

Review

ENHANCEMENT OF DAMPING PERFORMANCE OF POLYMERS BY
FUNCTIONAL SMALL MOLECULES

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Abstract The addition effects of organic small molecular substances *N,N'*-dicyclohexyl-benzothiazyl-2-sulfenamide (DZ) and 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) on the dynamic mechanical properties of chlorinated polyethylene (CPE), chlorinated polypropylene (CPP), acrylate rubber (ACM) and their blends were investigated. In the case of compatible systems such as CPE/DZ and ACM/AO-80, the height of the loss tangent ($\tan\delta$) peak of a matrix polymer (CPE or ACM) increases, and its peak position shifts to a higher temperature with the addition of DZ or AO-80. By contrast, for incompatible CPE/AO-80, a novel transition appeared above the glass transition temperature of CPE. This additional transition was assigned to dissociation of the intermolecular hydrogen bond between the α -hydrogen of CPE and the hydroxyl groups of AO-80 within the AO-80-rich domain. This will provide a new concept for developing damping material. However, the minimum value between two $\tan\delta$ peaks is lower. It was found that the temperature dependence of $\tan\delta$ could be improved by adding chlorinated paraffin (CP) or ACM to CPE/AO-80. In addition, another ternary system of ACM/CPP with more AO-80 was found to be a very good self-adhesive damping material because of the appearance of a novel transition due to an interfacial layer of ACM/CPP.

Keywords Organic hybrid, Second-order transition, Damping, Hydrogen bond, Self-restoration

INTRODUCTION

During recent years there has been an increased interest in the use of polymers for low-frequency vibration and sound wave damping in the sonic and ultrasonic ranges. This is because noise has become a serious form of pollution. Materials with high damping properties therefore find numerous applications, especially in aircraft, automobiles, skyscrapers, and appliance industries. Polymers are useful damping materials near their glass-transition temperature where coordinated chain molecular motion dissipates mechanical energy as heat. Based on the damping theory^[1–5], the magnitude of damping can be determined from dynamic mechanical behavior. The loss modulus E'' or the loss tangent $\tan\delta$ is a measure of damping properties. From an engineering point of view, there are basically two kinds of damping modes of interest when using polymers. A large E'' value is desired for extensional damping, while a large $\tan\delta$ value is desired for constrained-layer damping. Whenever any type of constraining layer is applied on the top of a damping layer, the constrained-layer damping is likely to exceed the extensional damping. This is because the shear deformation in the damping layer with constrained layers dissipates more energy than the bending deformation in an extensional damping layer. Therefore, in the present study, our focus was limited to how to enhance the $\tan\delta$ value of some polymers or their blends.

In general, the glass transition temperature of a given homopolymer at a given frequency can be shifted within a certain range by many conventional means^[6], such as the addition of plasticizers and fillers, and copolymerization. However, the value of $\tan\delta$ peak due to glass transition becomes relatively lower for conventional polymeric damping materials.

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Received July 2, 2001; Revised August 27, 2001; Accepted September 3, 2001

In addition, homopolymers generally have narrow glass-transition regions (20–30°C)^[7]. However, in real damping applications, wide variations of temperature and frequency are usually experienced; thus, damping systems with a high and broad $\tan\delta$ range are required. In general, for outdoor or machinery applications, good damping materials should exhibit a high loss factor ($\tan\delta > 0.5$) over a temperature range of at least 60–80°C^[8]. The height and width of the loss peak for a given polymer are mutually related. Therefore, it is impossible to obtain a loss peak that is both high and wide at the same time^[9]. Consequently, the narrow temperature range of the glass transition causes most homopolymer damping materials to fail^[10]. One solution to this problem lies in the selection of a multi-component polymer system with controlled degree of miscibility. Copolymers^[11] and mechanical blends^[12] have been used as the multi-component system, but dynamic mechanical spectroscopy has often shown limited damping capability as indicated by two narrow peaks. Another method of broadening the damping peaks is through the use of interpenetrating polymer networks (IPNs)^[13–21]. However, so far, the polymer blends and IPN are restricted to the general glass transition. To obtain a better damping property, a new concept for designing damping materials is needed.

On the other hand, the use of hydrogen bonding for designing multiphase materials has recently attracted attention because a wide variety of dynamic functional systems can be obtained by such an approach^[22]. We considered here a new blend system consisting of polar polymer and functional organic small molecule based on a concept of the sophisticated use of hydrogen bonds in order to develop a new damping material organized by the intermolecular hydrogen bonds. In the present study, we extensively investigate the additive effect of functional organic small molecules on the dynamic mechanical properties of polar polymers. Thereupon, to obtain a broader damping property, we suggest some ways to modify the damping properties of an organic hybrid consisting of CPE and hindered phenols by a third compound such as chlorinated paraffin and acrylate rubber. Finally, we attempted to develop a new type of super-damping material with self-adhesiveness.

EFFECTS OF SMALL-MOLECULE ADDITIVES ON DYNAMIC MECHANICAL PROPERTIES OF POLYMERS^[23–28]

The polymers used were chlorinated polyethylene (CPE, with chlorination degree of 40 wt%, acrylate rubber (ACM), chlorinated polypropylene (CPP, with chlorination degree of 26 wt%). The additives used were some multi-functional organic small molecular substances such as amines and phenols, the chemical structure of which is shown in Fig. 1. The polymers were kneaded by two mixing rollers for 5 min at a temperature above their glass-transition temperatures and then the additives in their powder form were blended into the kneaded polymers. The kneaded samples were melted for 3 min and were pressed in a laboratory hot press at 160°C, which is above the melting point of additives, for 7 min under a pressure of 180 kg/cm², then they were quenched in an ice-water bath.

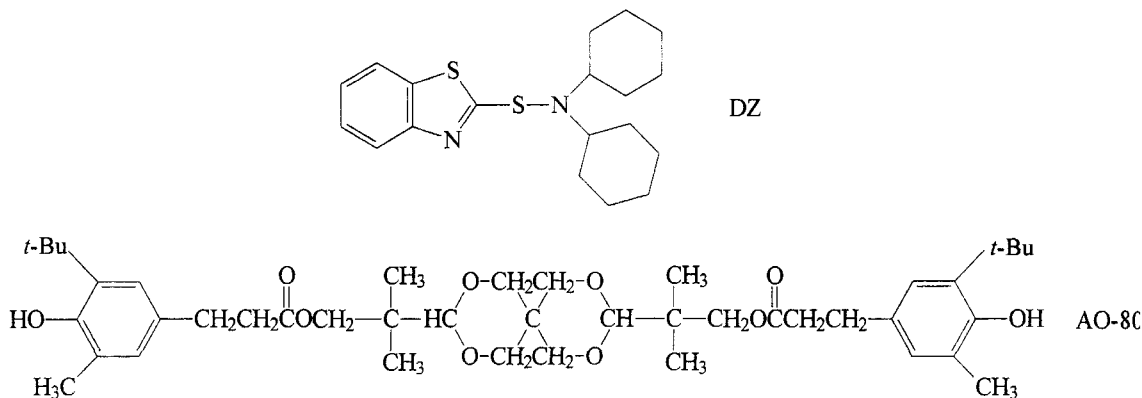


Fig. 1 Chemical structures of organic small molecule substances used

Figure 2 shows the temperature dependence of $\tan\delta$ for various CPE/DZ hybrids. All CPE/DZ hybrids show a single $\tan\delta$ peak, demonstrating that DZ is compatible with CPE. In addition, the peak position of the $\tan\delta$ peak shifts to higher temperatures with increasing DZ content. This behavior is contrary to that of poly(vinyl chloride) plasticized by diethylhexyl succinate^[29]. Hence the DZ molecules are called an antiplasticizer. Moreover, it is noteworthy that the addition of DZ to CPE leads to not only a shift of peak temperature to room temperature, but also a dramatic increase in the maximum value of the $\tan\delta$ peak. In particular, it is of obvious interest that the value of $\tan\delta$ of CPE/DZ (80 wt%) at room temperature is about 5, which is markedly higher than that of common damping materials (< 2). A dramatic increase of the maximum value of $\tan\delta$ peak has also been observed in many hybrids such as ACM/AO-80 (see Fig. 3) and CPP/AO-80. This signifies that the organic hybrids of polymers and small molecules have the potentiality to become a new type of damping materials having very high performance.

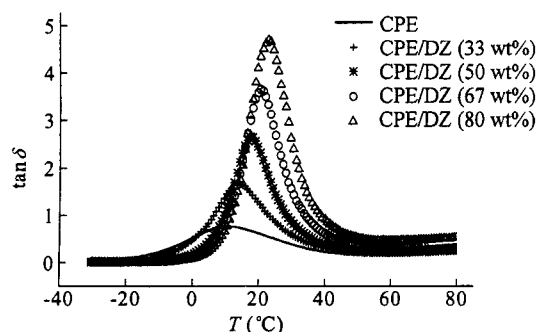


Fig. 2 Temperature dependence of $\tan\delta$ at 11 Hz for CPE/DZ

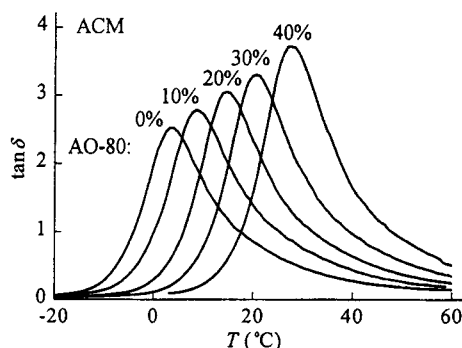


Fig. 3 Temperature dependence of $\tan\delta$ at 11 Hz for ACM/AO-80

The storage modulus E' , loss modulus E'' , and $\tan\delta$ for CPE/AO-80 (40 wt%) were plotted as a function of temperature in Fig. 4. The CPE/AO-80 samples clearly exhibits two transitions. According to recent studies^[25–28], there is a CPE-rich matrix phase and an AO-80-rich domain for CPE/AO-80, and the first transition is attributed to the glass transition of the CPE-rich matrix phase, whereas the second transition is associated with the AO-80-rich domain.

One can define five domains on the E' curve showed in Fig. 4, two dispersion regions in which E' decreases rapidly with temperature, as well as three regions that show a much more gentle decrease in slope, resembling the behavior in the region of a plateau. As can be seen from Fig. 4, the E' in the intermediate plateau is larger than that of pure CPE, whereas the E' in the last plateau is smaller than that of pure CPE. The former implies that the AO-80-rich domains act as the junction of crosslink points. In addition, the Payne effect^[30], that is, the decrease of the E' with increasing strain amplitude and the appearance of the E' maximum at a strain of 0.6%, has been observed for the CPE/AO-80 hybrids^[31]. This result demonstrates that the AO-80-rich domains within matrix polymer CPE form a network. Therefore, CPE/AO-80 can be considered as a thermoplastic IPN. On the other hand, the latter implies that AO-80 molecules at relatively high temperature act as only a plasticizer, because those AO-80 molecules organized by hydrogen bond are released. Therefore, the second dispersion observed in the E' curve is assigned to the dissociation of the intermolecular hydrogen bond between CPE and AO-80 within the AO-80-rich domain. For this reason, the second peak of E'' can be attributed to the dissociation of the intermolecular hydrogen bonds within the AO-80-rich domains, as well as the flow motion of the AO-80 molecules within the AO-80-rich domains and the reptation motion of the CPE chains within the CPE-rich phase accompanied by this dissociation.

For the observed two $\tan\delta$ peaks, the magnitude of the second peak is of particular interest. In several binary systems that have incomplete phase separation, such as a polymeric blend^[32], a block polymer^[33], or a polymer-diluent system^[34], a new transition originating from the cooperative motion of two components has been reported.

Such transition is relatively weak because the contacts of segment-segment or segment-diluent caused by the van der Waals force are very weak. From Fig. 4, another interesting finding is that though the strength of the two transitions is the same for CPE/AO-80 (50%), the height of the second $\tan\delta$ peak originating from the AO-80-rich domain is larger than that of the first $\tan\delta$ peak corresponding to the glass transition of the CPE-rich continuous phase. This implies that the energy dissipation due to dissociation of the intermolecular hydrogen bonds is larger than that due to general friction between polymer chains. From this comparison, it was also confirmed that the second peak is different from the glass transition^[25, 35]. The relatively large transition caused by the dissociation of the intermolecular hydrogen bonds is of importance in many applications such as damping material.

Figure 5 shows the temperature dependence of $\tan\delta$ for CPE/AO-80 samples. The CPE/AO-80 samples clearly exhibit two peaks at 21 °C and 74 °C, respectively. As the content of AO-80 is increased, the height of the first peak shows a gradual decrease after an initial increase. The initial increase is due to some of the AO-80 molecules also being dissolved in the CPE matrix^[25]. In contrast, the height of the second peak increases remarkably by the addition of AO-80. Because of the appearance of the second peak, development of wider applications is conceivable, including the use as a damping material. However, it should be mentioned that for these systems, the values in the middle of the two peaks are all relatively low.

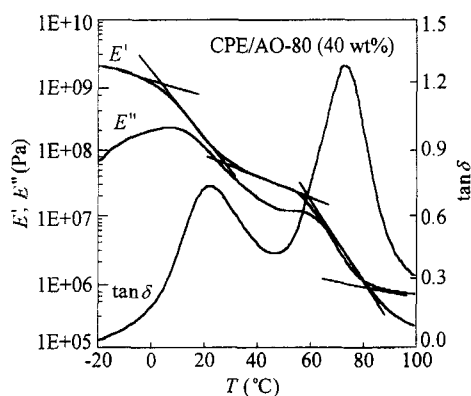


Fig. 4 Temperature dependence of E' , E'' , and $\tan\delta$ at 110 Hz for CPE/AO-80 (50 wt %).

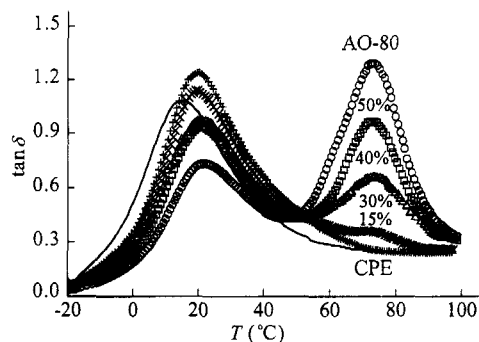


Fig. 5 Temperature dependence of $\tan\delta$ at 110 Hz for CPE/AO-80

MODIFICATION OF DYNAMIC MECHANICAL PROPERTIES BY THIRD COMPOUND^[36, 37]

In order to obtain a very broad, almost rectangular transition range with values for the area under the linear $\tan\delta$ curve, it is important to control the molecular mixing in CPE/AO-80 hybrids. From Fig. 3, we found that the position of the $\tan\delta$ peak of ACM/AO-80 just appears at the intermediate region of the two peaks for CPE/AO-80. In addition, it is also noteworthy that hydrogen bonds exist in organic hybrids consisting of AO-80 and CPE^[26] or ACM^[25]. Thus, when ACM is added to such organic hybrids consisting of CPE and AO-80, and if a linking between the chains of CPE and the chains of ACM through bi-functional AO-80 molecules is achieved, the compatibility of CPE and AO-80 will be improved, which can lead to a broad damping behavior.

The effects of replacing a part of the matrix polymer CPE by ACM on the temperature dependence of $\tan\delta$ are shown in Fig. 6. With an increase in the ACM content, the first peak decreases, whereas the second peak increases. It is possible that these two peaks are to be the same in height at a certain content of ACM (about 88/12). Moreover, it is remarkable that the value in the middle region of the two peaks also increases with an increase in the ACM content. Thus, it can be concluded that the temperature dependence of $\tan\delta$ for CPE/AO-80 can improve by replacing a part of the matrix polymer CPE with ACM, which is compatible with AO-80. In particular, it is noteworthy that as a result the same performance as that of many IPNs can be obtained^[13-21]. Therefore, this is considered to be a good approach to improve the temperature dependence of $\tan\delta$ of the CPE/AO-80 hybrids.

On the other hand, a CPE with two peaks of molecular weight distribution exhibited a relatively high minimum value in two $\tan\delta$ peaks^[27]. This finding suggests that the temperature dependence of $\tan\delta$ could be improved by adding a chemically identical and short chain compound such as CP to a binary system of CPE and AO-80.

The additive effects of CP (the number after CP denotes the chlorination degree) on the temperature dependence of $\tan\delta$ of CPE/AO-80 are shown in Fig. 7. The addition of CP can increase the height of not only the first peak but also the second peak. In addition, the $\tan\delta$ value in the middle of two peaks should be noted, too. For a number of polymers that do not contain diluents, Heijboer^[38] analyzed the relationship between the temperature coefficient of the elastic modulus and the $\tan\delta$ value, finding that the $\tan\delta$ value is proportional to the temperature coefficient of the elastic modulus at a corresponding temperature where there is no transition process. For CPE/CP/AO-80 the $\tan\delta$ value in the middle of two peaks increases in the order CPE < CPE/CP50 < CPE/CP70, as shown in Fig. 7, whereas the slopes of the E' curves at identical temperatures are in the same order, i.e. CPE < CPE/CP50 < CPE/CP70 as shown in Fig. 8, when the AO-80 content is the same. Thus, a relationship between the temperature coefficient of E' and the $\tan\delta$ value was also seen in such multiple-phase system. Moreover, an increase in the E' slope caused by the addition of CP implies that part of the CP molecules are dissolved in AO-80-rich domains in which the CP molecules are mobile.

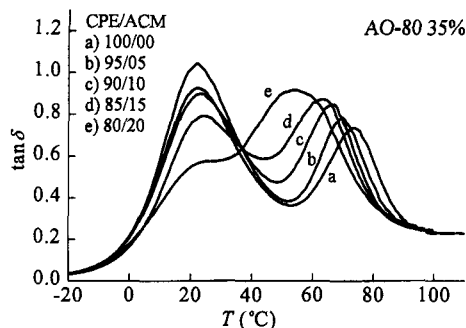


Fig. 6 Temperature dependence of $\tan\delta$ at 110 Hz for CPE/AO-80 and CPE/ACM/AO-80

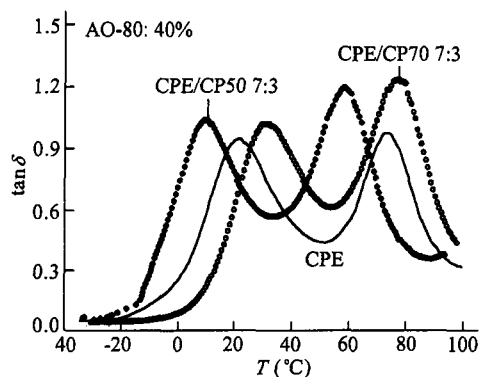


Fig. 7 Temperature dependence of $\tan\delta$ at 110 Hz for CPE/AO-80, CPE/CP50/AO-80 and CPE/CP70/AO-80

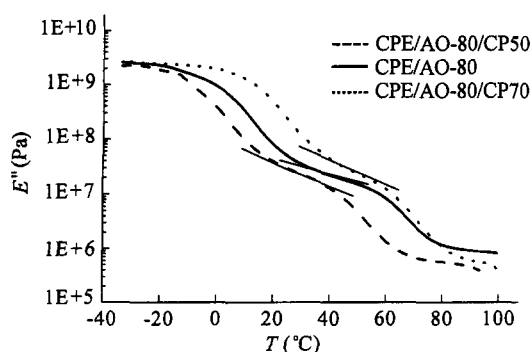


Fig. 8 Temperature dependence of E' at 110 Hz for CPE/AO-80, CPE/CP50/AO-80 and CPE/CP70/AO-80

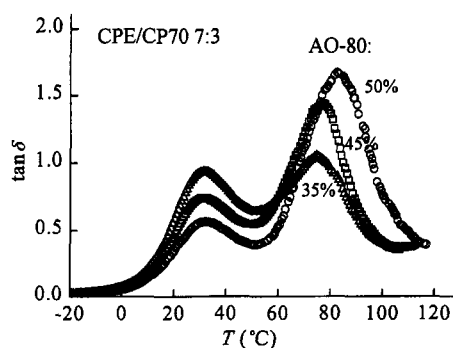


Fig. 9 Temperature dependence of $\tan\delta$ at 110 Hz for CPE/CP70/AO-80

As another comparison, as seen from Fig. 9, the $\tan\delta$ value in the middle of the two peaks decreases with increase of AO-80 when the blend ratio of CPE/CP70 is the same (7:3). In contrast, as seen from Fig. 10, the order of the slopes of the E' curves at identical temperatures agreed with that of the $\tan\delta$ values. The decrease in the E' slope implies that the mobility of the CP molecules within the AO-80-rich domains is suppressed by the

AO-80 molecules.

The plot of $\tan\delta$ value in the middle of the two peaks against the slope ($d\ln E'/dT$) of the E' curve at an identical temperature is illustrated in Fig. 11. This correlation agrees with the relationship reported by Herjboer^[38] for a pure polymer below its glass-transition temperature.

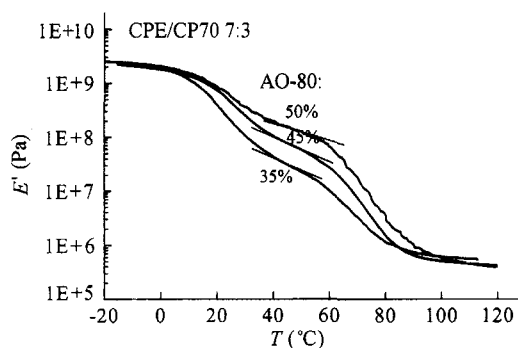


Fig. 10 Temperature dependence of E' at 110 Hz for CPE/CP70/AO-80

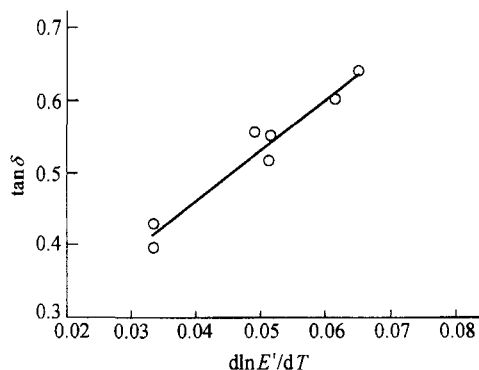


Fig. 11 Height of the $\tan\delta$ peak as a function of the slope ($d\ln E'/dT$) of the E' curve for CPE/CP/AO-80

Furthermore, as seen in Fig. 7, both the first and second transition shift to a higher (CP70) temperature or lower (CP50) temperature with respect to CP. In addition, the extent of the shift also varies according to the kind of CP. In the case of CP50, the shift of the second transition is larger than that of the first transition, indicating that the AO-80-rich domain is preferentially plasticized by the CP molecules. However, in the case of CP70, more molecules are dissolved into a CPE-rich phase.

Finally, it was noted that the $\tan\delta$ value in the middle of the two peaks increases with CP, as shown in Fig. 7. In general, for outdoor or machinery applications, good damping materials should exhibit a high loss factor ($\tan\delta > 0.5$) over a temperature range of at least 60–80°C. As can see in Fig. 7, for CPE/CP70(7:3)/AO-80 (40 wt%), the temperature range in which the $\tan\delta$ value is larger than 0.6 is over 70°C. This behavior is also matched in many IPN systems. Therefore, this is considered to be a new approach to improving the temperature dependence of $\tan\delta$ of CPE/AO-80 hybrids.

PHASE MODIFICATION AND FUNCTION DISCOVERY OF ACM/CPP BLENDS BY AO-80^[25]

ACM and CPP exhibited a relatively high $\tan\delta$ value (> 1.5 , as shown in Fig. 12), their blends therefore might have the possibility of being used for damping materials. However, the $\tan\delta$ curve of their blends has shown limited damping capability as indicated by two narrow peaks (as shown in Fig. 12), because they are immiscible. To obtain a very broad transition range with high value for the area under the $\tan\delta$ curve, it is very important to modify the compatibility of ACM/CPP blends. From Fig. 12, it was found that AO-80 increases the $\tan\delta$ value of ACM and CPP, indicating that AO-80 interacts strongly with both ACM and CPP. Consequently, the addition of AO-80 containing two hydrogen-bonding acceptors to their blends makes it possible to compatibilize the ACM and CPP phases.

The additive effects of AO-80 on the $\tan\delta$ curves of a polymer blend of ACM and CPP (7:3) are shown in Fig. 13. The ACM/CPP blends without AO-80 exhibit two relatively small peaks. The addition of 30% AO-80 enhances the $\tan\delta$ peaks and leads to a shift of ACM peak to higher temperature without affecting the location of CPP peak. These results suggest that the AO-80 molecules may preferentially mix with ACM polymers, because the intermolecular interaction of AO-80 with ACM is greater than that with CPP, as demonstrated in Fig. 12. Furthermore, when the AO-80 content is higher than 30%, these ternary blends show only one peak apparently, implying that ACM and CPP are compatibilized by AO-80, which acts as a new type of compatibilizer. This is due to the formation of a super-molecular network where ACM and CPP chains are coupled by an AO-80 molecule that is a bi-functional hydrogen bonding acceptor, as shown in Fig. 14.

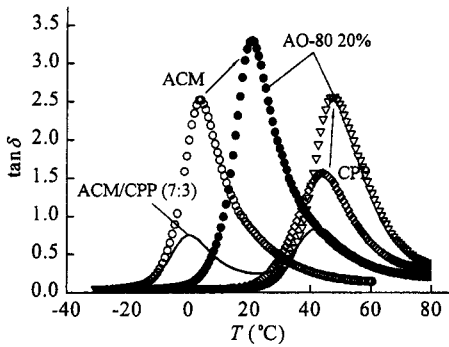


Fig. 12 Temperature dependence of $\tan \delta$ at 11 Hz for ACM, ACM/AO-80 (20 wt %), CPP and CPP/AO-80 (20 wt %).

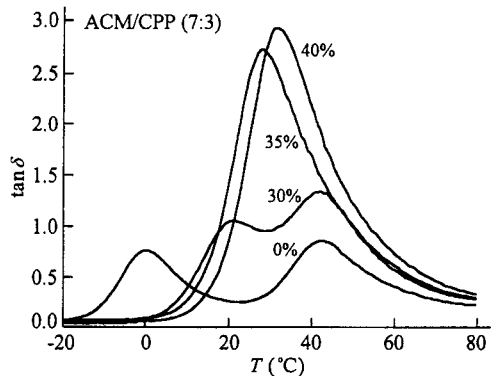


Fig. 13 Temperature dependence of E' at 11 Hz for ACM/CPP (7:3) with AO-80

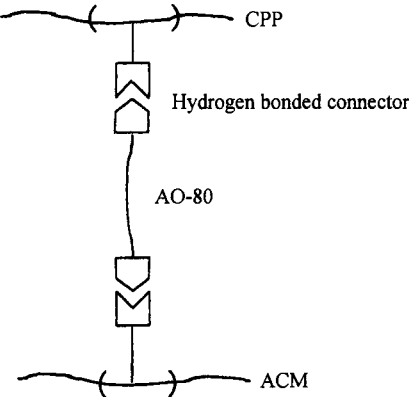


Fig. 14 Schematic structure of supermolecular network at an interface between ACM and CPP

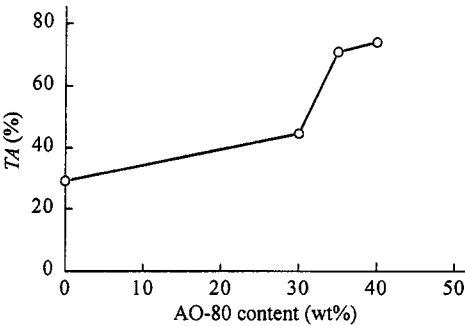


Fig. 15 Dependence of the peak area under the $\tan \delta$ temperature curves of various samples on the AO-80 content

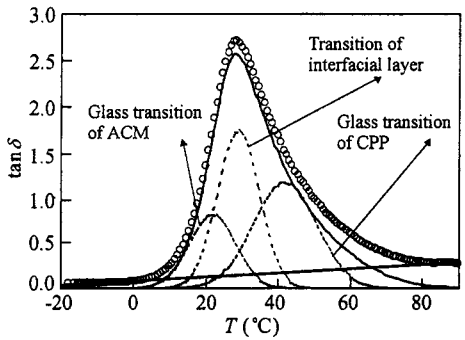
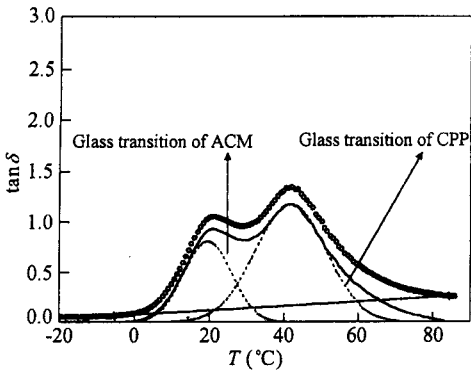


Fig. 16 Normalized $\tan \delta$ -temperature curve of (a) ACM/CPP (7:3) with AO-80 (30%), and (b) ACM/CPP (7:3) with AO-80 (35%)
Gaussian curves are shown using dotted lines.

Thereupon, we shift our attention to the magnitude of the peak area under the $\tan \delta$ -temperature curves, which is abbreviated as $TA^{[9, 39]}$. The additive effects of AO-80 on the TA values of ACM/CPP are summarized in

Fig. 15. A sharp increase of $T\alpha$ value was observed around 30%–35% AO-80 content. This may be due to the appearance of a new $\tan\delta$ peak between the glass transitions of ACM and CPP, and it can be attributed to the formation of an interfacial layer between ACM and CPP.

As is evidenced in Fig. 13, the shape of $\tan\delta$ versus temperature curves is drastically changed at around 30%–35%. A blend of 30% AO-80 distinctly shows two peaks, whereas that of 35% AO-80 seems to have a broad peak with multi-components. In order to analyze the $\tan\delta$ peaks quantitatively, we performed a mathematical deconvolution process, in which the Gaussian curves are used. As it can be seen in Fig. 16, one relatively larger peak appears between two peaks corresponding to the glass transition of ACM and of CPP for the ACM/PP (7:3) with 35% AO-80. This peak is probably associated with the interfacial layer between ACM and CPP. Considering that this novel peak is detectable only from the DMA but not from the DSC curve, this novel peak could be attributed to the cooperative motion between the AO-80 molecules and the ACM chains as well as the CPP chains. The existence of an interfacial layer between ACM and CPP when the AO-80 content is higher than 35% has been demonstrated by scanning electron microscopy observation of those samples^[25].

We also found that the addition of AO-80 could impart highly desirable self-adhesiveness to ACM/PP blends. Therefore, such a ternary blend can be regarded as a new type of super-damping material for use in damping of low vibration and sound waves.

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