Experimental and Theoretic Studies on Sound Transmission Loss of Laminated Mica-Filled Poly(vinyl chloride) Composites

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ABSTRACT: The effects of dioctyl phthalate and inorganic filler, mica, on the sound insulation property of poly(vinyl chloride) (PVC) were investigated in this work. The stiffness and mass laws, which are the common theoretic tools to predict the soundproof properties of materials, were used to analyze the sound transmission loss (STL). The experimental results revealed that the stiffness and mass laws can describe well the sound insulation property of PVC/mica composites. The stiffness and surface density are important factors influencing the improvement of STL. With the increase of content of mica, STL and resonance frequency, f_{mn}, of PVC/mica composites increase. Moreover, the change of STL in the stiffness-controlled region is more obvious than that in the mass-controlled region, because the addition of mica in PVC leads to a greater increase in the stiffness. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 122: 1427–1433, 2011

Key words: sound transmission loss; stiffness; surface density; PVC; mica

INTRODUCTION

With the development of modern industry and traffic system, noise pollution has become one of many factors that affect human health and environment worldwide. Therefore, the techniques using sound absorption and insulation materials to reduce ambient noise have received much attention.[1,2] Herein, the viscoelastic polymer materials show the great potential for insulating or damping the sound and vibration due to their unique combination of inherent damping, light-weight, and processable ability compared to metal and inorganic materials[3–5]

However, most polymer materials, which have lower modulus and surface density compared to metal and inorganic materials, cannot be solely used to insulate sound because of their poor sound insulation performance. At present, the application of polymer material in the field of sound insulation has been focused on structure designing of polymeric and metal or inorganic materials, such as constrained layer.[6–8] Here, viscoelastic polymer materials are used as interlayer to increase sound transmission loss (STL) because of their unique mechanical, high damping properties, and low density. Yoon et al.[9,10] studied damping properties and STL of a series of polyurethane composites as damping layer and concluded that the damping loss is the key to energy consuming when the wave of sound transmits through the polymer. Such a sandwich structure, which separates the substrate layer, can weaken the sound transmission in the resonance and coincidence regions. On the basis of above works, materials with multilayered structure have been designed.[11–13] In these materials, the multilayered interfaces lead to the multiple reflection of acoustic waves, and the internal layers have multiform deformation, such as tension, compressing, and shearing that can dissipate more acoustic energy.

However, the layered structure designing of different materials brings some disadvantages: unstable interface, high fabrication cost, and density. Therefore, improving the soundproof ability of single polymer by simple filling modification is attracting researchers' attention.[14–17] Moreover, with the extensive applications of polymer materials in the construction, transportation, and other fields, a higher STL of polymer material itself is also required.

The acoustic insulation ability of single polymer material mostly depended on the surface density, stiffness, and uniformity of material. In general, the addition of inorganic filler in polymer can easily increase these properties. Lee et al.[16] found that the principal factor, influencing the improvement of sound insulations of ABS/CNT composites, was

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enhancement of stiffness by CNT additives. Moreover, consumption and reflection of sound energy also have effect on STL of material. A kind of sound insulation material based on the polymer containing hollow glass microspheres was also studied.[17] The result showed that the dilatation vibration of the microspheres plays a key role in the sound attenuation of the polymer composites in the quasi-static region.

Although the methods to improve the acoustic insulation ability of polymer have been reported, the theoretic study on the acoustic insulation ability of single polymer material is still scarce. The stiffness and mass laws are the common theoretic tools to predict the soundproof properties of some building materials.[18,19] In this work, the acoustic insulation ability of poly(vinyl chloride) (PVC) with different stiffness and surface density was investigated using stiffness and mass-controlled laws. PVC was selected as soundproof matrix material because of its wide use as building and automotive material and its good compatibility with most filler. Mica, a platelike filler, was filled into PVC to adjust its stiffness and surface density. Mica has been reported to improve the vibration damping property[20] and gas barrier ability[21] of polymer composites due to its excellent mechanical, electrical insulation, and thermal properties. However, there was rare report regarding the effect of laminar mica on acoustic insulation property.

EXPERIMENTAL

Materials

PVC (SG-5) with a density of 1.20 g/cm^3 was obtained from Tianjin Dagu Chemical Factory, China. Crystallitic mica (1250 mush) with a density of 1.92 g/cm^3 was provided by Sichuan Tianwei Industry Co., China. Dioctyl phthalate (DOP) was from Luoyang Jiarui Plastic Industrial Co., China. Other additives including lead salt stabilizer, polyethylene wax, and stearic acid were commercial products.

Sample preparation

PVC/mica composites were prepared by blending PVC and mica with various proportions (0, 10, 20, 30, 40, and 50 wt %) for a period of 8 min in a twin roll mixer at 160°C and followed by compression molding at 160°C and 15 MPa pressure for 5 min, then naturally cooled to room temperature under pressure. The sheets were then removed and die cut for the appropriate tests. The contents of processing additives (DOP, combined lead salt stabilizer, lubricant, and processing acid) were, respectively, 0–30, 5, 2, and 2 phr (parts per hundred parts of PVC resin).

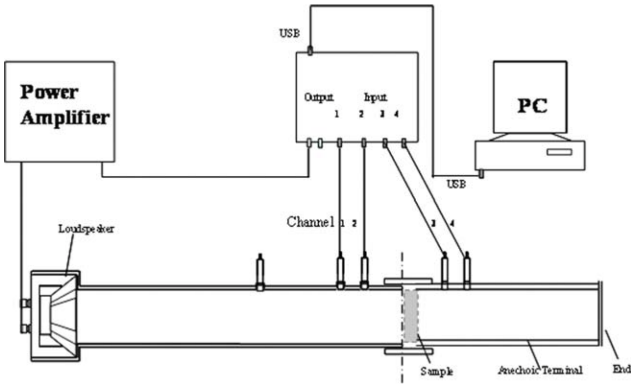

Figure 1 System connection of sound transmission loss testing. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Measurements and characterization

STL is a key quantification of the effectiveness of acoustical insulation for engineering applications and then simply defined as the transmission coefficient (τ) expressed in decibels (dB) according to Eq. (1) below[22]:

$$\mathrm{STL} = 10 \times \log_{10}\left(\frac{1}{\tau}\right) \tag{1}$$

A four microphone impedance tube was provided by BSWA Technology Co., Beijing for the measurement of STL, as shown in Figure 1. Two separate tubes with diameters of 60 and 30 mm were used to measure STL. The frequency ranges on the two tubes were assumed to be 100 Hz to 2.5 KHz for large tube and 1.6 KHz to 6.3 KHz for the small tube per the manufactures impedance testing specifications. An open-ended tube condition and a hard close-ended tube condition were used for both sized tubes. The transfer function method was used to calculate the STL.[23] The dimensions of the specimens were 30 and 60 mm in diameter and 1 mm in thickness. The results were measured as the average of three times.

Tensile modulus of PVC and composites were measured on an SANS CMT4104 Electronic Universal Testing Machine (Shenzhen, China) with dumbbell specimen dimensions of 25 × 6.5 × 1 mm^3 and a crosshead speed of 5 mm/min (GB/T 1040–2006) at 23°C. The results were reported as the average of five samples.

The bulk densities (ρ_m) of the fabricated specimens were obtained through measuring their masses and volumes, and the surface densities of unit area ($\overline{\rho}$) were defined as the ratio of bulk density and thickness of specimens.

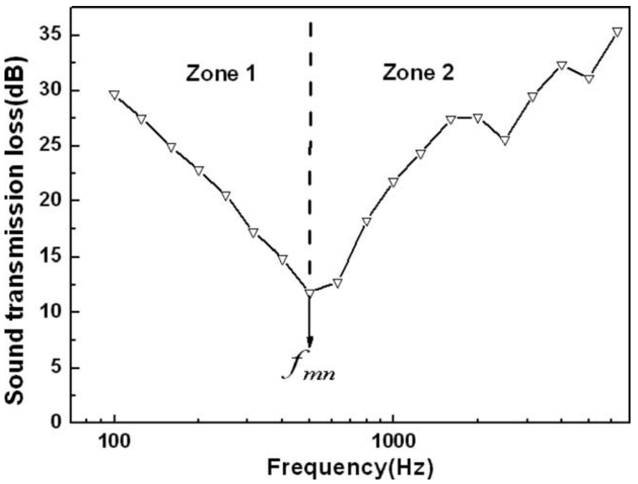

Figure 2 Typical STL versus frequency curve of PVC panel with 30 phr DOP.

Liquid nitrogen cryogenic fractured specimens were used for scanning electron microscope (SEM) observation. After the fractured sections were sputter coated with gold, the morphology observation was taken on a JSM-5900LV (Japan) SEM at an accelerating voltage of 20 kV.

RESULTS AND DISCUSSION

Effect of DOP content on the STL of PVC

Figure 2 shows the dependence of STL of PVC with 30 phr dioctyl phthalate (DOP) on the sound frequency. In this typical curve of STL versus sound frequency (f), a lowest STL value appears at the resonance frequency, f_{mm}, which is determined by the physical parameters and boundary conditions of the specimen. Moreover, the curve can be divided into two zones according to sound frequency. In zone 1 ($f < f_{\mathrm{mn}}$), STL decreases quickly with increasing frequency due to resonance effects. On the contrary, STL increases gradually with the increase of frequency in zone 2 ($f > f_{\mathrm{mn}}$).

Many researchers showed that besides the frequency, the stiffness and mass (or density) of materials can affect greatly the acoustic insulation ability.[16,24,25] In zone 1, the stiffness plays a critical role in the sound insulation, and STL increases with the increasing stiffness. In zone 2, STL of material complies with the mass law and increases with the increase of the surface density of unit area of the panel. Therefore, zones 1 and 2 are also referred to as stiffness-controlled and mass-controlled stages.

As an important processing additive, DOP cannot only improve effectively processability of PVC but also change its stiffness in a large scope. According to the definition,[25] the stiffness of materials, B, can be expressed by the following equation

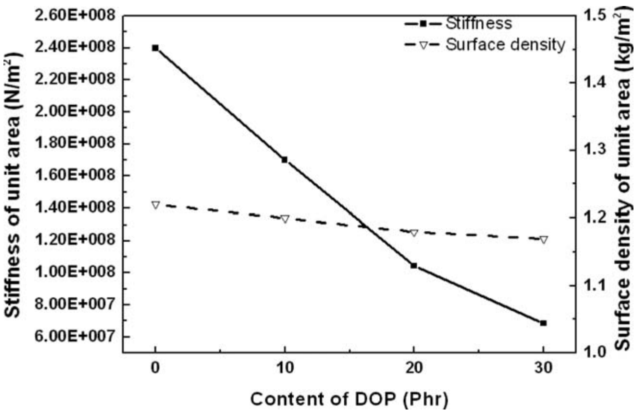

Figure 3 The stiffness and surface density of unit area of PVC with different DOP content.

$$B = \frac{1}{12} \times \frac{Eh^3}{1 - \mu^2} \tag{2}$$

where E, h, and μ are elastic modulus, thickness, and Poisson ratio of materials, respectively.

Figure 3 shows the quantitative relationship of DOP content, stiffness, and surface density of unit area of PVC. It can be found that the stiffness of PVC obviously decreases with increasing DOP content. When the content of DOP changes from 0 to 30 phr, the stiffness reduces by 71%. According to the above discussion, such a large decrease in the stiffness would lead to an obvious change of STL in stiffness-controlled region.

Figure 4 shows the effect of DOP content on STL in the sound frequency range of 100–6300 Hz. It can be clearly found that in zone 1, the curves of STL versus frequency move to the direction of lower frequency with the increase of DOP content in PVC. These changes mean that increasing DOP content

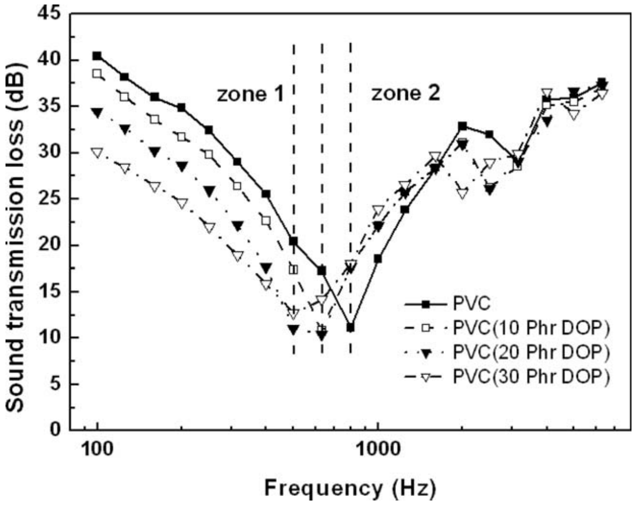

Figure 4 The STL versus frequency curves of PVC with different DOP content.

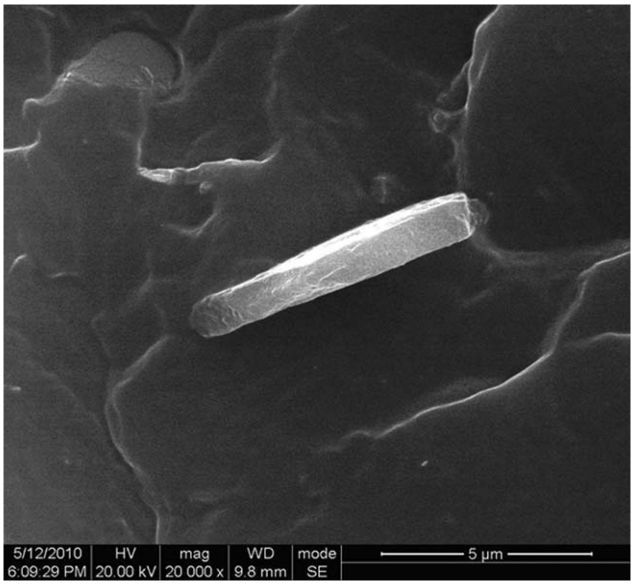

Figure 5 SEM micrograph of mica-filled PVC.

causes simultaneously the decrease of STL and f_{mn}. However, in zone 2, the change of STL is indistinctive. This is attributed to the very small change of the surface density of materials with different DOP content, as shown in Figure 3.

STL of PVC/mica composites

The PVC composites containing different content mica were prepared through melting blend. To ensure the processability, 30 phr DOP was incorporated in composites. Laminar mica can be exfoliated to form platelets in PVC matrix during melting processing because of their good compatibility,[26] as shown in Figure 5. Some works showed the effect of this plate morphology on the vibration damping of specimen.[27] When the specimen is vibrating, the shear takes place in the damping layer between the mica platelets, which increase the mechanical loss of energy being converted into heat. In this part, the effects of mica on the physical properties and STL of PVC were discussed.

Some works reported that the presence of mica platelets restricts the mobility or deformability of PVC molecules and consequently leads to an increase of elastic modulus.[28–30] According to Eq. (1), the stiffness also increases with the addition of mica. Additionally, mica was used here as a kind of high-density filler in this work. Its introduction into PVC can also increase the surface density of PVC composites. Table I shows that the elastic modulus, stiffness, and surface density of unit area of samples increase with increasing the mica content from zero to 50 wt %. The stiffness and surface density are improved by 75 and 27%, respectively. The increase in stiffness is much greater than that in surface density.

In the same way, the dependence of STL of PVC/Mica composites on frequency can be divided into stiffness-controlled (zone 1) and mass-controlled (zone 2) regions as shown in Figure 6. In stiffness-controlled region, STL can be calculated approximately by the following equation[19]:

$$STL\ (\text{stiffness}) = 10\log\left\{\left(\frac{\pi\bar{\rho}f}{\rho c}\right)^2\left[1-\left(\frac{f_{mn}}{f}\right)^2\right]^2\right\} \quad (3)$$

where ρ is the density of air and c is acoustic speed in air.

For single panel, the resonance frequency (f_{mn}) can be evaluated by Eq. (4).

$$f_{mn} = 0.45C_p h\left[\left(\frac{m}{a}\right)^2+\left(\frac{n}{b}\right)^2\right] \quad (4)$$

where a, b are the dimensions of panel; m, n are integers 1, 2 … and C_p is the speed of longitudinal wave in material, which can be calculated by Eq. (5).

$$C_p = \frac{E}{[\rho_m(1-\mu^2)]} \quad (5)$$

In our experiment, the tested samples in impedance tube were cut to 60 and 30-mm diameter circular plate. So, the f_{mn} can be calculated by the following equation:

$$f_{mn} = 0.45\frac{Eh}{[\rho_m(1-\mu^2)]}\left[\left(\frac{m}{\sqrt{\pi}r}\right)^2+\left(\frac{n}{\sqrt{\pi}r}\right)^2\right] \quad (6)$$

where r is the radius of panel.

According to eqs. (3) and (6), STL in stiffness-controlled region increases with the increase of the elastic modulus of material and decreases with the increase of measured frequency. This result is consistent with the above discussion.

TABLE I
The Effects of Mica Content on the Elastic Modulus, Stiffness, and Surface Density of Unit Area of PVC/Mica Composites

Content of mica (wt %)	Elastic modulus (MPa)	Stiffness of unit area ($\times 10^7 \mathrm{N/m^2}$)	Surface density of unit area ($\mathrm{kg/m^2}$)
0	721	6.85	1.22
10	741	7.04	1.28
20	820	7.8	1.33
30	900	8.57	1.4
40	1173	11.2	1.48
50	1260	12	1.56

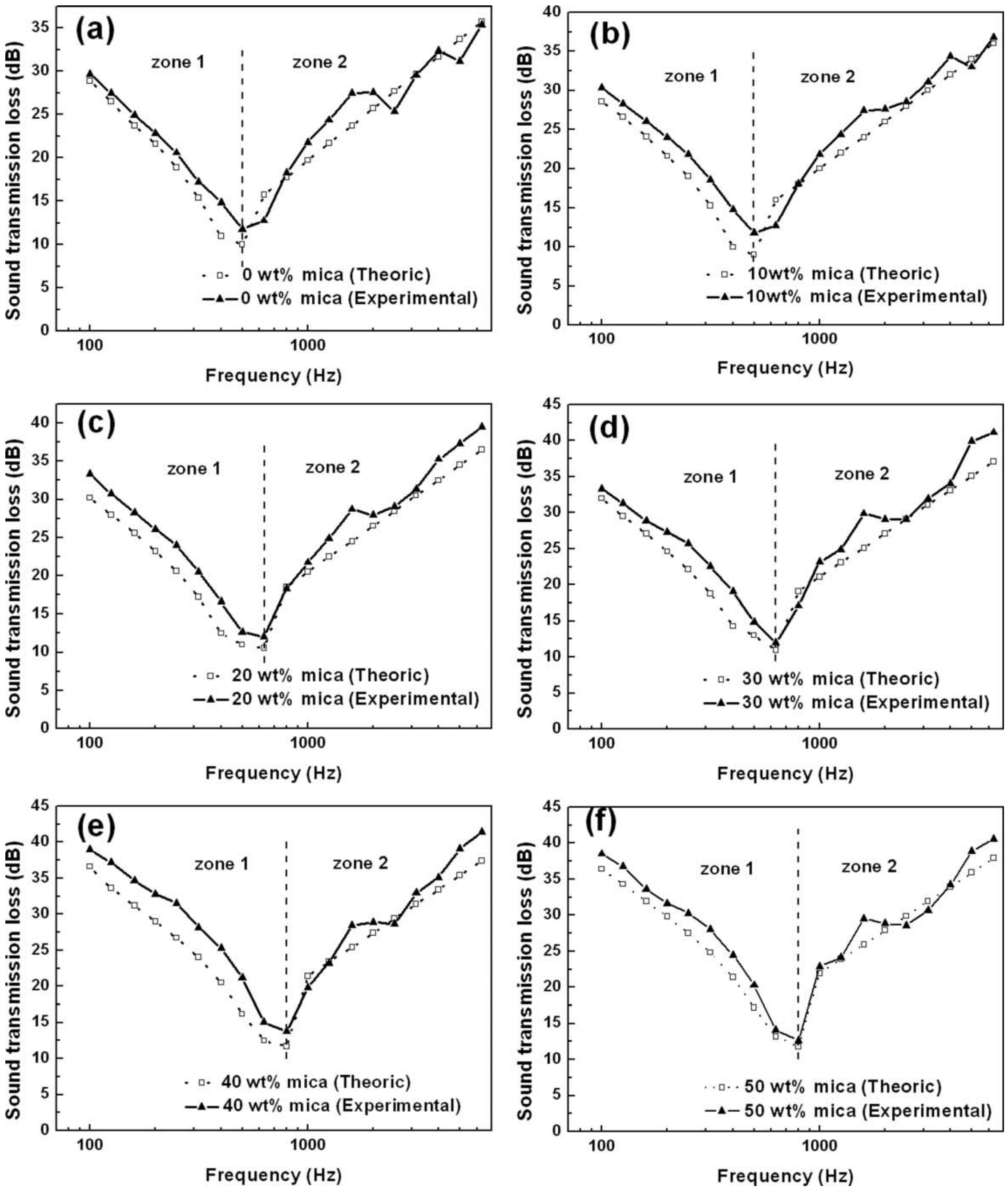

Figure 6 (a–f) Theoretic and experimental results of the STL for PVC/mica composites with different mica contents.

TABLE II
The Experimental and Theoretic Resonance Frequency (f_{mn}) of PVC/Mica Composites with Different Mica Contents

Content of mica (wt %)	Experimental (Hz)	Theoretic (Hz)
0	500	538
10	500	540
20	630	562
30	630	586
40	710	723
50	710	735

In mass-controlled region (zone 2), when sound wave impacts a specimen, the specimen vibrates according to the change of the atmospheric pressure. The vibration energy dissipates during the transmitting process from inside to outside of the specimen and increases according to the increase of the weight of the material.[31] This relation is called the mass law of sound insulation, that is, Eq. (7), by which STL in zone 2 can be calculated.

$$STL\ (\text{mass}) = 10\log\left(\frac{\pi\overline{\rho}f}{\rho c}\right)^2 = 20\log\overline{\rho} + 20\log f - 42 \tag{7}$$

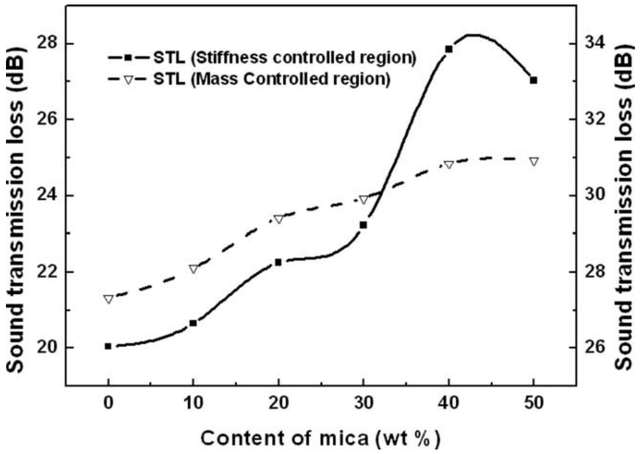

Figure 7 Average STL values of PVC composites with different mica contents in the frequency range of 100–6300 Hz.

Thus, the theoretic STL and f_{mn} values of PVC composites with different mica content in both stiffness-controlled and mass-controlled regions, which are illustrated in Figure 6(a–f) and Table II, can be obtained according to eqs. (3), (6), and (7). At the same time, the experimental values of STL and f_{mn} measured by impedance tube are also illustrated in Figure 6(a–f) and Table II. It can be found from Figure 6(a–f) that, for all samples with different mica content, the values of STL obtained through theoretic calculation are very approximate to those obtained through experimental measurement. This indicates that the stiffness and mass laws can describe well the acoustic insulation ability of PVC/mica composites and provide a reliable guidance for the design and optimization of soundproof composite with fillers. It is also noted from Figure 6(a–f) that the experimental values of PVC/mica composites are slightly greater than the theoretic values. This difference may be attributed that the sound damping by reflection or scattering would be increased when the layered mica fillers act as another wall for sound blocking. Moreover, when the fillers are dispersed in the wall of PVC, we can anticipate that the energy dissipation as heat through hysteresis can be increased, because the wall of PVC should move together with the massive inorganic when it recovers its deformation caused by sound waves.[32] So, relative to the matrix, the composites filled with mica have many contributions coming from the frictional loss and the platelet structure, which may increase the sound insulation capacity.

The change of STL with frequency of all curves in Figure 6(a–f) shows the similar trend. In stiffness-controlled region (zone 1), the STL decreases quickly with increasing sound frequency and reaches the minimum at f_{mn} due to resonance results. And then, the STL begins to increase with increasing of sound frequency according to mass law when sound frequency is higher than f_{mn}. Table II listed the experimental and theoretic f_{mn} values of PVC/mica composites with different mica contents. It can be found that the experimental and theoretic values are relatively close. The differences may originate from the restraints on rotation of edges of the fixture during the measurement. The addition of mica affects greatly f_{mn}, which increases with the increase of content of mica due to the improvement of stiffness of composites.

For a more comprehensive comparison of the acoustic insulation ability of PVC/mica composites with different mica contents, the average values of STL of PVC/mica composites in stiffness-controlled and mass-controlled regions were plotted in Figure 7. It can be found that the average STL value in stiffness-controlled region increases obviously with increasing mica content up to 40 wt % and then has a small drop with the further increase of filler content. The average STL value increases by 40% with the addition of 40 wt % of mica in PVC, because the stiffness of composites increases greatly as shown in Table I. Therefore, in stiffness-controlled region, it might be concluded that the increasing stiffness plays a principal role in the improvement of the STL of PVC/mica composites. However, with the further increase of the content of mica, the uniformity of composites becomes worse. This results in a decrease in STL.

On the other hand, the average STL in mass-controlled region also increases with increasing mica content due to the increase of surface density of composites. However, the maximum increase of STL is only 12.8%, because the surface densities of composites are not greatly improved with increasing mica content, as shown in Table I. Therefore, the addition of mica leads to more significant change of the acoustic insulation ability in the stiffness-controlled region.

CONCLUSIONS

In this research, the STL of PVC with different DOP content was investigated. It is found that the STL decreases with increasing of DOP content due to the reduction of stiffness. Moreover, various amounts of mica were introduced into the PVC matrix to improve the sound insulation property. The results show that the STL value of PVC/mica is improved obviously because of the introduction of mica, especially in stiffness-controlled region. The increased average STL can be a significant value in practical noise reduction. By the stiffness and mass laws, the sound insulation efficiency for soundproof composite can be evaluated, and contribution of mica on sound insulation is confirmed. It is demonstrated that the increased stiffness and the unique structure of microconstrained layer as the addition of flake mica induce the improvement of acoustic attenuation. As a future work, the practical applications of the thin and light-weight sound insulation material will be developed.

APPENDIX

Parameters used in the article.

Symbols	
τ	sound transmission coefficient
ρ_m	bulk density of specimen
$\bar{\rho}$	surface density of unit area of specimen
ρ	density of air
f	sound frequency
f_{mn}	resonance frequency
B	stiffness of specimen
E	elastic modulus
h	thickness of specimen
μ	Poisson ratio
c	acoustic speed in air
a, b	dimensions of specimen
m, n	integers 1, 2...
C_p	speed of longitudinal wave in material
r	radius of specimen in impedance tube

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