

Note

EFFECTS OF PHENOL RESIN ADDITIVE ON DYNAMIC MECHANICAL PROPERTIES OF ACRYLATE RUBBER AND ITS BLENDS*

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Abstract The dynamic mechanical properties of a new blend system consisting of phenol resin and polar polymer (acrylate rubber and/or chlorinated polypropylene) were investigated. It was found that the addition of phenol resin to acrylate rubber and its incompatible blend can cause a remarkable improvement in the temperature dependence of the loss tangent. As a result, the present blends are very good damping materials.

Keywords Acrylate rubber, Chlorinated polypropylene, Phenol resin, Damping

INTRODUCTION

Polymers are useful vibration damping materials near their glass-transition temperature where the molecular motion of chain segments dissipates mechanical energy as heat. Homopolymers generally have narrow glass-transition temperature regions (20–30 °C)^[1]. When temperature becomes a significant variable, the narrow temperature range of the glass transition causes the impairment of most homopolymer damping materials^[2]. Therefore, polymer systems with a high and broad damping range are required. The glass transition of a given homopolymer at a given frequency can be controlled by many conventional means, such as the addition of plasticizers and fillers^[3]. Another method is through the use of multi-component systems such as copolymers^[4], mechanical blends^[5] and interpenetrating polymer networks (IPNs)^[6].

Recently, it has been found that organic hybrids consisting of polar polymers such as chlorinated polyethylene (CPE) and functional organic small molecule substance 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) show a novel transition above the glass-transition temperature of matrix polymer^[7]. This fact will provide a new concept for developing damping material. However, the minimum value between two $\tan\delta$ peaks is lower. To obtain a very broad, almost rectangular, transition range with high values for the area under the linear $\tan\delta$ curve, it is important to control the molecular mixing in organic hybrids. When chlorinated paraffin (CP)^[8] or acrylate rubber (ACM)^[9] is deliberately added to CPE/AO-80, a relatively broad $\tan\delta$ curve has been obtained. In another recent study^[10], we also investigated the effects of the addition of AO-80 on the dynamic mechanical properties of a blend of ACM and chlorinated polypropylene (CPP). It has been found that a ternary blend of ACM/CPP (7:3) with AO-80 (35 wt%) showed a novel $\tan\delta$ peak between two peaks corresponding to ACM and CPP. This peak is attributed to an interfacial layer where a super-molecular network is formed.

In this study, we considered a new blend system consisting of phenol resin (PF) and polar polymer (ACM

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and/or CPP) based on a concept of the sophisticated use of hydrogen bonds. The effects of PF additive on the dynamic mechanical properties were investigated. The final goal is to obtain a cheap good damping material.

EXPERIMENTAL

The ACM used in this study was a commercial product (Nippon Zeon Co., Nipol AR42W); CPP with 29.5% of the weight content of chloride was a thermoplastic resin (Nippon Paper Industries Co., Superchlon 803MWS). The PF was a thermoplastic resin used as paint or adhesive agent (Hitanol 1401; Hitachi Chemical Co., Ltd.), with a glass transition temperature of 85 °C. To control the glass-transition temperature of base polymer, triphenylphosphate (TPP) was used as the plasticizer of PF. The ACM or/and CPP were kneaded by mixing rollers for 5 min at 90 °C and then PF and TPP were added to the kneaded ACM and CPP, and their mixture. The kneaded samples were molten for 3 min and then were pressed in a laboratory hot press at 160 °C for 7 min under a pressure of 20 MPa. Finally, the samples were cooled to obtain films by quenching in an ice-water bath.

The dynamic mechanical measurements were performed using a dynamic mechanical analyzer (Rheology Co., DVE-V4) on sample specimens of the following dimensions: 20 mm long, 5 mm wide, and 1 mm thick. The temperature dependence of dynamic tensile moduli was measured at a constant frequency of 11 Hz and a heating rate of 3 °C/min.

RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the loss tangent ($\tan\delta$) for ACM and ACM/PF (50/50). ACM shows a very high $\tan\delta$ peak, implying that ACM has a good damping property. However, it should be mentioned that the effective damping range of ACM is relatively narrow. Therefore, in order to obtain a broader damping property, a modification of ACM is needed. As shown in the figure, ACM/PF blend shows a single $\tan\delta$ peak, implying that ACM is compatible with PF. We noticed that the addition of PF to ACM leads to an improvement in the temperature dependence of $\tan\delta$. In general, for outdoor or machinery applications, a good damping material should exhibit a high loss factor ($\tan\delta > 0.5$) over a temperature range of at least 60–80 °C^[11]. The ACM/PF (50/50) blend showed not only relatively high $\tan\delta$ peak (2.3), but also its temperature range of $\tan\delta > 0.5$ is 76 °C, resulting in that ACM/PF mixture is a promising damping-material.

In general, the $\tan\delta$ peak of a compatible polymer blend is low and broad comparing with each component. In the case of ACM/PF blend, its $\tan\delta$ peak is broader, but its height hardly changes. According to our previous studies^[8, 12, 13], in the case of CPE/AO-80 that showed two transitions, though the strength of the two transitions is the same for CPE/AO-80 (50/50), but the height of the second $\tan\delta$ peak originating from the dissociation of the intermolecular hydrogen bonds in the AO-80-rich domain is larger than that of the first $\tan\delta$ peak due to the glass transition of the CPE-rich continuous phase. This implies that the energy dissipation due to the dissociation of the intermolecular hydrogen bonds is larger than that due to general friction between polymer chains. Therefore, the good damping properties of ACM/PF blends can be attributed to the formation of the intermolecular hydrogen bonds between ACM and PF.

On the other hand, it is well known that the phase diagram of a given polymer blend is determined by the molecular weight of their components. In the case of ACM/PF that exhibited the upper critical solution temperature form (UCST), the temperature dependence of $\tan\delta$ can be improved by increasing the molecular weight of PF component or/and cross-linking of ACM component.

Figure 2 shows the temperature dependence of $\tan\delta$ for ACM, CPP and their blend. Both ACM and CPP exhibited a relatively high $\tan\delta$ value (> 2.3). The peak area under the $\tan\delta$ temperature curves, which is abbreviated as TA, can be considered to be a damping index, which is a measure of the energy dissipation of a transition^[6]. Both ACM and CPP showed relatively large TA value (52.9 and 43.4 °C), therefore they can be considered to be a good damping homopolymer. If the TA value of ACM/ CPP blend obeys the simple additive mixing rule reported by Ogawa *et al.*^[11] for a compatible copolymer, their blends will have a possibility be used as damping materials. However, the $\tan\delta$ curve of ACM/ CPP (75/25) has shown a limited damping capability as

indicated by two narrow peaks (its TA value is only 29.8°C). This is because the blends of ACM and CPP are immiscible.

In order to obtain a very broad transition range with high value for the area under the $\tan\delta$ curve, it is of great importance to modify the compatibility of ACM/CPP blends. In a previous study^[10], AO-80 was found to be a good compatibilizer for the ACM/CPP blends. PF having similar hydroxyl group also is conceivable as a compatibilizer of the ACM/CPP blends. The additive effects of PF and TPP on the temperature dependence of $\tan\delta$ of a polymer blend of ACM and CPP (75:25) are shown in Fig. 3. As shown in the figure, ACM/CPP (75/25) blend exhibits two relatively small peaks. The addition of 20 phr PF and 12 phr TPP apparently raise the $\tan\delta$ peak of CPP. PF resin enhanced the value of the $\tan\delta$ peak of ACM and CPP, indicating that PF interacts strongly with both ACM and CPP. Consequently, addition of such PF containing hydrogen-bonding acceptors into their blends makes it possible to compatibilize ACM and CPP phase. Moreover, The addition of 40 phr PF and 24 phr TPP leads to a larger temperature shift of ACM peak but did not affect the position of CPP peak. This result suggests that the PF component may preferentially mix with ACM polymers.

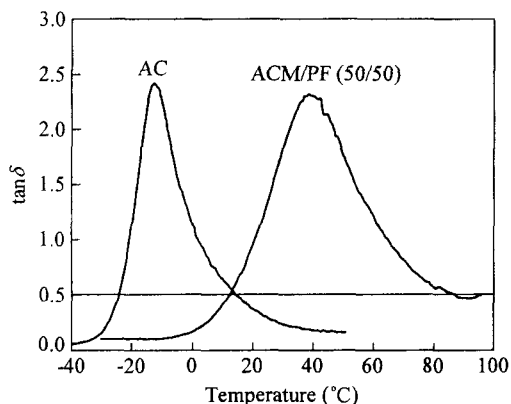


Fig. 1 Temperature dependence of $\tan\delta$ at 11 Hz for ACM and ACM/PF (50/50)

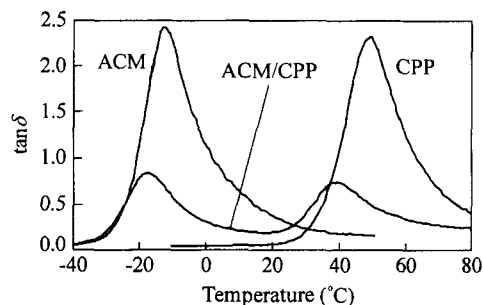


Fig. 2 Temperature dependence of $\tan\delta$ at 11 Hz for ACM, CPP and their blend

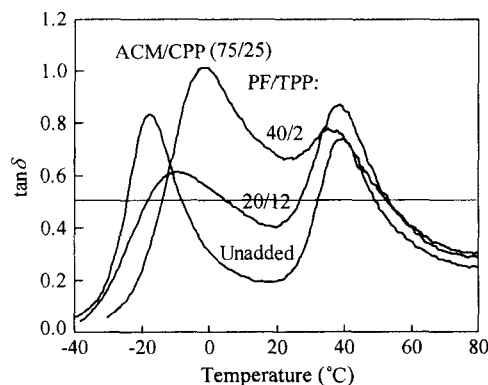


Fig. 3 Temperature dependence of $\tan\delta$ at 11 Hz for ACM/CPP (75/25) blend with PF and TPP

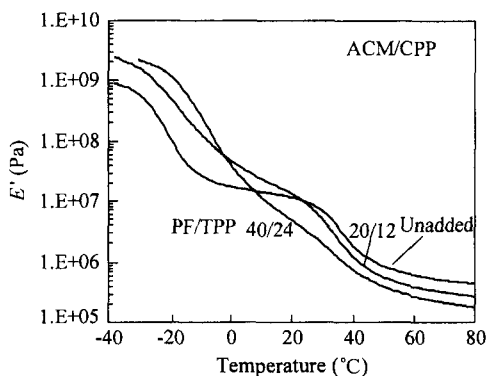


Fig. 4 Temperature dependence of E' at 11 Hz for ACM/CPP (75/25) blend with PF and TPP

It is noteworthy that for ACM/CPP/PF/TPP (75/25/40/24) blend, the temperature range of $\tan\delta > 0.5$ is 67°C. A comparison with many IPNs^[15–17] having two overlapping peaks shows that this system exhibits top-ranking TA value. Thus, the present blends make it as a very good damping materials.

Figure 4 shows the temperature dependence of storage modulus E' . The E' curves of ACM/CPP blends have

two distinct dispersions around -20 and 40°C , while that of the blend with 40 phr PF shows a single dispersion. This behavior is similar to the temperature dependence of E' of many IPNs.

CONCLUSIONS

The dynamic mechanical properties of the multi-component systems of ACM and its blend with PF were investigated. The addition of PF to ACM and its incompatible blend can cause a remarkable improvement in the temperature dependence of $\tan\delta$. As a result, the present blends are very good damping materials.

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