPPB, which should be due to the lower content of the EPR and EP segmented copolymer in CPPJ than that in CPPB. For a multiphase system, when its phases are more compatible to each other, they disperse together in smaller scale and their polymer chains' movements affect each other more strongly, so the 2 phases' glass transition temperatures are closer to each other. Therefore, DMA results again prove that the compatibility be-

Table IV   The T_g of the Impact Copolymer Polypropylene Detected by DMA

Item	CPPB	CPPJ
T_g PP (°C)	26	17
T_g EPR (°C)	−46	−33

Figure 3 DSC heating scanning curves of ICPP.

Figure 4 DSC heating scanning curves of CPPJ crystallizing isothermally in an oil oven for about 24 h at different temperatures: (a) 133, (b) 139, (c) 145, and (d) 151°C.

tween the dispersed particle and the matrix of CPPJ is better than that of CPPB. This is corresponding to the difference of the characteristic chain structure between these 2 impact copolymer PPs.

Characteristic Transition of ICPP

Figure 3 shows the heating DSC curves of the ICPP, and the results are listed in Table V. Figures 4 and 5 show the heating DSC curves of ICPPs crystallizing in oil oven at 133, 139, 145, and 151°C for 24 h.

The thermal behavior of the EP block copolymer has been investigated in several works.[29,30] The first transition of the EP block copolymer corresponds to the melting of polyethylene blocks at 130°C, and the second is due to the melting of PP blocks at 162°C according to Ke's observation.[29] Cooling DSC curves of mixed homopolymers show a single exothermic peak, while cooling DSC curves of block copolymer show 2 exothermic peaks at 158 and 119°C.[30]

DSC results demonstrate that the thermal behavior of the ICPP is different from the EP block copolymer, the EP random copolymer, and simple PP blends. Both heating and cooling DSC curves of CPPB show 2 peaks. The strong one should relate to the melting (or crystallizing) of the PP crystal. The other weak one corresponds to the transition of the PE block crystal. But there is only 1 peak for both the heating and cooling curves of CPPJ. The scanning curves of isothermally crystallizing samples are similar to the heating curves of corresponding quenched samples: there are 2 melting peaks for CPPB and 1 melting peak for CPPJ. The melting peaks broaden and move to higher temperature as T_c increases from 133 to 151°C, and their T_m is higher than the T_m of quenched impact copolymer PP. Moreover, the melting DSC curve of CPPB crystallizing at 145°C shows 2 peaks of PP melting, one of which is a shoulder to another. The PP melting peak of CPPB crystallizing at 151°C be-

Table V   DSC Results of the Impact Copolymer Polypropylene

Sample	T_m of PP (°C)	T_m of PE (°C)	T_c of PP (°C)	T_c of PE (°C)
CPPB	165.6	110.1	111.2	84.6
CPPJ	162.6	—	112.3	—

Figure 5 DSC heating scanning curves of CPPB crystallizing isothermally in oil oven for about 24 h at different temperatures: (a) 133, (b) 139, (c) 145, and (d) 151°C.

comes very broad. ICPP comprises multiple components, and in the process of crystallization, EPR, the EP segmented copolymer, and the EP block copolymer are pushed away from the growth front of the crystal to some extent. As the T_c increases, more and more EPR, the EP segmented copolymer, and the EP block copolymer are forced out of the crystal, so there is a more perfect crystal, which melts at comparatively higher temperature; thus, the PP melting peak broadens and moves to higher temperature, as shown in Figures 4 and 5. The DSC results also indicate that the T_m of the CPPB samples is higher than that of CPPJ samples crystallizing at the same conditions. Furthermore, this difference gets obvious as T_c increases. The melting peaks of CPPB crystallizing at 145 and 151°C even show shoulder. On one hand, the difference comes from the fact that the ethylene content in fraction D of CPPJ is higher than that of CPPB, so the chain structure of CPPJ sample is comparatively imperfect. On the other hand, because the difference of chain structure and composition between each 2 fractions of CPPJ is smaller than that of CPPB, the compatibility among the fractions of CPPJ is better than that of CPPB; thus, it is more difficult for EPR, the EP segmented copolymer, and the EP block copolymer to be pushed away from the growth front of the PP crystal in CPPJ than in CPPB.

Figure 6 DSC heating scanning curves of fraction A of the ICPP.

Characteristic Transition of the Fractions of ICPP

To make things clear, it is necessary to check the characteristic transitions of their fractions. Fourier transform infrared (FTIR) results have shown in our previous work that EPR is noncrystalline and the other 3 fractions can crystallize,

Figure 7 DSC heating scanning curves of fraction B of the ICPP.

Figure 8 DSC heating scanning curves of fraction C of the ICPP.

Table VI   DSC Result of Fraction A

ICPP	T_1 (°C)	T_2 (°C)
CPPB	−44.8	36.0
CPPJ	−33.5	41.7

should be attributed to the long PP segment or ethylene segment that may be arranged in a long-range order. This transition temperature of fraction A of CPPJ is higher than that of CPPB. This may result from the PP content and the length of the PP segment and ethylene segment of CPPJ's fraction A being higher than those of CPPB's fraction A.

Figure 7 and Table VII show that fraction B has 1 transition at about 40°C and 2 weak melting transitions. The transition at about 40°C should be due to the motion of polymer segment in the crystalline region, which shows the existence of obvious crystalline imperfection.[27,28] T_{m1} (about 107°C) and T_{m2} (about 130–140°C) should be corresponding to the PE segment and the PP segment crystal's melting respectively. The 2 melting peaks are small and appear at lower temperature than the melting peak of corresponding homopolymer, which means that the crystal in fraction B is very imperfect. Being consistent with the results of FTIR and ^{13}C nuclear magnetic resonance (^{13}C-NMR), DSC results also show that fraction B is an EP segmented copolymer. The second melting peak of CPPB's fraction B is stronger than that of CPPJ's fraction B. The first transition at about 40°C of CPPB's fraction B is weaker than that CPPJ's fraction B. These are due to CPPB's fraction B having a longer PP segment than CPPJ's fraction B.

Figure 8 and Table VII show that fraction C also has 1 glass transition (41–50°C) and 2 melting transitions. The first transition appears at a little higher temperature than that of fraction B,

more or less.[24] Here, their thermal properties are shown again by DSC in Figures 6–9.

Figure 6 shows that for fraction A, the main glass transition appears around −40°C. According to several authors,[21,31–33] it should be the glass transition of EPR rubber. DSC results of fraction A are compiled in Table VI. It can be seen that T_g of CPPJ's fraction A is higher than T_g of CPPB's fraction A, which is consistent with the DMA result. The weak transition at about 36°C

Figure 9 DSC heating scanning curves of fraction D of the ICPP.

Table VII   DSC Results of Fraction B and Fraction C

ICPP		T (°C)	T_{m1} (°C)	T_{m2} (°C)
Fraction B	CPPB	45.6	107.1	137.6
	CPPJ	47.8	105.7	132.2
Fraction C	CPPB	50	119.0	149.4
	CPPJ	44	118.6	149.1

Figure 10   Polarized light microscopy photograph of ICPP crystallizing isothermally at different temperatures: (a) CPPJ at 148°C for 24 h; (b) CPPJ at 140°C for 24 h; (c) CPPJ at 132°C for 48 h; (d) CPPB at 148°C for 24 h; (e) CPPB at 140°C for 24 h; (f) CPPB at 132°C for 48 h.

but it is not so significant as that of Fraction B. T_{m1} is about 119°C and T_{m2} is about 150°C; all of them are about 20°C higher than those of fraction B, but they are still lower than the melting temperature of the corresponding homopolymers. These melting peaks are stronger than those of fraction B. Fraction C has longer ethylene and PP blocks than fraction B, so there is less crystalline imperfection in its crystal, and the melting points of fraction C are higher than those of fraction B.

In DSC curves of fraction D, there is only 1 simple melting peak of PP ($\sim$ 164°C), which is narrower than the original sample's. That confirms that fraction D consists mainly of PP, which is same as the results of FTIR and C^{13}-NMR.

These results show that from fraction A to fraction D, the glass transition becomes weaker and weaker, and the melting peaks become stronger and stronger and move to higher temperature

gradually. The crystallinity and the crystalline perfection increase from fraction A to fraction D. That is corresponding to the change of chain structure from fraction A to fraction D. Moreover, the change of thermal behavior from fraction A to fraction D of CPPJ is somewhat smaller than that of CPPB. This should lie in that the difference of chain structure and composition between CPPJ's fractions is smaller than that between CPPB's fractions. Interestingly, this is corresponding to the good compatibility between dispersed phase and matrix in CPPJ.

The Crystalline Morphology of Impact PP

Polarized light microscopy photographs of isothermal crystallizing ICPP are shown in Figure 10. It is obvious that only CPPB crystallizing at 148°C grew spherulites with black cross. In other

Figure 11 Polarized light microscopy photograph of ICPP fraction A′ (without fraction A) crystallizing isothermally at different temperatures: (a) CPPJ at 148°C for 24 h; (b) CPPJ at 140°C for 24 h; (c) CPPJ at 132°C for 48 h; (d) CPPB at 148°C for 24 h; (e) CPPB at 140°C for 24 h; (f) CPPB at 132°C for 48 h.

conditions, only a mass of crystalline clot is observed. DSC results have shown that the main crystalline component is fraction D. EPR, the EP segmented copolymer, and maybe the EP block copolymer also disperse inter- and intraPP spherulites, the main part of which should be pushed away from the spherulite growth front. So the growth of spherulite is hindered by them. Simple PP blends can form spherulites even at a relatively lower temperature.[1,5,10,18,19] That means that it is more difficult to push away the noncrystalline component from the crystal growth front for the ICPP than for the PP simple blends. So we can deduce that it is more compatible between the fractions of the ICPP than between rubber and PP of simple PP blends. This is consistent with the perfect dispersity of toughening particles, as shown in Figure 1. It has been shown that the improved compatibility in the ICPP

comes from the continuous change of chain structure between its fractions. Fraction C and part of fraction B may act as a compatibilizer. The crystal of CPPJ is comparatively little, due to the good compatibility between the fractions of CPPJ.

To demonstrate the effect of toughening component on the morphology more clearly and surely, we observed isothermal crystalline state of fraction A′ and fraction D using polarized light microscopy (PLM). Their photographs are shown in Figures 11 and 12, respectively. Similar to that of the ICPP shown in Figure 10, as the crystallization temperature (T_c) increases, their spherulites get larger. When fraction A was separated out, the sample (fraction A′) of CPPJ can grow larger crystal, and fraction A′ of CPPB can grow spherulite at relatively low T_c (140°C). Black particles are still obvious intraspherulite, which means that not only fraction A disperses inter-

Figure 12 Polarized light microscopy photograph of the ICPP fraction D crystallizing isothermally at different temperatures: (a) CPPJ at 148°C for 24 h; (b) CPPJ at 140°C for 24 h; (c) CPPJ at 132°C for 48 h; (d) CPPB at 148°C for 24 h; (e) CPPB at 140°C for 24 h; (f) CPPB at 132°C for 48 h.

and intraPP spherulite. The crystallization photograph of fraction D shows that there is almost no black particle seen after fractions A, B, and C being separated away, and its spherulites get perfect, almost similar to iPP. Those suggest that both fraction A (EP rubber), fraction B (EP segmented copolymer), and fraction C (EP block copolymer) comprise the black particles in the crystal. Comparing crystallization photographs of fraction D of CPPB with those of CPPJ, we can see that fraction D's spherulites of CPPB are bigger than that of CPPJ. It may be due to the higher noncrystallizable ethylene content in fraction D of CPPJ.

These results indicate that the crystalline morphology of the ICPP is determined by the chain structure of its fractions. Due to the good compatibility between fractions A, B, C, and the matrix, it is hard to grow big and perfect spherulites in

the ICPP. Apart from the effect of the nucleation agent, the smaller differences of chain structure and composition between the ICPP's fractions lead to better compatibility between the ICPP's fractions and then more little crystal or spherulite in CPPJ.

CONCLUSIONS

From results of DMA, SEM, DSC, and PLM on the ICPP, we come to the following conclusions.

The chain structure of the fractions of the ICPP changes gradually from fraction A to fraction D, so their morphology changes from noncrystalline to semicrystalline. The characteristic chain structure leads to good compatibility between the fractions of the ICPP. Moreover, the smaller the differences of chain structure and morphology

among the fractions of ICPP, the better the compatibility among the fractions of ICPP.

Due to the different morphology and chain structure of its fractions, the ICPP shows a multiphase structure. Fraction A and fraction B disperse as a toughening particle. Fraction C and part of fraction B act as a compatibilizer. Fraction D is the matrix.

Because of the good compatible structure, it is hard to grow spherulites in the ICPP. Noncrystalline components disperse inter- and intraspherulites. As the fraction A, B, and C were separated away one by one, it gets easy to grow spherulite little by little. Furthermore, crystal and spherulites grown in CPPJ are smaller than that grown in CPPB under the same crystallizing condition.

The mechanical properties of the impact copolymer will be discussed in the next work and the relationship of properties to the characteristics of morphology and chain structure will be suggested.

REFERENCES

1. Yang, D. C.; Zhang, B. L.; Yang, Y. K.; Fang, Z.; Sun, G. F. Polym Eng Sci 1984, 24, 612.
2. Jang, B. Z.; Uhlmann, D. R.; Vander Sande, J. B. J Appl Polym Sci 1985, 30, 2485.
3. Van Gisbergen, J. G. M.; Hoeben, W. F. L. M.; Meijer, H. E. H. Polymer 1991, 31, 1539.
4. Van Gisbergen, J. G. M.; Meijer, H. E. H.; Lemstra, P. J. Polymer 1989, 30, 2153.
5. Choudhary, V.; Varma, H. S.; Varma, I. K. Polymer 1991, 32, 2534.
6. Choudhary, V.; Varma, H. S.; Varma, I. K. Polymer 1991, 32, 2541.
7. D'Orazio, L.; Mancarella, C.; Martuscelli, E.; Sticotti, G.; Massari, P. Polymer 1993, 34, 3671.
8. Chiang, W. Y.; Yang, W. D.; Pukanszky, Bela Polym Eng Sci 1992, 32, 641.
9. D'Orazio, L.; Mancarella, C.; Martuscell, E. Polymer 1991, 32, 1186.
10. Prentice, P.; Williams, J. G. Plastics Rubber Proc Appl 1982, 2, 27.
11. Sun, Z. H.; Yu, F. S.; Qi, Y. C. Polym Acad 1991, 2, 143.
12. Bartczak, Z.; Galeski, A.; Martuscelli, E.; Janik, H. Polymer 1985, 26, 1843.
13. Fernando, P. L.; Williams, J. G. Polym Eng Sci 1981, 21, 1003.
14. Narisawa, I. Polym Eng Sci 1987, 27, 41.
15. Bramuzzo, M. Polym Eng Sci 1989, 29, 1077.
16. Manson, J. A.; Sperling, L. H. Polymer Blends and Composites; Plenum Press: New York, 1976.
17. Bucknall, C. B. Toughened Plastics; Applied Science Publishers: London, UK, 1977.
18. Prentice, P. Polymer 1982, 23, 1189.
19. Yeh, P. L.; Birley, A. W.; Hemsley, D. A. Polymer 1985, 26, 1155.
20. Sun, Z.; Yu, F.; Qi, Y. Acta Polym Sinica 2 1991, 143.
21. Wang L.; Huang, B. J Polym Sci Part B Polym Phys 1990, 28, 937.
22. Besomles, M.; Menguel, J.-F.; Delmas, G. J Polym Sci Part B Polym Phys 1988, 26, 1881.
23. Karger-Kocsis, J.; Kiss, L.; Kuleznev, V. N. Polym Commun 1984, 25, 122.
24. Cai, H.-J.; Luo, X.-L.; Ma, D.-Z.; Wang, J.-M.; Tan, H.-S. J Appl Polym Sci to appear.
25. Ming, J. Physical Chemistry in Polymer Alloys; Sichuan Educational Press: Chengdu, China, 1988.
26. Lofthouse, M. G.; Burroughs, P. J Thermal Anal 1978, 13, 439.
27. Frank, H. P. Polypropylene; Gordon and Breach: New York, 1968.
28. Beldwin, E. P.; VerStrate, G. Rubber Chem Technol 1972, 45, 709.
29. Ke, B. Organic Analysis Vol. 4; Interscience: New York, 1960.
30. Inoue, M. J Polym Sci A1 1963, 3427.
31. Paukkeri, R.; Lethtinen, A. Polymer 1994, 35, 1673.
32. Maurer, J. J. Rubber Chem Technol 1965, 38, 979.
33. Ronarc'h, D.; Audren, P.; Moura, J. L. J Appl Phys 1985, 58, 466.