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Effects of Fillers and Other LIPN on Damping of PS/P(EA-nBA) LIPNs

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Polystyrene/Poly(Ethyl acrylate-n-Butyl acrylate) (PS/P(EA-nBA) latex interpenetrating polymer networks (LIPNs) were synthesized by a two stage emulsion polymerisation technique. The effects of fillers and PMMA/P(EA-EMA) LIPN on damping of the LIPNs have been investigated. The results indicate that inorganic fillers produce increases in $\tan \delta$ values and broaden the damping temperature range. Among the fillers, mica has shown good result on damping properties. Blending of PS/(EA-nBA) LIPN with PMMA/P(EA-EMA) LIPN produces similar results as the inorganic fillers. Damping materials whose $\tan \delta$ values exceed 0.5 over 82°C temperature range between -21.3 and 60.7 has been obtained.

KEY WORDS Latex, interpenetrating polymer network (IPN), dynamic mechanical properties, damping, polystyrene/poly(ethyl acrylate-n-butyl acrylate).

INTRODUCTION

Noise is a very serious form of pollution. It can be reduced by controlling the damping characteristics of the material. Many kinds of damping materials are used in controlling the sound levels. The principle is that the application of the damping materials to the vibrating surface will convert the mechanical energy into heat which is dissipated within the damping materials rather than being radiated to the air. Among these materials, Interpenetrating Polymer Networks (IPNs), which may be defined as a combination of two polymer in network form,¹ are an important class of vibration damping materials because of their broad glass transition.^{2,3,4} The most common type of polymeric materials such as simple homopolymers or copolymers and their blends are, however, ineffective for reduction of noise because of their limited damping capability indicated by one or two narrow dynamic mechanical loss peaks.^{5,6,7} Therefore there has been an increasing interest in exploring the potential of IPNs for sound and vibrating damping in recent years. We synthesized PS/P(EA-nBA) LIPNs and to further improve the damping properties of the LIPNs, the effects of fillers and other LIPN on damping of PS/P(EA-nBA) were investigated.

EXPERIMENTAL

Materials

Styrene (S) was supplied by BeiJing 50950 chemical plant; Ethyl acrylate (EA), n-Butyl acrylate (nBA), Methyl methacrylate (MMA), Ethyl methacrylate (EMA), by TianJin third chemical reagent plant; Sodium lauryl sulfate (SLS), by TianJin Dongfang chemical plant; Fillers (TiO_2 , Mica, CaCO_3 , Talc powder), by TianJin chemical reagent market; Trimethylpropane triacrylate (TMPTA), by TianJin Institute of Chemical Reagent. Before use, the monomers were distilled under reduced pressure.

Syntheses

Latex IPNs were synthesized by a two stage emulsion polymerisation technique. First, the deionized water and emulsifiers (SLS) were introduced into a flask equipped with stirrer and reflex condenser at 60°C , then the monomer I containing 1% TMPTA and three-fifths of the initiator (0.2% $\text{K}_2\text{S}_2\text{O}_8$) were gradually added by a dropping funnel. After that, the reagents were heated to 80°C and the rest of initiator added. After 2 hours at 80°C , the contents were cooled to room temperature. This produced the starting latex (Network I) for the second stage.⁷ Next, the monomer II containing 1% TMPTA and three-fifths of initiator (0.2% $\text{K}_2\text{S}_2\text{O}_8$) were added as in the first stage at 60°C , but with no new emulsifier. The network I was fully swollen with the monomer II. The reaction was completed under the same condition as in the first stage, and the resulting product was a LIPN. Finally, various amounts of fillers or PMMA/P(EA-EMA) LIPNs were incorporated into the PS/P(EA-nBA) LIPNs.

Measurement of Damping Properties

Rheovibron DDV-III-EA dynamic viscoelastomer (TOYO Baldwin Co. Ltd) was used to obtain the damping properties of the system. The heating rate was $2^\circ\text{C}/\text{min}$. and the frequency was 35 Hz.

RESULTS AND DISCUSSION

Effect of Various Fillers

Ten wt percent inorganic fillers such as TiO_2 , Mica, CaCO_3 , Talc powder, were incorporated into 30/70 PS/P(EA-nBA) LIPN with an EA/nBA ratio of 50:50. The dynamic mechanical properties data are presented in Figure 1 and Table I. From the data, it can be seen that the damping values of filled LIPN markedly increased in a broad temperature range over values found with the unfilled LIPNs. $\tan \delta_{\text{max}}$ in Table I is damping peak height at $\tan \delta$ - T curve, these enhance in different degrees for different filler, the order of fillers to increase the damping values of PS/P(EA-nBA) LIPN is mica > TiO_2 > CaCO_3 > Talc powder. Table I shows that for the unfilled sample, the $\tan \delta$ values exceed 0.4 over a temperature range of

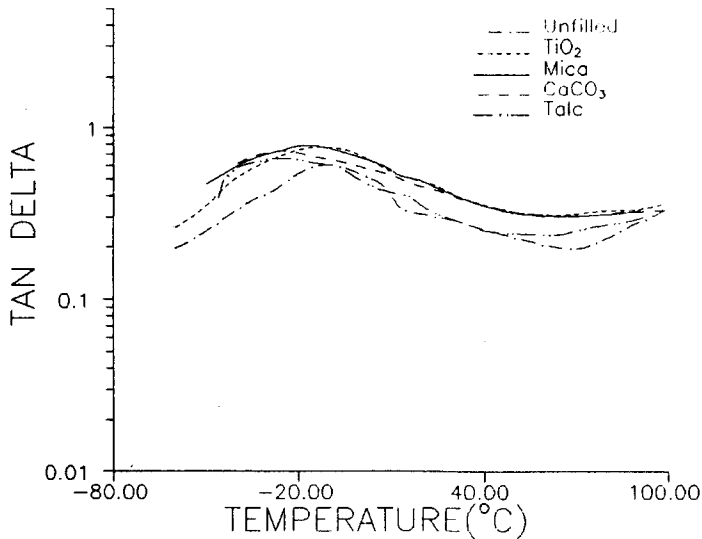


FIGURE 1 Effect of various fillers on the damping of PS/P(EA-nBA) LIPNs.

TABLE I

The damping properties of filled LIPNs

Sample	tan δ max peak height	Rate of tan δ max increase (%)	Temperature range of tan $\delta > 0.4$ (°C)		
			T1	T2	ΔT
Unfilled	0.607	—	-33.2	14.7	47.9
TiO ₂	0.774	27.5	-45	33	78
Mica	0.793	30.6	-55	32.9	87.9
CaCO ₃	0.736	21.3	-51	29.3	80.3
Talc powder	0.661	8.3	-46	19	65

47.9°C. This range is 78°C for TiO₂-filled sample, 87.9°C for mica-filled sample; and 65°C for Talc powder-filled sample. Thus the order of the fillers to broaden the damping range is Mica > CaCO₃ > TiO₂ > Talc powder. This is in agreement with the data of Silverstein *et al.*⁸ The large increase in damping values for filled LIPN indicate that mechanisms other than friction between particles or between particles and polymer contribute to energy dissipation.

In mica-filled LIPN, the mica platelet at one side of the LIPN damping layer can be considered as a vibrating substrate, while the platelet at the other side of damping layer acts as a constrained layer. When the specimen is vibrating, the shear takes place in the polymer damping layer between the platelets, which increases the mechanical loss of energy, being converted to heat. Moreover, the fact that damping peak in tan δ - T curve of filled LIPN is broadened suggests that the inhomogeneous distribution of filler in the system is inhomogeneous.

Effect of Filler Content

The dynamic mechanical data of mica-filled 35/65 PS/P(EA-nBA) LIPN vary with mica content as shown in Figure 2 and Table II. The results indicate that the

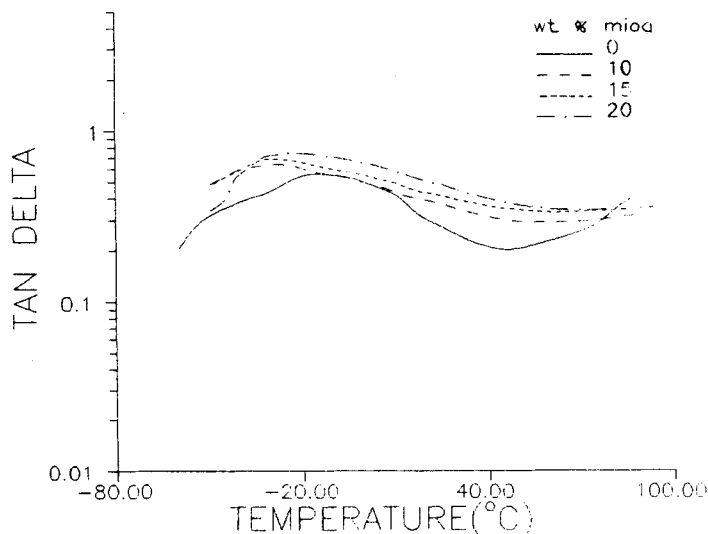


FIGURE 2 Effect of filler content on the damping of PS/P(EA-nBA) LIPNs.

TABLE II

Damping properties changing with mica content

Content (wt%)	tan δ max peak height	Rate of tan δ max increase (%)	Temperature range of tan $\delta > 0.4$ (°C)		
			T1	T2	ΔT
0	0.561	—	-37	12	19
10	0.641	14.3	-55	21	76
15	0.685	22.1	-52	27	79
20	0.745	32.8	-48	41.5	89.5

damping values and width of damping peak increase with increasing mica content in the LIPN. For example, with a 20 wt percent mica-filled LIPN, the tan δ increase reaches 32.8% and the width of damping peak (tan $\delta > 0.4$) extends over a 89.5°C temperature range. It is difficult to prepare the sample that the filler content in the LIPN exceeds by 20 wt percent, however it could be inferred that the damping properties decrease if mica content is too high in the LIPN. The aggregation of particles confines polymer damping performance because of excessive filler in the LIPN. In addition the damping peak of mica-filled LIPN shifts down in tan δ - T curve contrasting with unfilled LIPN. It may be due to strong adhesion between the polymer and filler, resulting in the increase of chain-segment relaxation time.

Effect of PMMA/P(EA-EMA) LIPN

The blend prepared by mixing 35/65 PS/P(EA-nBA) LIPN with 40/60 PMMA/P(EA-EMA) LIPN in an 80/20 ratio has the dynamic mechanical properties shown in Figure 3 and Table III. In tan δ - T curve of 35/65 PS/P(EA-nBA) LIPN, the damping peak is located at -16°C and tan δ values exceed 0.4 over a 49°C tem-

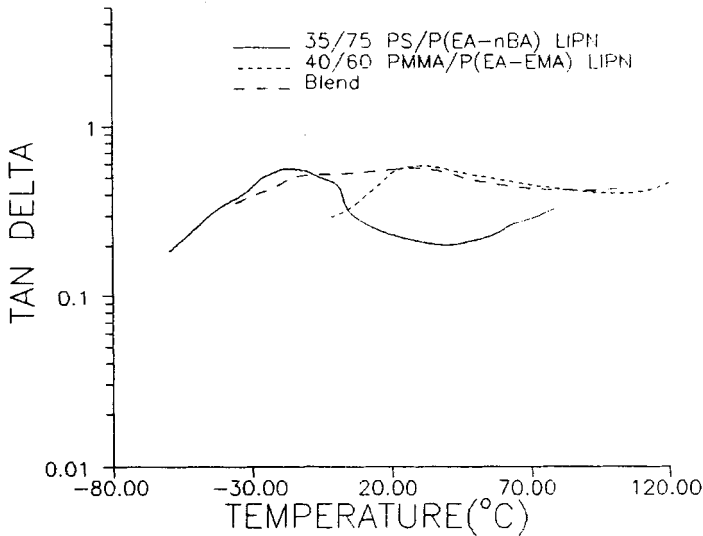


FIGURE 3 Damping properties of the blend of PS/P(EA-nBA) LIPN with PMMA/P(EA-EMA) LIPN.

TABLE III

Damping properties of the LIPN blend

Sample	tan δ max peak height	Temperature range of tan $\delta > 0.4$ (°C)		
		T1	T2	ΔT
PS/P(EA-nBA) LIPN*	0.561	-37	12	49
PMMA/P(EA-EMA) LIPN†	0.583	11.5	100	11.5
Blend	0.563	-27.5	100	127.5

*EA/nBA ration of 50/50 in network I.

†EA/EMA ration of 65/35 in network II.

perature range between -37°C and 12°C . The damping values are higher in lower temperature region, but lower in the room temperature region. The damping peak of PMMA/P(EA-EMA) LIPN is at 31.1°C and the tan δ values are greater than 0.4 over a 111.5°C temperature region between 11.5°C and 100°C . The blending of PS/P(EA-nBA) LIPN with PMMA/P(EA-EMA) LIPN, results in a broader damping peak, tan δ values between 0.4 and 0.563 are noted over a 127.5°C temperature range between -27.5°C and 100°C . This is attributed to the semi-miscibility of PS/P(EA-nBA) LIPN and PMMA/P(EA-EMA) LIPN, and is consistent with results of two LIPN blends reported by Satgurunathan *et al.*¹⁰

Effect of Filler on Damping of LIPN Blend

Ten wt percent mica was added to the blend of 35/65 PS/P(EA-nBA) LIPN with 40/60 PMMA/P(EA-EMA) LIPN. The dynamic mechanical properties of this system are shown in Figure 4. The tan δ values of the blend exceed 0.5 over a 82°C temperature range between -21.3 and 60.7°C , which is superior to those of unfilled

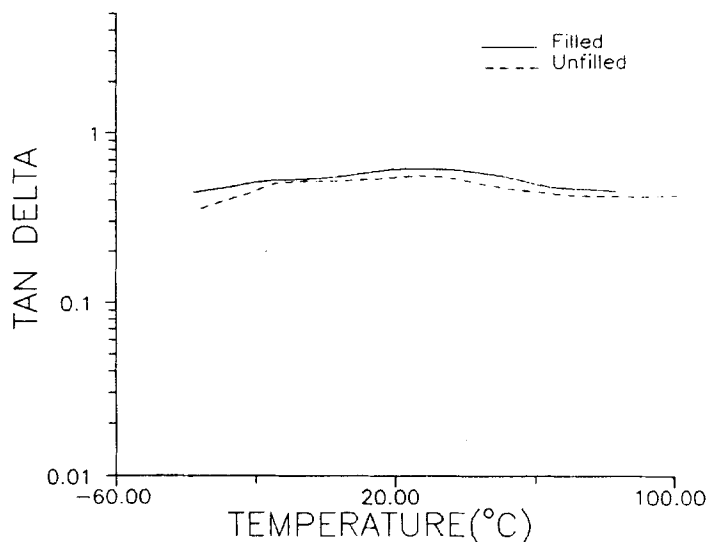


FIGURE 4 Damping properties of the mica-filled LIPN blend.

LIPN blend. It indicates that mica is an effective material to enhance the damping properties over a broad temperature range.

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