

Damping Properties of Ethylene-Vinyl Acetate Rubber/Polylactic Acid Blends

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Abstract

In this research, ethylene-vinyl acetate rubber (EVM)/polylactic acid (PLA) = 80/20 by weight blend was compounded with silica in a Haake torque rheometer. The effects of hindered phenol (AO-60), super branched polyol, petroleum resin C9, polyvinyl chloride (PVC) and acrylic rubber (ACM) on the damping properties of blends were investigated by dynamic mechanic analyzer (DMA). The results showed that 20 phr super branched polyol significantly increased the damping factor of PLA to widen the effective damping temperature range from 42.1°C to 102.5°C. 15 phr AO-60 and 10 phr petroleum resin C9 both dramatically raised the blend's damping factor to broaden the effective damping temperature range to 98.0°C and 102.6°C, respectively. ACM and PVC are compatible with EVM, and both improved the damping properties of EVM/PLA blends.

Keywords

DMA, EVM/PLA, Damping, Organic Hybrid

1. Introduction

Polymeric damping materials are functional materials having polymer matrices and are widely used for vibration and noise reduction [1]-[3]. The height and width of the $\tan\delta$ peak at the glass transition zone are the two main factors for assessing the damping ability of a material. In general, homopolymers possess efficient damping ability in a temperature range of only 20°C - 30°C around the glass-transition temperature T_g [4]. However, for outdoor or machinery applications, good damping materials should exhibit a high loss factor ($\tan\delta \geq 0.3$) over a wide temperature range [5]-[7]. In order to meet this particular requirement, typically, a low T_g and a high T_g polymer are combined together by blending (including solution and mechanical blending) or as block or graft copolymers, as interpenetrating networks, etc. [8]. Organic hybrid polymers are also an effective way to improve

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the damping properties of polymers [9]-[13].

Ethylene-vinyl acetate rubber (VA content of more than 40%, EVM) [14] [15] is a polar saturated rubber with a large number of polar ester side groups, which impart a high loss factor (ca. 0.93). The glass transition zone of EVM is between -5°C and 30°C , which is a typical temperature range for damping applications. EVM is also inherent flame retardant making it an ideal material for damping elements. However, the effective damping temperature range (EDTR) is too narrow to make it suitable for many applications. As a result, it is not widely used. Polylactic acid (PLA) [16]-[18] is a biodegradable and environment-friendly polymer with abundant ester groups along the main chain. PLA has a high damping factor ($\tan\delta = 2.2$), and its glass transition lies between 55°C and 70°C . Adding PLA can widen the damping temperature range of EVM, and it is an effective way to design a damping material with abroad temperature range [19]-[21].

In this work, EVM/PLA = 80/20 was chosen as blend matrix having abundant ester groups (hydrogen bond accepters). Hindered phenol AO-60, super branched polyol (made in our laboratories) and petroleum resin C9 with molecular weights of 1177, 1101 and 2000 were chosen as the organic hybrid agents for EVM/PLA blends due to their mode rate molecular weight as well as the abundant hydroxyl groups (in AO-60 and the polyol), which could act as hydrogen bond donors. PVC and ACM both with high damping factors were used as a third polymer component in the EVM/PLA blends to examine the effects on the damping properties. The aim was to prepare high performance damping materials with wider damping temperature ranges.

2. Experimental

2.1. Materials

Ethylene-vinyl acetate rubber (EVM): Levapren 700, vinyl acetate content of 70 wt%, Lanxess Deutschland GmbH, Leverkusen, Germany; polylactic acid (PLA): 2003D, Nature Works, USA; dicumyl peroxide (DCP): Rhenocure VC-40CC, 40% wt on an inert carrier; triallylisocyanurate (TAIC): Rhenofit TAIC/s; polycarbodiimide (PCD): Stabaxol P; DCP, TAIC and PCD were all provided by Rhein Chemie Qingdao, China; silica 1165 MP: specific surface area $165\text{ m}^2/\text{g}$, Rhodia, Qingdao, China; AO-60 (antioxidant 1010, Hindered phenol): Haihua Qingdao, China; super branched polyol was made in our laboratory (as shown in **Figure 1**); petroleum resin C9: Puyang Chemical Co. Ltd., Henan, China; ACM: East Asia, Japan; PVC: Zhongtai, Xinjiang, China.

2.2. Blend Recipe

The variable elements of the blend recipe include: EVM 80 phr, PLA 20 phr, PCD 2 phr, SiO_2 30 phr, DCP 1.5 phr, TAIC 0.5 phr; AO-60, polyol, C9, ACM and PVC.

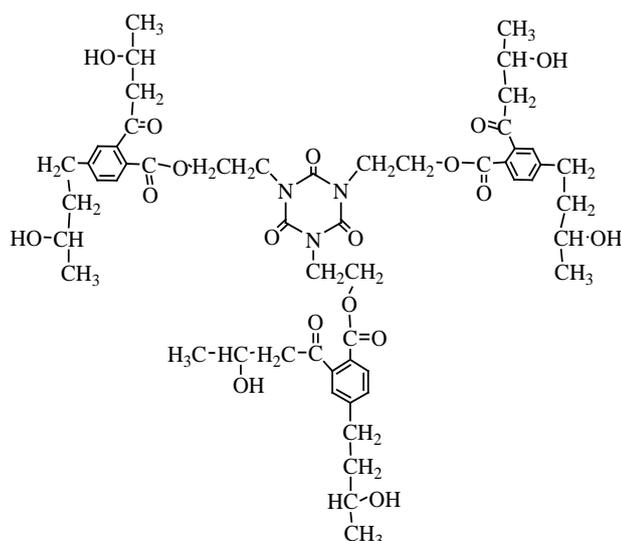


Figure 1. Structure of the super branched polyol.

2.3. Sample Preparation

EVM and PLA were first mixed in a Haake Rheomix 30,000 s mixer at a rotor speed of 50 rpm for 5 min at 160°C. Then, premixed filler and additive were added and mixed for 7 more minutes. The blends were taken out of the mixer and processed at room temperature with DCP and TAIC on an SK-160B two-roll mill manufactured by Shanghai Plastics and Rubber Machinery Factory, China, before finally being molded into sheets in a VC-150T-FTMO-3RT vacuum press manufactured by Jiaxin Electric Company, China, at 170°C for 10 min.

2.4. Measurements

The dynamic mechanical analysis was carried out using a Netzsch DMA 242 Dynamic Mechanical Analyzer, manufactured by Netzsch Company, Germany, from -60°C to 180°C at a rate of 3 K/min and a fixed frequency of 1 Hz in a double cantilever deformation mode.

3. Result and Discussion

3.1. The Effects of AO-60 on the Damping Properties of EVM/PLA Blends

Figure 2 shows the $\text{Tan}\delta$ -T curve of blends with 15 phr of AO-60. Some key data from Figure 1 are summarized in Table 1. It can be seen that AO-60 significantly increased the blend's $\text{tan}\delta$ and the effective damping temperature range (EDTR) of EVM/PLA was broadened from 42.1°C to 98.0°C. The two glass transition temperatures (peak1 and peak2) were shifted closer together. It was concluded that AO-60 has strong hydrogen bond interaction with both polymers in the EVM/PLA blend [9] [10]. During the dynamic mechanical testing, hydrogen bonds dissociate and reform, consuming a lot of energy. Thus, the loss factor increases and the damping properties improve. In addition, during heating, AO-60 molecules move at lower temperature and more easily due to their lower molecular weight (1177) compared with the polymer molecules. This leads to an increase intermolecular friction and more effective damping [10].

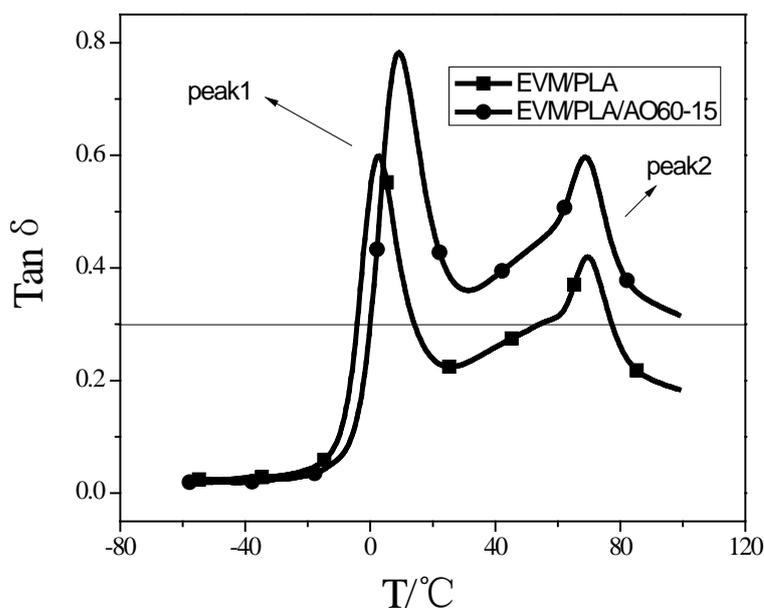


Figure 2. Effects of AO-60 on the damping factor of EVM/PLA blends.

Table 1. Effects of AO-60 on the damping factor of blends.

Composites	Peak1/°C	$\text{Tan}\delta_{\max}$ value 1	Peak2/°C	$\text{Tan}\delta_{\max}$ value 2	EDTR/°C ($\text{Tan}\delta \geq 0.3$)
EVM/PLA	2.5	0.594	69.7	0.418	42.1
EVM/PLA/AO60-15	9.7	0.779	69.2	0.596	98.0

It also can be seen that the difference between T_g of EVM and T_g of PLA (peak1-peak2) became smaller indicating that AO-60 acts as a compatibilizer for EVM and PLA.

3.2. The Effects of Polyol on the Damping Properties of EVM/PLA Blends

Figure 3 shows the Tan δ -T curve of blends with 20 phr of polyol. Some key data from **Figure 3** are summarized in **Table 2**. It can be seen that the polyol dramatically increased the damping factor of PLA and broadened the effective damping temperature range of EVM/PLA blends from 42.1°C to 102.5°C. The T_g of PLA shifted a little to lower temperature but that of EVM remained unchanged. We infer that the polyol has stronger hydrogen bond interaction with PLA than with EVM [22] and the improvement of the damping properties of the blend can be essentially attributed to the intermolecular friction between the polyol and the polymer matrix due to the polyol's inherent dynamic mechanical behavior [23].

3.3. The Effects of Petroleum Resin C9 on the Damping Properties of EVM/PLA Blends

Figure 4 shows the Tan δ -T curve of blends with 10 phr amounts of C9. Some key data from **Figure 5** are summarized in **Table 3**. It can be seen that 10 phr C9 significantly increased the blend's damping factor and widened the effective damping temperature range from 42.1°C to 102.6°C. Both T_g shifted to higher temperature. It is known that there are no hydrogen bond donors in C9. Thus, the improvement to the damping properties of the blend with C9 must derive from its own dynamic mechanical behavior due to its moderate molecular weight, which is comparable with that of AO-60 [24].

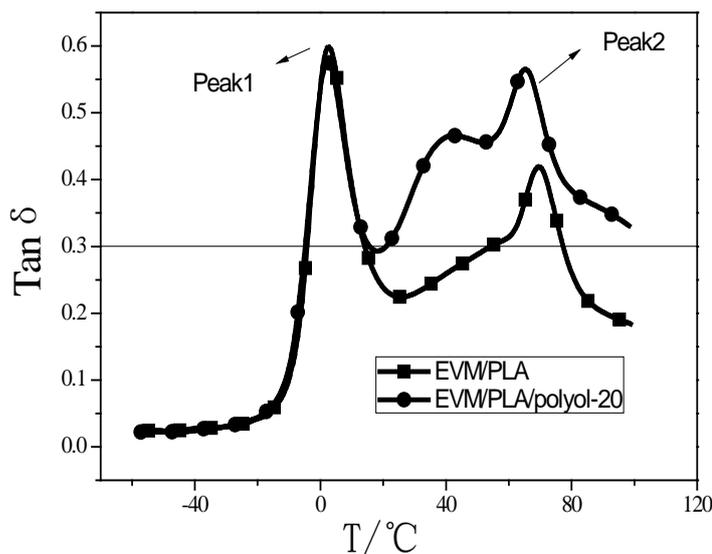


Figure 3. Effects of polyol on the damping factor of EVM/PLA blends.

Table 2. Effects of polyol on the damping factor of blends.

Composites	Peak1/°C	Tan δ_{\max} value 1	Peak2/°C	Tan δ_{\max} value 2	EDTR/°C (Tan $\delta \geq 0.3$)
EVM/PLA	2.5	0.594	69.7	0.418	42.1
EVM/PLA/polyol-20	2.8	0.575	64.8	0.564	102.5

Table 3. Effects of C9 on the damping factor of blends.

Composites	Peak1/°C	Tan δ_{\max} value 1	Peak2/°C	Tan δ_{\max} value 2	EDTR/°C (Tan $\delta \geq 0.3$)
EVM/PLA	2.5	0.594	69.7	0.418	42.1
EVM/PLA/C9-10	6.9	0.699	72.4	0.533	102.6

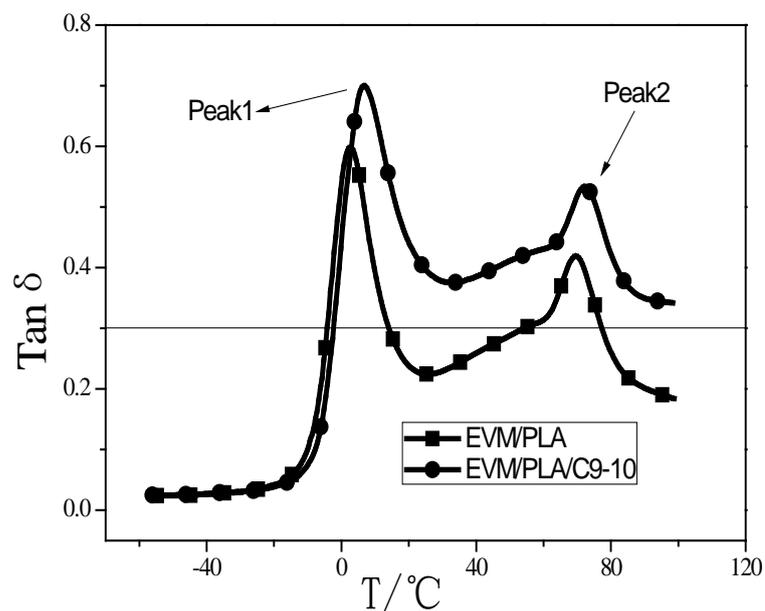


Figure 4. Effects of C9 on the damping factor of EVM/PLA blends.

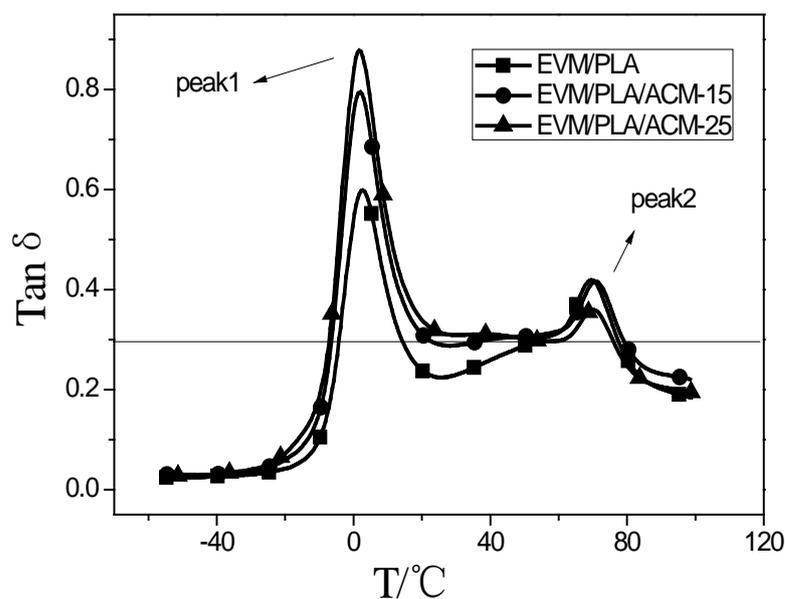


Figure 5. Effects of ACM on the damping factor of EVM/PLA blends.

3.4. The Effects of ACM on the Damping Properties of EVM/PLA Blends

The $\text{Tan}\delta$ - T curves of blends with different amounts of ACM are shown in Figure 5. Some key data from Figure 5 are summarized in Table 4. It can be seen that 15 phr ACM broadened the effective damping temperature range from 42.1°C to 57.7°C due to increasing the breadth of EVM's damping peak. 25 phr ACM further widened the effective damping temperature range of blend to 81.7°C due to further increasing the breadth EVM's damping peak. Moreover, the T_g of EVM shifted to slightly lower temperature with the addition of ACM.

The glass transition temperature of ACM is about -15°C and ACM has similar molecular structure to EVM, they are both polar. Only a single glass transition temperature could be observed around Peak1. Furthermore, peak 1 was increased dramatically in area with increasing ACM content. All this suggests that ACM and EVM are compatible.

3.5. The Effects of PVC on the Damping Properties of EVM/PLA Blends

The $\text{Tan}\delta$ -T curves of blends with different amounts of PVC are shown in **Figure 6**. Some key data from **Figure 6** are summarized in **Table 5**. It can be seen that 15 phr PVC increased the blend's damping factor and broadened the effective damping temperature range from 42.1°C to 76.9°C. The Tg of EVM was shifted to higher temperature. 25 phr PVC shifted the Tg of EVM to even higher temperature but did not further increase the damping value.

The glass transition temperature of PVC is at ca. 78°C, from previous studies [10] it is known that PVC exhibits good compatibility with EVM700. Thus, no new relaxation peak between peak1 and peak2 is observed in **Figure 6** and the EVM Tg peak simply shifts to higher temperature. The Tg peak of PLA didn't move, which can be taken as evidence that PVC and PLA are not compatible.

4. Conclusion

15 phr AO-60 significantly increased the damping factor of an EVM/PLA blend to broaden the effective damping temperature range from 42.1°C - 98°C. The 20 phr super branched polyol was able to significantly raise PLA's damping factor without shifting the Tg of the polymers but nevertheless broadening the effective damp-

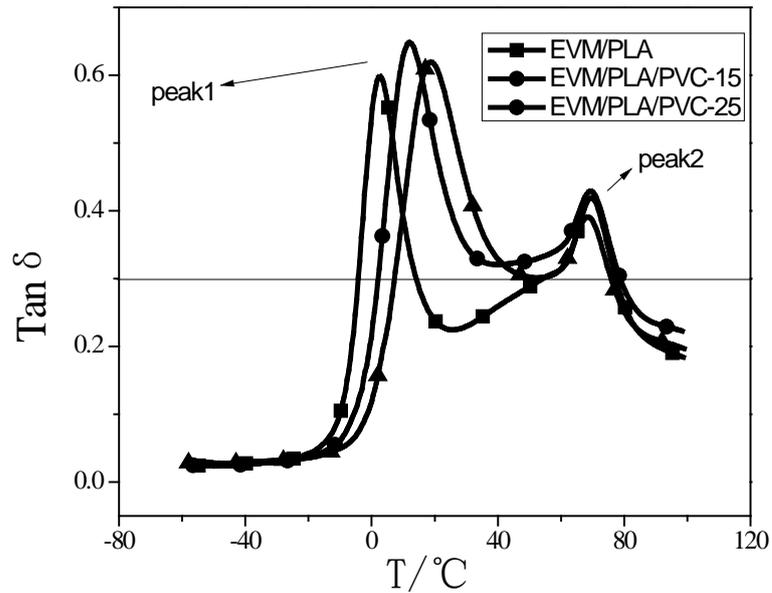


Figure 6. Effects of PVC on the damping factor of EVM/PLA blends.

Table 4. Effects of ACM on the damping factor of blends.

Composites	Peak1/°C	Tanδ _{max} value 1	Peak2/°C	Tanδ _{max} value 2	EDTR/°C (Tanδ ≥ 0.3)
EVM/PLA	2.5	0.594	69.7	0.418	42.1
EVM/PLA/ACM-15	1.5	0.792	69.7	0.419	57.7
EVM/PLA/ACM-25	1.7	0.878	70.2	0.359	81.7

Table 5. Effects of PVC on damping factor of blends.

Composites	Peak1/°C	Tanδ _{max} value 1	Peak2/°C	Tanδ _{max} value 2	EDTR/°C (Tanδ ≥ 0.3)
EVM/PLA	2.5	0.594	69.7	0.418	42.1
EVM/PLA/PVC-15	11.6	0.651	69.7	0.433	76.9
EVM/PLA/PVC-25	19.0	0.616	68.6	0.391	68.3

ing temperature range to 102.5°C. 10 phr petroleum resin C9 significantly increased the damping factor of EVM/PLA blends to broaden the effective damping temperature range from 42.1°C to 102.6°C. 25 phr ACM increased the damping factor of EVM to broaden the effective damping temperature range to 81.7°C; 15 phr PVC increased the damping factors of EVM and PLA to widen the effective damping temperature range to 76.9°C and shifted the T_g of EVM to higher temperature. ACM and PVC are both compatible with EVM700.

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