

Review

EFFECTS OF MOLECULE AGGREGATION STATE OF HINDERED PHENOL ON DYNAMIC MECHANICAL PROPERTIES OF CHLORINATED POLYETHYLENE*

Chi-fei Wu*

Institute of Material Science and Technology, East China University of Science and Technology, Shanghai 200237, China

Abstract Hindered phenol compound 3,9-bis[1,1-dimethyl-2[β -(3-*tert*-butyl-4-hydroxy-5-methylphenyl)propionyloxy]ethyl]-2,4,8,10-tetraoxaspiro[5,5]-undecane (AO-80) is a polymorphous material with different physical structures. The initial AO-80 is highly crystalline, whereas AO-80 obtained by cooling from its molten state is an amorphous material. Annealing treatment below the melting point of AO-80 results in structural development. The mixture of chlorinated polyethylene (CPE) and vitrified AO-80 particles exhibits a dramatic change in the dynamic mechanical properties during heat treatment at 130°C. This change can be attributed to the decomposition of the vitrified AO-80 particles and the hybridization of two constituents. The vitrified AO-80 particles can crystallize again in a CPE matrix by annealing at 100°C, but this crystal is different from that of the initial AO-80 in its microstructure. In addition, the incorporation of CPE chains caused a dramatic increase in the modulus. As a result, the AO-80 crystal particles that contain some CPE chains act as multifunctional cross-links and the CPE/AO-80 hybrid was found to be a new type of elastomer.

Keywords Hindered phenol, Polymorphous, Dynamic mechanical properties, Chlorinated polyethylene

INTRODUCTION

The dynamic mechanical properties of elastomeric materials have been discussed extensively in the literature. Many of these studies have been carried out on rubber blends and filler-filled systems. However, much less attention has been paid to the examination of the effects of addition of organic small molecular substances on the dynamic mechanical properties of elastomers.

In recent studies^[1-5], we have extensively investigated the effects of adding organic small molecular substances, such as 3,9-bis[1,1-dimethyl-2[β -(3-*tert*-butyl-4-hydroxy-5-methylphenyl)propionyloxy]ethyl]-2,4,8,10-tetraoxaspiro[5,5]-undecane (AO-80), to some polar polymers such as chlorinated polyethylene (CPE). (AO-80) is a common antioxidant, its chemical structure is shown in Fig. 1 Most of these studies indicated that the aggregation state of small molecule substances plays an important role in determining the dynamic mechanical properties of the matrix polymers.

On the other hand, some studies^[6-8] have revealed that most antioxidants are polymorphous substances with different physical structures (glassy, different crystal modification); and their aggregation states depend strongly on the preparation conditions and the thermal history^[9]. Therefore, control of the aggregation states of small molecules in a matrix polymer is possible.

In this review, the aggregation states of AO-80 were elucidated. Therefore, for a mixture of CPE and vitrified AO-80 particles, the aggregation state of small molecules in a matrix polymer and its influence on

* This work was financially supported by the "The National High Technology Research and Development Program" of the Ministry of Science and Technology of China (the registered number is 2002AA333020).

*Corresponding author: Chi-fei Wu (吴驰飞), E-mail: wucf@ecust.edu.cn

Received October 31, 2002; Revised December 19, 2002; Accepted December 30, 2002

dynamic mechanical properties during heat treatment at a temperature above and below the melting point of AO-80 were extensively investigated in detail.

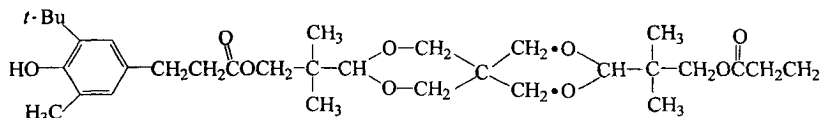


Fig. 1 Chemical structure of 3,9-bis[1,1-dimethyl-2{β-(3-*tert*-butyl-4-hydroxy-5-methylphenyl)propionyloxy}ethyl]-2,4,8,10-tetraoxaspiro[5,5]-undecane, which is abbreviated as AO-80

MICROSTRUCTURE OF PURE AO-80^[10, 11]

The thermal properties of initial AO-80, vitrified AO-80 and vitrified AO-80 annealed at 100°C are shown in Fig. 2. The initial AO-80 showed a very large melting peak with a maximum at 119°C and $\Delta H = 97$ J/g, while the vitrified AO-80 showed a second-order transition at 42°C. It was found that heat treatment at 100°C caused a change from an amorphous state to a crystalline state. With an increase of annealing time (up to 134 h), the specific heat of the second-order transition at 42°C decreased, whereas two other melting peaks increased. The temperature positions of the two melting peaks are notable. The higher melting peak is higher than the melting point (T_m) of initial AO-80, whereas the lower melting peak is lower than T_m . The results suggest that AO-80 exhibits at least three solid phases (polymorphs). With a further increase in annealing time up to 153 h, the lower melting peak appeared as a smaller shoulder, but the higher melting peak was enhanced and shifted to a higher temperature, suggesting that there is a structural transformation from a relatively unstable crystal to a more stable crystal during annealing (134–153 h). When the annealing time was further increased, the higher melting peak became higher and sharper. A comparison of the melting enthalpy of vitrified AO-80 annealed at 100°C for 588 h (68 J/g) and that of the initial AO-80 (97 J/g) showed that the crystallization of AO-80 from an amorphous state at 100°C is incomplete even after 588 h. In summary, the process in annealing treatment for 588 h can be classified into four regimes: (1) formation of an ordered intermediate structure (~ 74 h)^[16], (2) growth of two kinds of crystals (74–134 h), (3) transition from a relatively unstable crystal to another more stable crystal (134–193 h) and (4) further growth of a highly stable crystal (~ 193 h).

In order to determine the effects of annealing temperature, annealing treatment of vitrified AO-80 at 80°C was also carried out. Figure 3 shows the change in thermal behavior of vitrified AO-80 as a function of annealing time. The sample annealed at 80°C for 50 h showed an endothermic peak just above the glass transition temperature, a shoulder and an endothermic (melting) peak. This characteristic implies that AO-80 in this case is a medley of several elements containing glass, paracrystal (shoulder) and crystals (endothermic peak). The tendency of the change in DSC curves caused by more prolonged annealing is similar to that in the case of annealing at 100°C. However, the T_m of the crystal formed at 80°C is lower. This result suggests that AO-80 has many crystalline modifications.

WAXD traces were recorded to confirm the existence of many crystalline modifications. Figure 4 shows the X-ray diffraction traces of various samples. It was found that the diffraction profiles of the initial AO-80, vitrified AO-80 that were annealed at 80°C and 100°C are very different. The initial AO-80, which was cultured from a dilute solution, showed many stronger diffraction peaks, and the trace consisting of these peaks is a typical diffraction profile of a crystal of a low-molecular-weight compound. In the case of vitrified AO-80 annealed at 100°C for 134 h, its diffraction trace was slightly different from those samples annealed for 153 h and 588 h, supporting the speculation that the crystalline modifications corresponding to the two melting peaks (108 and 120°C) shown in Fig. 2 are different. In addition, the diffraction trace of vitrified AO-80 annealed at 80°C was not the same as that of vitrified AO-80 annealed at 100°C or the same as that of initial AO-80. This result also verified that AO-80 has many crystalline modifications.

A brief summary of the structure of AO-80 is given below. AO-80 is a polymorphous material. The initial AO-80 is a complete crystal, whereas AO-80 obtained by cooling from its melting state is an amorphous solid. The vitrified AO-80 particles can crystallize again by annealing treatment, and the structure of this crystal depends on the annealing condition.

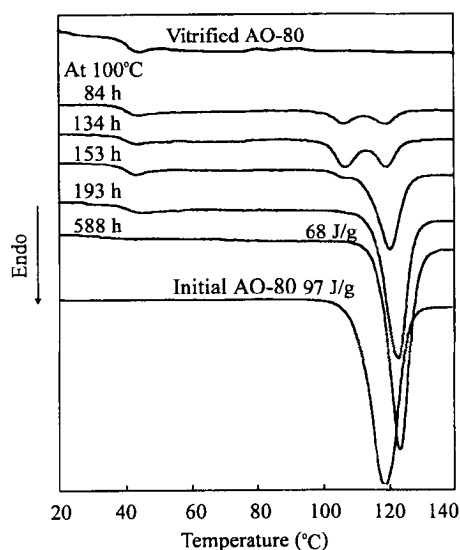


Fig. 2 DSC curves of vitrified AO-80, initial AO-80, and vitrified AO-80 annealed at 100°C

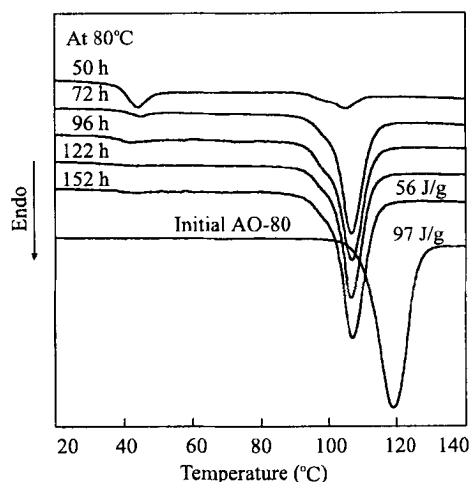


Fig. 3 DSC curves of vitrified AO-80, initial AO-80, and vitrified AO-80 annealed at 80°C

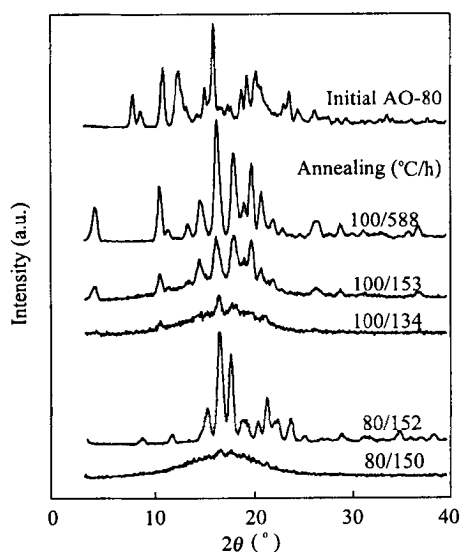


Fig. 4 Wide-angle X-ray diffraction traces of initial AO-80 and vitrified AO-80 annealed at 80 and 100°C

STRUCTURAL CHANGE OF VITRIFIED AO-80 IN MATRIX CPE DURING THERMAL TREATMENT AT 130°C AND ITS INFLUENCE ON THE DYNAMIC MECHANICAL PROPERTIES^[12]

The dynamic mechanical spectra for CPE/vitrified AO-80 (60/40) is shown in Fig. 5. There are two significant peaks in the loss modulus E'' and loss tangent $\tan\delta$ curves and there are two dispersion regions in the storage

modulus E' over the entire experimental temperature range. Hereinafter, these two transitions are defined as β and α transitions at low and high temperature, respectively. The β transition can be attributed to the glass transition of the continuous CPE phase, while the α transition is associated with the dispersed AO-80 domain.

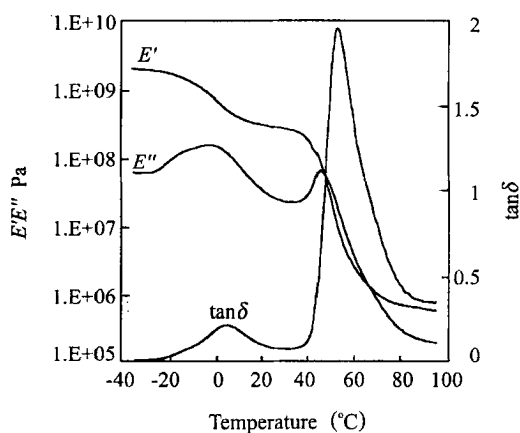


Fig. 5 Temperature dependence of E' , E'' , and $\tan\delta$ of CPE/vitrified AO-80 (60/40) blend at 11 Hz

A series of heat-treated samples were prepared for studying the effects of heat treatment on the dynamic mechanical properties of a mixture of CPE and vitrified AO-80 having a 60/40 composition. To obtain the heat-treated samples, the mechanical mixes of CPE and vitrified AO-80 were placed in a constant temperature chamber at 130°C and were rapidly quenched in an ice-water bath. The dynamic mechanical properties of these heat-treated samples are presented in Fig. 6. As shown in Fig. 6(c), the β peak monotonously increases and shifts to a high temperature as the heat-treatment time increases. Nishi *et al.*^[13] extensively investigated the changes of dynamic mechanical properties of poly(vinyl chloride) (PVC)-copolyester thermoplastic elastomer (Hy) mixtures during the heat treatment process. They found that for heat-treated samples, the α peak corresponding to the glass transition of Hy shifted to a high temperature but its height became lower as compared with melt-blended and solution-blended samples, and they attributed this change to a phase separation. Therefore, changes in dynamic mechanical properties due to the heat treatment at 130°C cannot always be attributed to phase separation, suggesting that there may be some other mechanisms. Because the dissolution of some of the AO-80 molecules in the CPE matrix can cause an increase in the β peak of a CPE/AO-80 hybrid^[3], it is most likely that the vitrified AO-80 particles dispersed in the CPE matrix have undergone dissolution upon isothermal annealing.

In addition, the change in the α peak caused by heat treatment is also remarkable. The untreated sample showed a very large peak. With increases in annealing time (up to 0.5 min), the α peak became small and shifted to a lower temperature. However, with further increases in annealing time (from 1 to 5 min), the α peak can resolved into two peaks, which are designated as α_1 and α_2 , respectively. Furthermore, when the annealing time was extended to 120 min, the α_1 peak became smaller, whereas the α_2 peak barely changed, and is the same as the α peak of melt-quenched sample. Therefore, the α_2 peak is derived from the dissociation of the intermolecular hydrogen bonding between the α -hydrogen of CPE and the hydroxyl groups of AO-80 within the AO-80-rich domain.

Here, let us consider the molecular mechanism of the α_1 peak, which is still unclear. We noted that the locations of the two α peaks were very different. This difference may be attributed to the structural difference of the vitrified AO-80 particles dispersed in the CPE matrix. It is assumed that the α_1 peak arises from pure AO-80 particles. This is conceivable because a relatively large heterogeneous AO-80-rich particle can be resolved into much smaller particles including pure AO-80 particles and even AO-80 molecules during the heat treatment process.

The effects of heat treatment on the compatibility between two components can be evaluated from the

variation in E'' with respect to temperature as presented in Fig. 6(b). However, because the relaxation spectrum $H(1/T)$ is approximately given by $E''(1/T)^{[14]}$, the discussion below will be restricted to $E''(1/T)$. Fig. 7 shows a plot of normalized E'' against reciprocal absolute temperature. It was found that as the heat treatment time increased, the peak in the $E''(1/T)$ curves corresponding to AO-80 was considerably broadened, indicating that the compatibility had been greatly enhanced. The enhancement of compatibility means that the heat treatment not only induces the dissolution of some of the AO-80 molecules into a continuous CPE phase but also the incorporation of some CPE chains into the AO-80 particles.

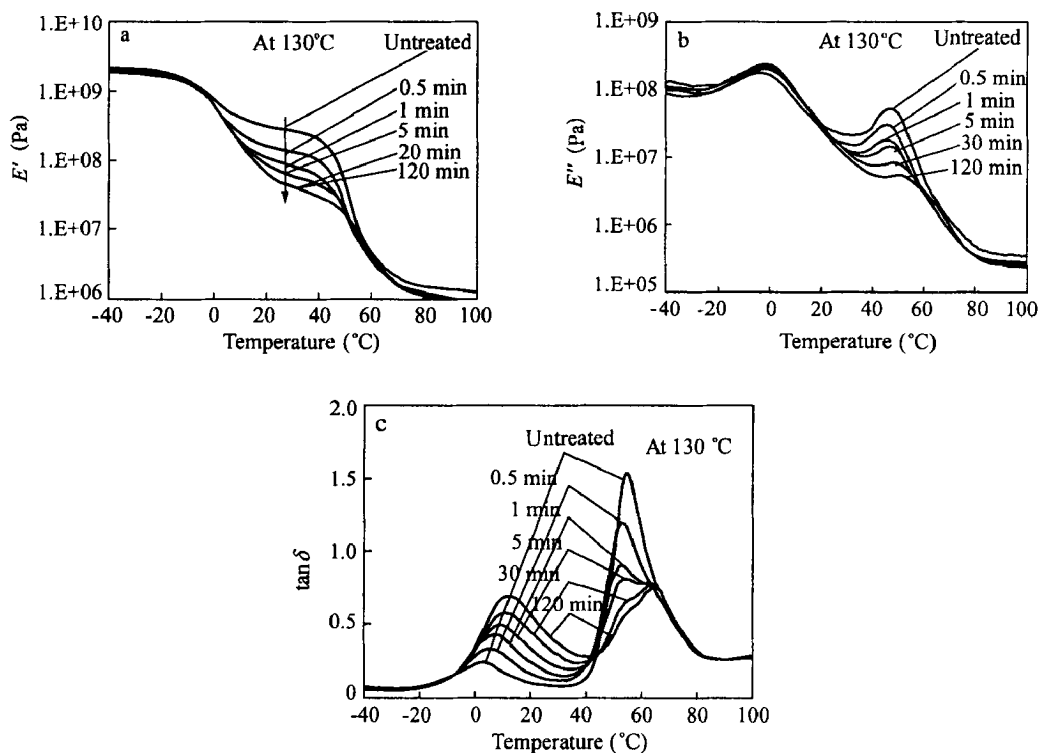


Fig. 6 Temperature dependence of (a) E' , (b) E'' , and (c) $\tan \delta$ for CPE/vitrified AO-80 (60/40) annealed at 100°C at 11 Hz

We have thus established from the analyses of the dynamic mechanical properties that the heat treatment process can be described by the decomposition of vitrified AO-80 particles and the hybridization of two constituents. This proposition will be further substantiated by the results of analysis of infrared spectra and SEM observations.

It is conceivable that the hybridization of two constituents resulting from heat treatment should cause significant changes in the nature and number of some hydrogen bonds. To confirm such changes, we performed FT-IR measurements of various samples. The spectrum of AO-80 shows two significant absorptions of the hydroxyl group (OH) stretching region ($3300\text{--}3700\text{ cm}^{-1}$) and the carbonyl group ($\text{C}=\text{O}$) stretching region ($1650\text{--}1850\text{ cm}^{-1}$). Since the bands for the $\text{C}=\text{O}$ group stretching vibration are difficult to interpret (too abundant), we focus our attention to the OH group-stretching region ($3300\text{--}3700\text{ cm}^{-1}$).

Figure 8 shows the infrared spectra centered near 3500 cm^{-1} corresponding to OH groups for the heat-treated samples. A salient feature of the spectrum of pure AO-80 is the presence of two bands associated with the OH group stretching vibration. The band at 3555 cm^{-1} is assigned to "free" (non-hydrogen-bonded) OH groups, whereas the band at 3470 cm^{-1} is assigned to "OH---OH" (hydrogen-bonded) interaction between AO-80 molecules. For the heat treated samples, another additional band clearly appeared at 3503 cm^{-1} , and this band can be attributed to an intermolecular hydrogen bond ($\alpha\text{H}\text{---}\text{OH}$). This illustrates that the domains of AO-80 within

the CPE matrix include CPE segments. As the heat treatment time increased, the band due to free OH and the band due to OH---OH decreased, while the intermediate band due to α H---OH increase. This result can be considered to be evidence of the hybridization of two constituents resulting from heat treatment.

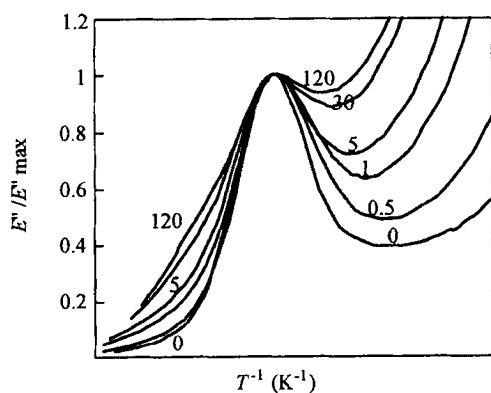


Fig. 7 Reciprocal absolute temperature dependence of normalized E'' for CPE/vitrified AO-80 (60/40) annealed at 100°C at 11 Hz

The numbers shown in the figure are heat-treatment time (min).

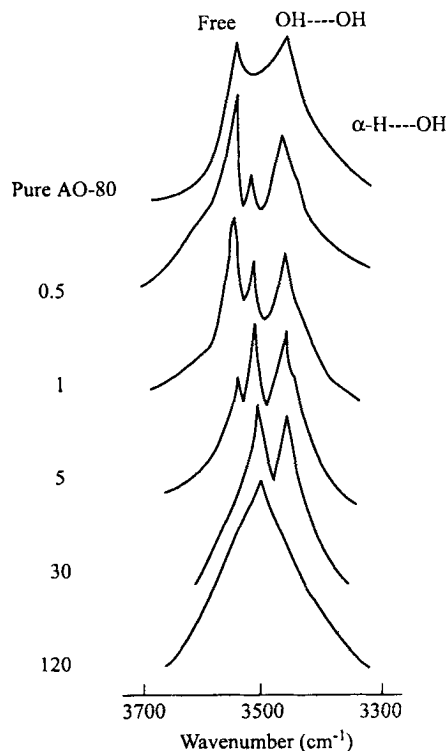


Fig. 8 Infrared spectrum of pure AO-80 and CPE/vitrified AO-80 (60/40) annealed at 100°C in the hydroxyl-stretching region

The numbers shown in the figure are heat-treatment time (min).

In addition to the hybridization of two constituents, the decomposition of vitrified AO-80 particles during the heat treatment process was also confirmed by electron microscopy. Figure 9 shows SEM micrographs of sample C. In an untreated sample, the vitrified AO-80 particles were clearly observed as white agglomerates ($> 10 \mu\text{m}$) consisting of relatively small primary particles ($2\text{--}5 \mu\text{m}$). As the heat treatment time increased, the sizes of the AO-80 particles significantly decreased, and they also became spherical in shape. Thus, decomposition of vitrified AO-80 particles due to heat treatment can be admitted by the existence of these facts.

STRUCTURAL CHANGE OF VITRIFIED AO-80 IN MATRIX CPE DURING ISOTHERMAL ANNEALING AT 100°C AND ITS INFLUENCE ON THE DYNAMIC MECHANICAL PROPERTIES^[15, 16]

Figure 10 shows the change in melting behavior of CPE/vitrified AO-80 (60/40) as a function of annealing time. It was found that annealing at 100°C caused a greater change in the aggregation state of AO-80 molecules within the CPE matrix. With an increase in annealing time (up to 60 min), the melting peak near 100°C increased in conjunction with a abrupt drop near 40°C due to the glass transition of amorphous AO-80. When the annealing time was extended to more than 120 min, the abrupt drop due to the glass transition of amorphous AO-80 disappeared, and the increase in the melting peak of AO-80 crystals stopped. Therefore, the value of the heat of fusion of CPE/vitrified AO-80 sample annealed at 100°C for 120 min can be regarded as a saturation value of

AO-80 crystal formed within the CPE matrix. This value is 64 J/g, which is smaller than either 97 J/g of the initial AO-80 or 68 J/g of the vitrified AO-80 treated at 100°C for 588 h. In addition, the melting points of the initial AO-80, the vitrified AO-80 treated at 100°C for 588 h and AO-80 crystals that formed within CPE matrix by annealing treatment are 119, 123 and 95–104°C, respectively. These results indicate that the form of the AO-80 crystal that formed within the CPE matrix is very different from the crystal form of either the initial AO-80 or the vitrified AO-80 treated at 100°C.

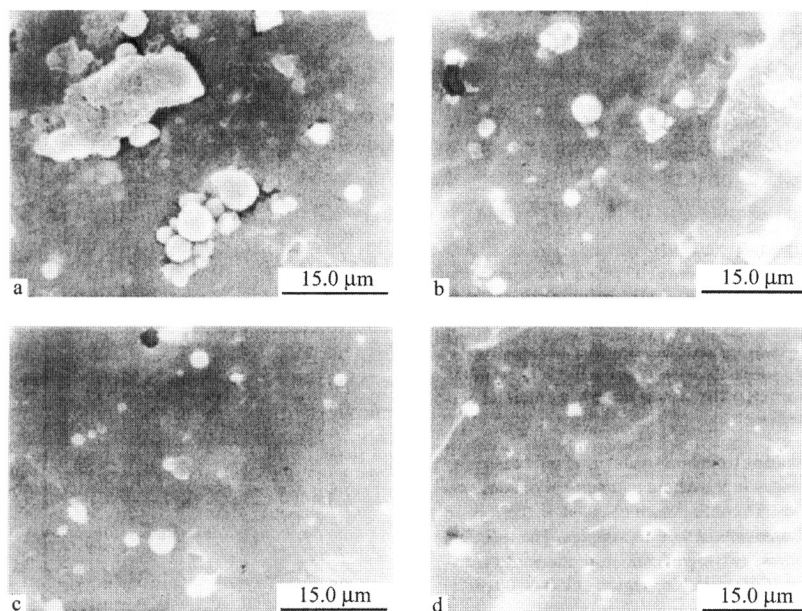


Fig. 9 SEM micrographs of CPE/vitrified AO-80 (60/40) treated at 130°C for (a) no treatment, (b) 1 min, (c) 5 min and (d) 120 min

WAXD traces were recorded to confirm the differences in the forms of the initial AO-80, the vitrified AO-80 treated at 100°C for 588 h and AO-80 crystals that formed within the CPE matrix by annealing treatment at 100°C for 22 h. Figure 11 shows the X-ray diffraction traces of these three samples. The WAXD trace obtained from a sample annealed at 100°C for 22 h was very different from both that of the initial AO-80 and that of the vitrified AO-80 treated at 100°C for 588 h. This finding supports the speculation that AO-80 is a polymorphous compound.

It is also notable that the crystallinity of AO-80 obtained from DSC and WAXD is different. As can be seen in Fig. 10, the particles of vitrified AO-80 within the CPE matrix recrystallized considerably during the annealing process. The saturation value of AO-80 crystal formed within the CPE matrix is 64 J/g, indicating that 94% of AO-80 molecules within the CPE matrix recrystallized during annealing at 100°C for 120 min. In contrast, the diffraction peak of a sample annealed at 100°C for 22 h ($>> 120$ min) is very small compared to those of the initial AO-80 and the vitrified AO-80 treated at 100°C, as shown in Fig. 11. For polymer systems, a broadening of the diffraction profile can be accounted for by either small crystallite size or distortion of the crystallographic lattice (or both)^[17]. This difference in crystallinity due to measurement method suggests that the AO-80 crystal formed within the CPE matrix is very imperfect.

More careful examination of the melting peak temperature of the AO-80 crystal that formed within the CPE matrix is needed. Although this temperature increased with increasing annealing time, it is still lower than the melting points of the initial AO-80 (119°C) and the vitrified AO-80 treated at 100°C for 588 h (121°C). The decline in the melting peak temperature of the AO-80 crystal that formed within the CPE matrix can be explained in terms of molecular interaction. To elucidate the nature of the interaction between CPE/AO-80, various samples

were subjected to FT-IR measurement.

In Fig. 12, the infrared absorption bands centered near 3500 cm^{-1} correspond to OH groups of the initial AO-80, the vitrified AO-80A, and various annealing samples. The band at 3555 cm^{-1} is assigned to “free” (non-hydrogen-bonded)OH, whereas the band at 3470 cm^{-1} is assigned to “OH---OH” (hydrogen-bonded) between AO-80 molecules. In contrast, for the vitrified AO-80, these bands disappeared, but another broader band clearly appeared at 3498 cm^{-1} . The appearance of the broader band implies that there may be many intermolecular interactions. For AO-80 recrystallized within the CPE matrix, with an increase in annealing time, the band (OH---O=C of AO-80) at 3498 cm^{-1} gradually lowered, whereas two new bands appeared at 3644 and 3612 cm^{-1} . The band at 3644 cm^{-1} is assigned to “free” (non-hydrogen-bonded)OH, whereas the band at 3612 cm^{-1} is assigned to “OH---OH (hydrogen-bonded)” between AO-80 molecules, which formed a crystal within the CPE matrix. As shown by Pimentel *et al.*^[18], the numerical value of band shift observed for ν_{OH} vibration can be roughly correlated with the O—H---O distance. The frequency of the IR stretching vibration band of both crystalline and annealed AO-80 thus gives a rough estimate of the dissociation energy of the hydrogen bond formed. A comparison of the two bands (3470 and 3555 cm^{-1}) of the initial AO-80 and the two bands (3612 and 3644 cm^{-1}) of AO-80 recrystallized within the CPE matrix shows that the vibration frequencies of AO-80 crystals formed within the CPE matrix are larger than the frequencies of the initial AO-80. This implies that the strength of the intermolecular hydrogen bond of AO-80 crystals formed within the CPE matrix is weaker than that of the initial AO-80. Thus, the fall of melting temperature of the AO-80 crystal that formed within the CPE matrix can be accounted for by the decrease in the strength of the OH---OH hydrogen bond. This is reasonable because the incorporation of some chains of CPE within an AO-80 crystal causes an increase in the intermolecular distance of AO-80 molecules.

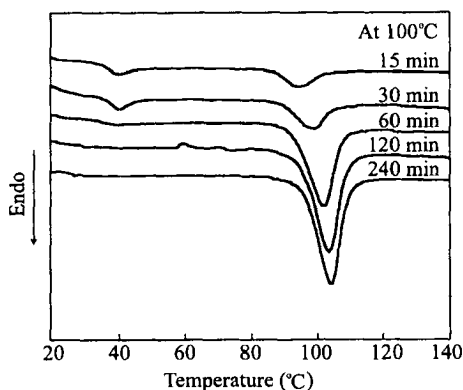


Fig. 10 DSC curves of CPE/vitrified AO-80 (60/40) annealed at 100°C
Scan rate was 10 K/min

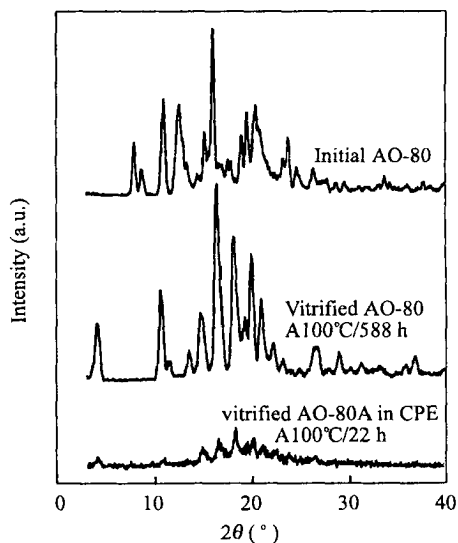


Fig. 11 Wide-angle X-ray diffraction traces of the initial AO-80, the recrystallized AO-80 (the vitrified AO-80 annealed at 100°C for 588 h) and vitrified AO-80 within CPE annealed at 100°C for 22 h

Figure 13 shows the effects of annealing treatment at 100°C on the temperature dependence of the storage modulus E' and tangent ($\tan\delta$) for the annealed CPE/vitrified AO-80 (60/40) samples. Untreated samples clearly exhibited two transitions, at about 7°C and 51°C , respectively. The first transition is attributed to the glass transition of a CPE-rich phase, and the second transition is attributed to the dissociation of intermolecular hydrogen bonds within the AO-80-rich domains. With an increase in annealing time (up to 60 min), the second

transition decreased but a new transition near 100°C appeared. The third transition is thought to be due to melting of the AO-80 crystal that formed within the CPE matrix during annealing treatment. We here focus on the influence of annealing treatment on the second transition.

There was no great change in the first peak of $\tan\delta$ with change in the annealing time, but the second peak decreased monotonously with an increase in annealing time. Since the second transition is associated with intermolecular hydrogen bonds between CPE and AO-80 within the AO-80-rich domain, the observed reduction in the second peak seems to be due to the decrease in the number of intermolecular hydrogen bonds during the crystallization process of AO-80 molecules due to annealing. The results demonstrated that the second transition caused by the dissociation of intermolecular hydrogen bonds was controlled through annealing treatment so that the number of intermolecular hydrogen bonds changes.

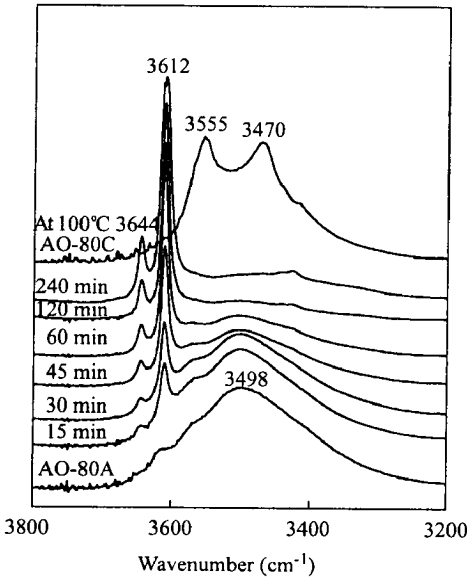


Fig. 12 Infrared spectra of the initial AO-80, the vitrified AO-80 and vitrified AO-80 within CPE annealed at 100°C in the stretching vibration frequency region of O—H groups

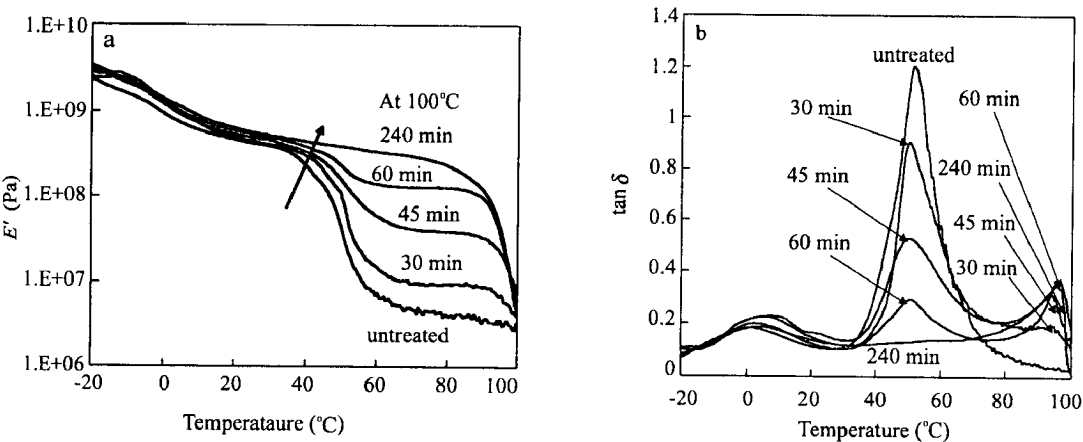


Fig. 13 Temperature dependence of (a) E' and (b) $\tan\delta$ for untreated CPE/vitrified AO-80 (60/40) and its annealed samples at 11 Hz

It should be noted that there was almost no change in the position of the second peak for all samples,

indicating that neither annealing time nor temperature influences the second transition temperature. This implies that the second transition temperature depends on the strength of the intermolecular hydrogen bonding but not on the number of intermolecular hydrogen bonds, which is thought to determine the height of the second peak.

The effects of annealing treatment on the temperature dependence of E' of annealed samples is also important for understanding the microstructure of the AO-80 crystal formed within the CPE matrix. A previous study^[12] on the changes in E' of CPE filled with vitrified AO-80 particles during annealing at temperature above the melting point of AO-80 has shown that because the vitrified AO-80 particles effectively act as multifunctional cross-links, a rubbery plateau can be observed in the modulus vs temperature curves at a temperature below the glass-transition temperature (T_g) of AO-80, but above T_g of the CPE matrix. The heat-treated samples showed a slight increase in modulus below the T_g of AO-80. This is thought to be due to the hardening of molecular chains accompanied by the crystallization of AO-80 molecules that dispersed within the CPE matrix. In addition, the second dispersion gradually decreased with an increase in the annealing time. This tendency agreed with the DSC observation of the second transition (Fig. 10).

The finding on the incorporation of some chains of CPE within AO-80 crystal is also very interesting from an industrial point of view, because the CPE cross-linked by an AO-80 crystal containing some CPE chains can be considered to be a new type of elastomer.

REFERENCES

- 1 Wu, C.F., Chin. J. Polym. Sci., 2001, 19: 455
- 2 Wu, C.F. and Akiyama, S., Chin. J. Polym. Sci., 2002, 20: 119
- 3 Wu, C.F., Yamagishi, T., Nakamoto, Y., Ishida, S., Nitta, K. and Kubota, S., J. Polym. Sci., Part B: Polym. Phys., 2000, 38: 2285
- 4 Wu, C.F., Mori, K., Otani, Y., Namiki, N. and Emi, H., Polymer, 2001, 42: 8289
- 5 Wu, C.F., Nomura, T., Mihara, T. and Hashimoto, K., U. S. Pat. 09/363749
- 6 Foldes, E., J. Appl. Polym. Sci., 1994, 51: 1581
- 7 Foldes, E. and Lohmeijer, J., J. Appl. Polym. Sci., 1997, 65: 761
- 8 Hasui, H., Furuhashi, T., Takanashi, K. and Omori, M., 1985, Jpn Patert 13018
- 9 Foldes E. and Turcsanyi, B., J. Appl. Polym. Sci., 1992, 46: 507
- 10 Wu, C.F., J. Non-Cryst. Solids, 2002, 315: 321
- 11 Wu, C.F., Polymer, 2003, 35: 286
- 12 Wu, C.F., Akiyama, S., Mabuchi, T. and Nitta, K., Polym. J., 2001, 33: 792
- 13 Nishi, T., Kwei, T.K. and Wang, T.T., J. Appl. Phys., 1975, 46: 4157
- 14 McIntyre, A.D., J. Appl. Polym. Sci., 1963, 7: 1291
- 15 Wu, C.F. and Akiyama, S., J. Polym. Sci., Part B: Polym. Phys., in press
- 16 Wu, C.F. and Akiyama, S., Polym. J., 2002, 34: 847
- 17 Buchanan, D.R. and Miller, R. L., J. Appl. Phys., 1966, 37: 4003
- 18 Pimentel, G.C. and Sederlorn, C. H., J. Chem. Phys., 1956, 24: 639