Acoustic Absorption Performance of Polyacrylic Composite Latex

KANGDE YAO,^{1,*} GUOXIANG CHENG,¹ TAO LU,¹ WENGSHENG ZHU,² QINGCHI SHUN,¹ TIYONG WANG,³ and DAWEI ZHANG³

¹Department of Materials Science and Engineering, Tianjin University, Tianjin, 300072, People's Republic of China; ²Equipment Institute of Lanzhou Oil Refinery, Lanzhou, 730060, People's Republic of China; and ³Department of Mechanical Engineering, Tianjin University, Tianjin, 300072, People's Republic of China

SYNOPSIS

In this work, copolymers and a multilayer core/shell P(BA/BA-MMA/MMA) composite latex of methyl methacrylate (MMA), butyl acrylate (BA), and ethyl acrylate (EA) were prepared by emulsion polymerization. The thermal and the mechanical properties of their films were characterized by differential scanning calorimeter (DSC) and dynamic mechanical analysis (DMA), while the acoustic absorption performance of dried films were estimated using the standing wave pipe method. The results show that a multilayer core/shell polymer of P(BA/BA-MMA/MMA) has many glass transitions in a broad temperature region, and in the frequency range of sound, this core/shell polymer has better acoustic absorption properties than the random copolymer of P(BA-MMA). © 1995 John Wiley & Sons, Inc.

INTRODUCTION

In recent years, as people lay more and more stress on environmental protection, the technology of damping and noise reduction has been developed to a great extent. In the respect of damping design, damping materials have been advanced from past unitary inorganic, metal, and organic materials to composites, and research activities have reached a submicroscopic level. Generally, damping materials, such as honeycomb structure materials and viscoelasticity ones, convert mechanical energy into a thermal one, which is turned into heat.

As to polymeric materials, mechanical loss performance can be used for a damping design. For example, Interpenetrating Polymer Networks (IPN) and polyblending of low and high T_g polymers^{1,2} have been extensively studied in an effort to obtain the desired broad range of mechanical energy absorption property. Yng-Long Lee et al.³ recently prepared a broader glass transition reign in a polyurethane elastomer dangling system that favored the broad-

ening of sound and vibration damping. S. Gato et al.⁴ have blended polyvinylidene fluoride (PVDF), lead lanthanum zirconate titanate (PLZT), ceramic powder or lead zirconate titanate (PZT), and conducting powder carbon black. On the effect of vibration and noise, ceramic powder changed mechanical energy into electric energy, and then conducting powder transferred electric energy into a thermal one, which was dissipated as heat, so an acoustic insulation and vibration isolation material was obtained. In addition, S. Egusa and coworkers⁵⁻⁷ have showed that paints prepared using PZT ceramic powder as a pigment and epoxy resin as a binder formed a thin film whose piezoelectric activity was high enough for vibration sensor and actuator applications. Therefore, the acoustic absorption performance of damping materials can be improved via these energy transformation mechanisms, and these materials will be developed into intelligent acoustic absorption systems and structures.

In this article, a multilayer core/shell P(BA/BA-MMA/MMA) composite latex was prepared by multistage emulsion polymerization, and its acoustic absorption performance was estimated using the standing-wave pipe method. The acoustic absorption

^{*} To whom correspondence should be addressed.

Journal of Applied Polymer Science, Vol. 58, 565-569 (1995)

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properties of the composite was discussed as well. As to the acoustic absorption properties of the composites of polyacrylate and piezoelectric ceramic powder, these will be discussed elsewhere.

EXPERIMENTAL

Materials

Methyl methacrylate (MMA), butyl acrylate (BA), ethyl acrylate (EA), and sodium dodecyl benzene sulfonate (SDBS) were chemical grade and used as received. Potassium persulfate (KPS) was analytical grade.

Emulsion Polymerization

Deionized water (150 mL) and 0.15 g SDBS were placed into a 500 mL four-necked glass flask equipped with a stirrer, a thermometer, a reflux condenser and a nitrogen duct. The system was heated to 75°C, and then a 46 mL BA monomer and a known amount of KPS aqueous solution were added dropwise into a flask under N₂ atmosphere. After dropping had been done for 30 min, the reaction was then held at 75°C for 2 h until blue fluorescence appeared in the solution. Hence, the seed latex was obtained.

Multilayer core/shell emulsion polymer was prepared by a comonomer mixture of 14 mL and 20 mL MMA or 20 mL MMA monomer stepwise, while a given amount of KPS aqueous solution was incorporated into the mixture. Feeding time was 0.5 h. The reaction was continued for 2 h at 75°C stepwise, and the system was cooled to room temperature at last.

The formulations for polyacrylic composite latex are shown in Table I.

Film Formation

The prepared emulsions were poured onto a 120 \times 120 \times 2 mm³ polyimide film. They were dried at room temperature (28 ± 3°C).

Differential Scanning Calorimeter (DSC) Analysis

Differential Scanning Calorimetry (DSC) spectra from $-28 \sim 127^{\circ}$ C was performed using a DSC-2C module made by America Perkin-Elmer Company. A heating rate of 10° C/min was used in all cases.

Table I	Recipe	for	Polyac	erylic	Composite	Latex
				•		

Sample			
Designation	1	2	3
BA/mL		60	46
BA + MMA/mL			14 ± 26
MMA/mL	40	40	20
EA/mL	60		
KPS/g	0.2	0.2	0.3
SDBS/g	0.15	0.15	0.15
Temperature/°C	75	75	75
Reaction Time/h	2.5	2.5	7.5

Dynamic Mechanical Analysis (DMA)

The dynamic mechanical behavior of films was investigated using a GDP-2 module (Jilin University). The polymer samples were coated evenly onto a braid bundle of glass fiber. The braid bundle was 50 \sim 60 mm in length. The vibration frequency of specimens was 2.5 \sim 5 Hz. All dynamic mechanical date were evaluated at a heating rate of 2°C/min from \sim 60 \sim 120°C under a nitrogen atmosphere.

Acoustic Absorption Coefficient Testing

A latex film with diameter in 10 cm and thickness in 500 \pm 5 μ m was put in a standing-wave pipe [shown in Fig. 1) to measure its acoustic absorption coefficient. The length of the standing-wave pipe was 1 m, and its interior diameter was 10 cm. The acoustic absorption coefficient was determined at a frequency range of 200 \sim 2000 Hz and a temperature of 30°C. In the standing-wave pipe method, the specimen was used as a reflector that was vertical to the propagation direction of the sound wave, and according to the theory of standing-wave interference, the acoustic absorption coefficient was calculated.

The acoustic pressure maximum $(P_{\rm max})$ and the acoustic pressure minimum $(P_{\rm min})$ were measured by moving the speaking trumper vehicle. The acoustic absorption coefficient was computed as follows.

$$s = P_{\rm max}/P_{\rm min} \tag{1}$$

$$\alpha = 4s/(1+s)^2 \tag{2}$$

where s is standing-wave ratio, and α is acoustic absorption coefficient.



Figure 1 Equipment of standing-wave pipe testing.

RESULTS AND DISCUSSION

Acoustic Absorption Coefficient Measurement

In order to make the acoustic absorption materials to dissipate vibration energy completely, they should have not only high damping, but also suitable elastic modules. Besides, these materials are required to have high strength and low density, as well as stable damping properties in a broad temperature range. Acoustic absorption coefficient is often utilized to characterize the effect of these factors on damping materials.

The standing-wave pipe method is usually used to measure the acoustic absorption effectiveness of multiporous materials. However, in our case, the method is used to measure that of polyacrylic composite latex that is an unmultiporous material (Fig. 2). The result shows that the acoustic absorption coefficient of the copolymer of MMA and EA and that of the copolymer of MMA and BA are different in the frequency range of $500 \sim 2000$ Hz, especially in the range of $1000 \sim 2000$ Hz. Therefore, for polyacrylic composite latex membranes, the method can be utilized to characterize their acoustic absorption properties.

Acoustic Absorption Performance of Polyacrylic Composites

Figure 3 shows the acoustic absorption performance of the copolymer of P(BA-MMA) (#2) and multilayer core/shell polymer of P(BA/BA-MMA/ MMA)(#3).#3 has higher acoustic absorption coefficient than #2 in the frequency range of $500 \sim 2000$ Hz, especially in the range of $1000 \sim 2000$ Hz.

The DMA spectra of copolymer of MMA and BA (#2) and multilayer core/shell polymer of P(BA/ BA-MMA/MMA)(#3) are plotted in Figure 4. The #2 specimen is a random copolymer that has a broad glass transition around 20°C. But the #3 specimen of P(BA/BA-MMA/MMA) has three glass transitions at -10, 30, and 94°C, respectively. DSC analysis also confirms the existence of two latter glass transitions whose center corners are 32 and 96°C, respectively. #3 exhibits higher mechanical loss at $-10 \sim 100$ °C, too.



Figure 2 Acoustic absorption coefficient vs. frequency for (1) P(EA-MMA), EA : MMA = 60 : 40 (V/V), (2) P(BA-MMA), BA : MMA = 60 : 40 (V/V).



Figure 3 Acoustic absorption coefficient vs. frequency for (2) P(BA-MMA), BA : MMA = 60 : 40 (V/V), (3) P(BA/BA-MMA/MMA), BA:(BA + MMA):MMA = 46: (14 + 20):20 (V/V).

The morphology of the above polymer of P(BA/ BA-MMA/MMA) may be explained by Figure 5, showing multilayer core/shell structure that can be maintained after film formation. The internal layer is poly(butyl acrylate) with the lowest T_g . The middle one is a copolymer of BA and MMA, whose T_g being 18 ~ 25°C is calculated by the FOX equation. The outer layer is a poly(methyl methacrylate) shell with the highest T_g .

The frequency of sound and mechanical vibration spans from 20 Hz to 20,000 Hz. In general, in the region of glass transition, viscoelasticity polymer chains have mobile freedom to some extent. In a certain range of frequency and temperature, the groups in the polymer chains can be coupled with vibration, then mechanical loss reaches maximum value. Thus, a damping material can be selected by matching its T_{g} within the region of the service temperature at a specific resonant frequency range. But the common damping materials have damping region in rather narrow range (about 30°C). To the system of the latex polymer, in order to gain a damping effect in a broad temperature range, the model of Figure 5 can be used to control the structure of the copolymer and its compatibility.⁸ Hence, as to #3, why it shows the shape in Figure 3, we guess is as follows. Appropriate microphase separation^{9,10} is produced, and multilayer composite polymer is characterized by its multicompositions corresponding multiglass transitions. Moreover, the middle layer P(BA-MMA) may have an effect of disturbance on the motion of P(MMA) and P(BA) mo-



Figure 4 Dynamic mechanical spectra of (2) P(BA-MMA), BA : MMA = 60 : 40(V/V), (3)P(BA/BA-MMA/MMA), BA:(BA + MMA):MMA = 46:(14 + 20):20(V/V), frequency: $2.5 \sim 5$ Hz.

lecular segments and make them penetrate each other. Thereby, forced compatibility¹¹ may be produced by internal reaction of phase boundary to improve the synergistic effect and enhance the relationship of the core and shell. In addition, the structure of the multilayer core/shell polymer P(BA/BA-MMA/MMA) is like a constrained layer damping configuration. The soft shell P(BA) plays a part as a damping layer, while the stiff core P(MMA) plays a role as a constraining layer. Because of the flexural and extensional motions of the damping layer and the shearing action of the constraining layer,^{1,12,13} #3 shows high damping and acoustic absorption properties.

An interesting phenomenon is that the overlap of Figures 2 and 3 shows that the acoustic absorption coefficient for #1 and #3 passes through a minimum at the same frequency of 1000 Hz. It is possible that at the glass transition conditions, because both temperature and frequency take a common effect on the damping properties of polymers, the conversion or degradation of mechanical energy to heat reaches its minimum value at 1000 Hz for system 2 and 3.



Figure 5 Model structure of acoustic absorption material.

As to a more detailed explanation, it needs further study.

CONCLUSIONS

The standing-wave pipe method can be used to study the acoustic absorption performance of polyacrylic composite latex. A multilayer core/shell polymer of P(BA/BA-MMA/MMA) has many glass transitions in a broad temperature region. Because it has appropriate microphase separation, it also has high mechanical loss. In the frequency range of sound, this core/shell polymer has better acoustic absorption properties than the random copolymer of P(BA-MMA).

The authors wish to thank the National Natural Science Foundation of China for the financial support of this research through grant No. 59273114.

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Received September 15, 1994 Accepted April 1, 1995