Acoustic Characteristics Parameters of Polyurethane/Rice Husk Composites

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The interest of this article lies in the proposition of develop the relationship of the adding of Rice Husk (RH) and acoustic characteristic parameters of Polyurethane (PU) foam. In this article, different amount of RH (2%, 5%, and 8%) was employed into the formation of PU foam system. A rapid method based on professional acoustic measurement was used to determine the acoustic characteristic parameters of the PU foam and PU-RH composites, such as porosity, flow resistance, tortuosity, and characteristic constants, and analyze the effect of RH on the acoustic characteristic parameters. The measured results indicated that the sound absorption coefficients of PU-RH composites increased at low frequency along with a small decrease at high frequency on the addition of RH. And adding more RH will be helpful to improve the low-frequency sound absorption performance of PU foam without increasing the thickness. With the increase of RH additive amount, the flow resistance decreased first and then increased, while the porosity declined slowly generally. Overall, the PU-RH composite containing 5% RH showed the most satisfactory absorption performance. POLYM. COMPOS., 40:2653–2661, 2019. © 2018 Society of Plastics Engineers

INTRODUCTION

In recent decades, with the rapid development of urban construction, industrial production, and transportation, people's awareness of environmental protection and requirements for quiet environment have been increasing. However, the noise problem has developed into one of the three world environmental problems that need to be dealt with. At present, the noise reduction methods that have been put into use in life mainly include the establishment of sound barriers on both sides of traffic roads, the building sound insulation performance of doors and windows, the laying of sound absorbing materials on major traffic roads, and the addition of noise reduction systems for vehicle. After practice, people found that using noise reducing materials with specific structure in the transmission route (adding traffic road sound barriers) is an economic, simple, and effective method.

Polyurethane (PU) is a type of polymer produced by reacting a polyhydric alcohol with an isocyanate to produce a repeating group in the main chain. PU porous materials have the advantages of one-time processing and molding, light weight, low price, ease of prepare, high efficiency insulation, and waterproof [1,2], as well as excellent mechanical properties, acoustic properties, and chemical resistance. PU is widely used in the fields of sound absorption, heat insulation, food industry, medical field, conference rooms, concert halls, stadiums, automotive interior parts, etc. [3,4]. At present, adding various reinforcing particles in PU porous materials is an important method on improving the acoustic properties. The effective fillers used are mainly powder substance such as metal powder and vermiculite powder, granular material as metal particles, glass beads, polymer microspheres, and rubber particles, flaky substances such as mica plate and fibrous substance as plant fibers and glass fibers. Most of these materials are organic or inorganic bubble-containing fillers, which can form a uniform cavity and increase internal friction to improve the sound absorption performance of the material. Mott [5] added rubber into PU system and studied the dynamic and acoustic properties. Zhou et al. [6] developed hybrid rubber/PU composite and acoustic measured results showed that the powder rubber/PU composite foam has a relatively uniform cell structure and good sound absorption

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and insulation properties. Scarpa [7] investigated the effect of iron particle in PU foam on the acoustic properties, and Wang [8] analyzed the influence of carbonyl iron particles on acoustic absorption properties of magnetic PU foam. Ono [9] prepared unidirectional fiber-reinforced PU foam and discussed the acoustic characteristics. Khanouki [10] used silicon dioxide nanoparticles to improve the acoustic damping of PU foams. Sung and Kim [11] investigated the influence of inorganic fillers surface characteristics on the acoustic performance of PU composites, and the fillers are talc, zinc borate, and aluminum hydroxide. Chuang et al. [12] chose carbon fibers and glass fibers to prepare fiber-reinforced PU foams and studied the acoustic and mechanical properties. Ji et al. [13] investigated the effect of the graphene on the sound absorption coefficients, airflow resistance, and cell size of PU foam. Xi et al. [14] added a proper amount of fillers such as metal powder, glass beads, and mica flakes to the PU and found that the internal loss of the material increased and the sound absorption performance of the PU composite improved. Li et al. [15] added mica filler into PU elastomer, and found the mica filler can improve its low-frequency sound absorption performance, and within the range of 40–80 mesh, the low-frequency sound absorption performance increased with the increase of the filler size.

However, most PU investigated currently is polyether PU that the degradation performance is very poor, and will cause irreversible damage to the environment. Many researchers try to increase the degradation speed of PU by adding some biodegradable substance, of which natural fibers, especially tea-leaf-fiber [16,17] attract people's attention. Heriyanto [18] studied the effect of ramie fiber on the acoustic performance of polyester-ramie fiber composites. Rizzo et al. [19] analyzed the influence of banana fibers and cellulose in PU composites on the thermal insulation and acoustic performance. Chen [20] studied the acoustic property of PU foam with addition of bamboo leaves particles. Lou et al. [21] prepared a series of PU composites with bamboo fiber and analyzed the sound insulation performance. Choe et al. [22] tried to improve the sound absorption coefficients of PU foam by adding chemically treated wood fibers and analyzed the influence of open porosity, airflow resistance, and tortuosity of the foam materials. Besides, hemp [23], sisal [24], flax [25], and oil palm [26] are also commonly used acoustic material additives.

It should be noted that more and more studies are focus on the potential of under-utilized waste materials-byproducts from agricultural products with the requirement of green energy conservation, such as wheat straw [27], rice straw [28], corncobs [29], and so on. Rice husk (RH) is one of the most common agricultural wastes in China, which has won the favor of many researchers. Petchwattana [30] and Panthapulakkal [31] studied the mechanical properties and the influence of coupling agents on polyethylene/RH composites. Deeptangshu [32] give a detail review of polymeric composites mixed with RH. Rozman [33] analyzed the mechanical and physical properties of PU-RH composites. Till date, only a few investigations considered the effect of RH on the sound absorption performance of PU foam. Wang [34] presented the influence of RH on the acoustic performance of PU.

However, the acoustic properties of porous materials mainly depend on their inherent characteristics. Studying the characteristic parameters of porous materials is not only beneficial to control and optimize the use of porous materials, but also help to choose the most appropriate materials for specific application needs. The measurements of these characteristic parameters are complex. Such as Johnson–Allard model requires 5 parameters, Lafarge–Allard model requires 6 parameters, and Biot–Allard model takes into account the elastic skeleton of the material and requires 9 parameters. Each parameter requires a separate device, so the whole process is complex, expensive, time consuming, and prone to large errors. In this article, a test bench developed by Ecole Central de Lyon, based on Johnson–Allard and Lafarge–Allard models, was used to measure all the parameters of acoustic propagation in porous media from the angle of acoustic measurement.

MATERIALS AND METHODS

Synthesis of PU-RH Composites

In this study, one step foaming method was selected to prepare PU and PU-RH composites with polyether polyol HEP-330N, polymer polyol TPOP-36/28, catalysts A33 (triethylene diamine, the concentration is 33%), and A-1 [mixture of 70% bis (2, dimethylamino ethyl) ether and 30% dipropylene glycol], deionized water, silicone oil, and modified MDI. The ratio of polyol and modified MDI is 60:40. You can check the details about the specific proportions and preparation methods in my previous article (Influences of Rice Hull in Polyurethane Foam on Its Sound Absorption Characteristics). The addition of RH was 2%, 5%, and 8%, respectively. The self-made foaming mold is made of stainless steel 304, the diameter is 100 mm. The mold for the preparation of PU and the filler RH was shown in Fig. 1.

Measurement of Sound Absorption Coefficients

Acoustic performance of the PU foam and PU-RH composites were measured with a four-microphone impedance tube based on ISO10534-2:2002 (E) through Transfer Function Method [35]. The diameter of the tube we used here is 100 mm, and the corresponding test frequency is 50–1,600 Hz. The incident sound wave was perpendicular to the surface of the foam rise direction. Each of the tests was repeated with at least five samples to obtain consistent and representative results. The thickness of the sample is 40 mm. The prepared test samples were shown in Fig. 2.

Measurement of Acoustic Characteristic Parameters

Test Philosophy. The test philosophy is based on the impedance tube method of sound absorption coefficients of

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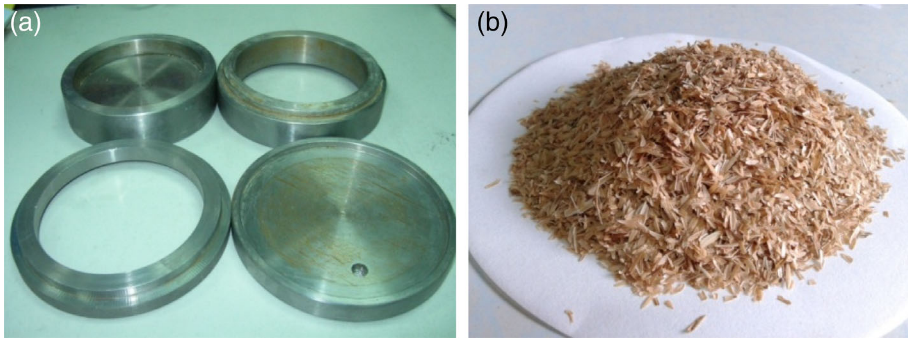

FIG. 1. The mold for the preparation of PU foam (a) and the filler RH (b). [Color figure can be viewed at wileyonlinelibrary.com]

FIG. 2. The test samples of PU and PU-RH composites. [Color figure can be viewed at wileyonlinelibrary.com]

porous materials. The sound waves generated by main acoustic source spread to the sample surface, a microphone located at the center of the tube is used to collect sound pressure and estimate the surface impedance of the sample, and further to determine the characteristic parameters based on sound propagation theory models in porous medium. The principle diagram is shown in Fig. 3.

At present, the most widely used rigid porous material model is Johnson–Allard and Lafarge–Allard models that can be characterized by a series of intrinsic parameters. While Johnson–Allard model is expressed by flow resistance σ, porosity Φ, tortuosity α_∞, thermal characteristic constant Λ, and viscous characteristic constants $\Lambda^{'}$. Two characteristic length are related to their shape factors s and $s^{'}$, respectively.

Viscosity effects [36] and thermal effect [37] in Johnson–Allard model

$$\rho_e(\omega) = \alpha_\infty \rho_0 \left(1 - \frac{j\sigma\Phi G(\omega)}{\rho_0 \alpha_\infty \omega}\right) \tag{5}$$

with

$$G(\omega) = \left(1 + \frac{4j{\alpha_\infty}^2 \eta \rho_0 \omega}{\sigma^2 \Phi^2 \Lambda^2}\right)^{1/2} \tag{6}$$

$$K(\omega) = \frac{\gamma P_0}{\gamma - (\gamma - 1) H(\omega)} \tag{7}$$

with

$$H(\omega) = \frac{1}{1 + \frac{8\eta}{j\Lambda'^2 \rho_0 P_r \omega}\left(1 + \frac{j\Lambda'^2 \rho_0 P_r \omega}{16\eta}\right)} \tag{8}$$

While Lafarge–Allard [38,39] model introduce a thermal permeability$k_0^{'}$,which is defined with viscous permeability similar to Darcy's law, can be expressed by the trapping constant$\Gamma$of porous materials

$$k_0^{'} = M^{'} / \Gamma \tag{9}$$

For cylindrical holes with a circular section

$$\Gamma = \frac{8}{\Phi \Lambda'} \tag{10}$$

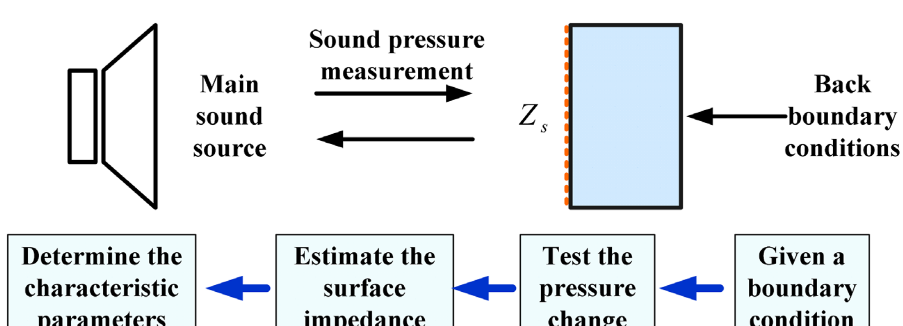

FIG. 3. Principle diagram of characteristic parameters of porous material. [Color figure can be viewed at wileyonlinelibrary.com]

Effective density does not consider saturated the viscous and inertia coupling between fluid and skeleton, therefore, its expression is not change due to import the heat permeability, and dynamic compression produce a new form contain $M^{'}$

$$K(\omega)=\frac{\gamma P_0}{\gamma-\frac{\gamma-1}{1-j\frac{\omega_0^{'}}{\omega}\left(1+j\frac{M^{'}}{2}\frac{\omega}{\omega_0^{'}}\right)^{1/2}}} \tag{11}$$

where, $\gamma$is the specific heat of air and $M^{'}=1$ for Johnson–Allard model.

The flow resistance σ of the sample is first determined according to the asymptotic approximation of the Lafarge–Allard's model. And the real part of the surface impedance of a porous material backed with condition of $P=0$ at low frequency is proportional to the flow resistance

$$Z_s=j\left(e\frac{\rho_0\alpha_\infty}{\Phi}\left(1+\frac{M}{4}\right)-3\frac{\sigma^2\Phi}{3P_0}e^3\right)\omega+\sigma e \tag{12}$$

The resistance is deduced through the thickness e of the material

$$\sigma=Re(Z_s)/e \tag{13}$$

Then the porosity $\emptyset$ can be determined according to the same theory with that of flow resistance. While the imaginary part of the surface admittance of a porous sample backed by a rigid wall depends on the frequency and the porosity as following

$$A=\frac{1}{Z_s}=\left[\frac{e\Phi}{\omega_0^{'}P_0}\left[\left(1+\frac{M^{'}}{4}\right)-\frac{1}{\gamma}\left(1+\frac{M^{'}}{4}\gamma\right)\right]+\omega_0\frac{\rho_0\alpha_\infty\Phi}{P_0{}^2}\frac{e^3}{3}\right]\omega^2+j\frac{\omega e}{P_0}\Phi \tag{14}$$

The porosity is then deduced by introducing the known static pressure P_0

$$\Phi=\frac{P_0}{e\omega}Im(A) \tag{15}$$

The values of the flow resistance and porosity can be obtained with above methods. In Johnson–Allard model, the other three parameters α_∞,s, and $s^{'}$can be acquired with least square method by fitting Johnson–Allard model with the surface impedance (or absorption coefficient) test curve of the sample. In case of the Lafarge–Allard model, parameters α_∞, Λ, $\Lambda^{'}$, and $k_0^{'}$ can be identified at the same time. They can be divided into two groups (α_∞,Λ) and ($\Lambda^{'}$,$k_0^{'}$), and determined with the least square method based on the fitting of effective density and dynamic compression.

Test Equipment. The self-made test bench in Ecole Centrale de Lyon is shown in Fig. 4, and prepared PU foam and PU-RH composites with a diameter of 100 mm under various conditions were cut to 55 × 55 mm (Fig. 5), test bend is 50–2,500 Hz. The test bend is 30–300 Hz for the test of flow resistance while 50–2,500 Hz for others parameters. The boundary conditions behind the sample can be rigid wall, different thickness of air layer, and soft impedance (Fig. 6).

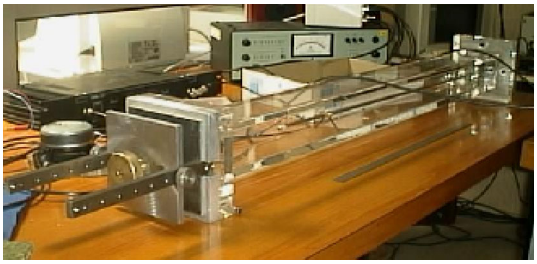

FIG. 4. Designed rapid test test-bed. [Color figure can be viewed at wileyonlinelibrary.com]

RESULTS AND DISCUSSIONS

Cell Morphologies

The pore structure of porous acoustic material should be tested before measuring the acoustic performance because the pore morphology is an important factor that affects their acoustic properties. Therefore, Fig. 7 showed the scanning electron micrograph of PU foam and Fig. 8 displayed the scanning electron micrograph of PU-RH composites. It can be seen that PU foam exhibits a typical porous morphology, the pores are connected to each other and the pore size distribution is relatively uniform. The larger pore is about 0.4–0.5 mm in diameter, and there are many smaller pores on the grain between the pores, the small pore is about 0.1–0.2 mm. Almost all the holes are interconnected, and the higher opening rate makes the PU foam has good sound absorbing property.

It was apparent that the pore size distribution was very uneven after the RH was added into the PU foam, the pores in the vicinity of RH became much smaller and numerous, and the pores far from the RH became larger and uneven. This is mainly due to the blocking effect of the RH in PU foam. The generated bubbles cannot be released quickly in the molding process of PU foam, and trapped by the RH, deposited close to the RH and turned into small holes. While the hindrance is less for the pores far away from the RH, the

FIG. 5. Prepared PU and PU-RH samples. [Color figure can be viewed at wileyonlinelibrary.com]

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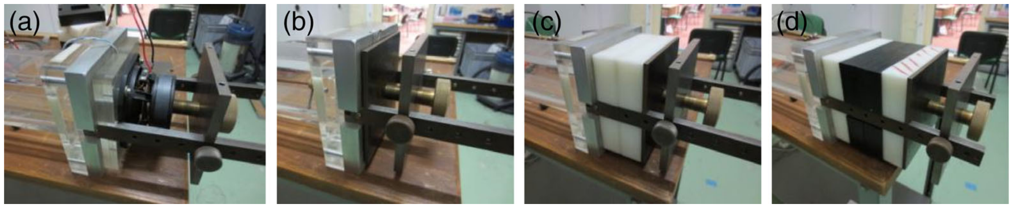


FIG. 6. Different boundary conditions behind the sample: (a) soft impedance; (b) rigid wall; (c) air layer of 60 mm; and (d) air layer of 120 mm. [Color figure can be viewed at wileyonlinelibrary.com]

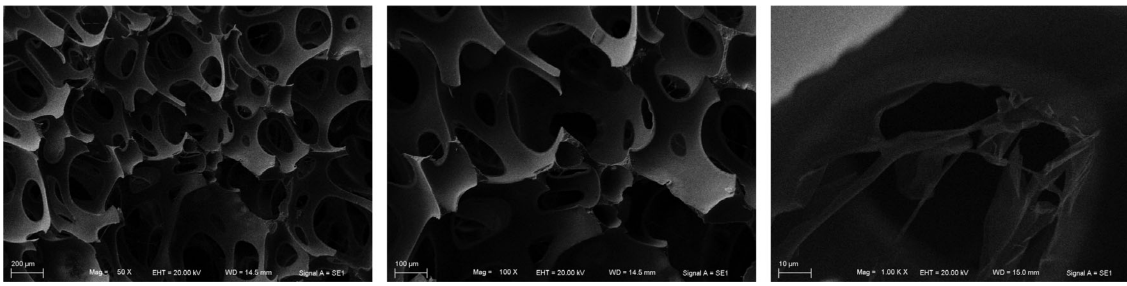
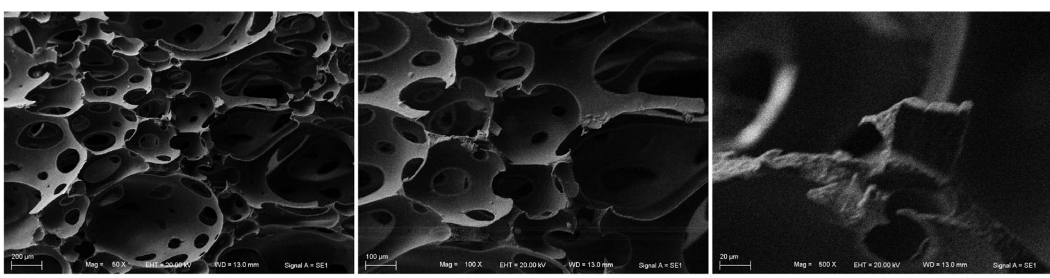
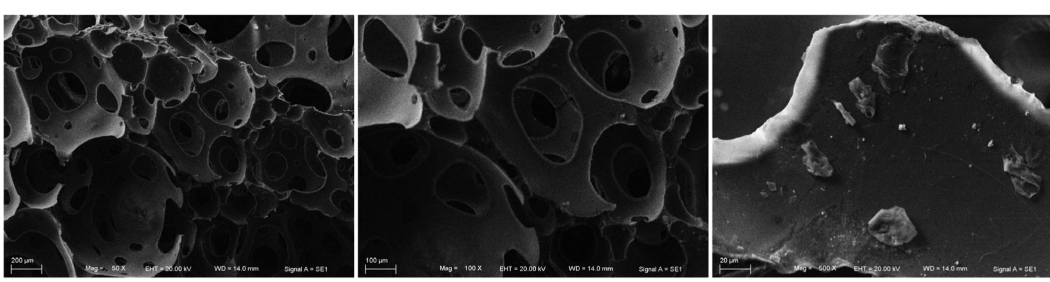


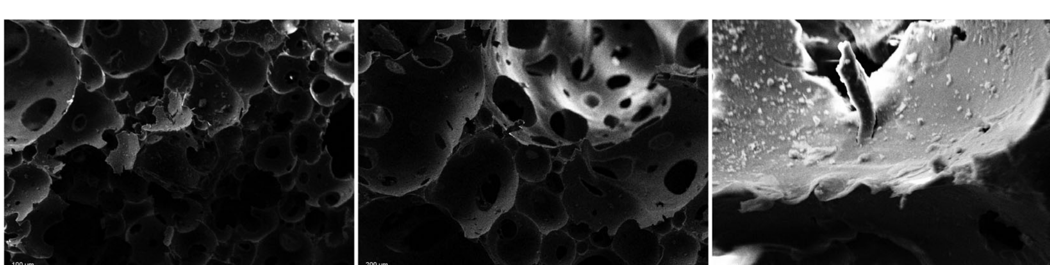
FIG. 7. Some SEM micrographs of PU foam.



(a) PU-RH composites with 2% RH



(b) PU-RH composites with 5% RH



(c) PU-RH composites with 8% RH

FIG. 8. Some SEM micrographs of PU-RH composites (a) PU-RH composites with 2% RH, (b) PU-RH composites with 5% RH, and (c) PU-RH composites with 8% RH.

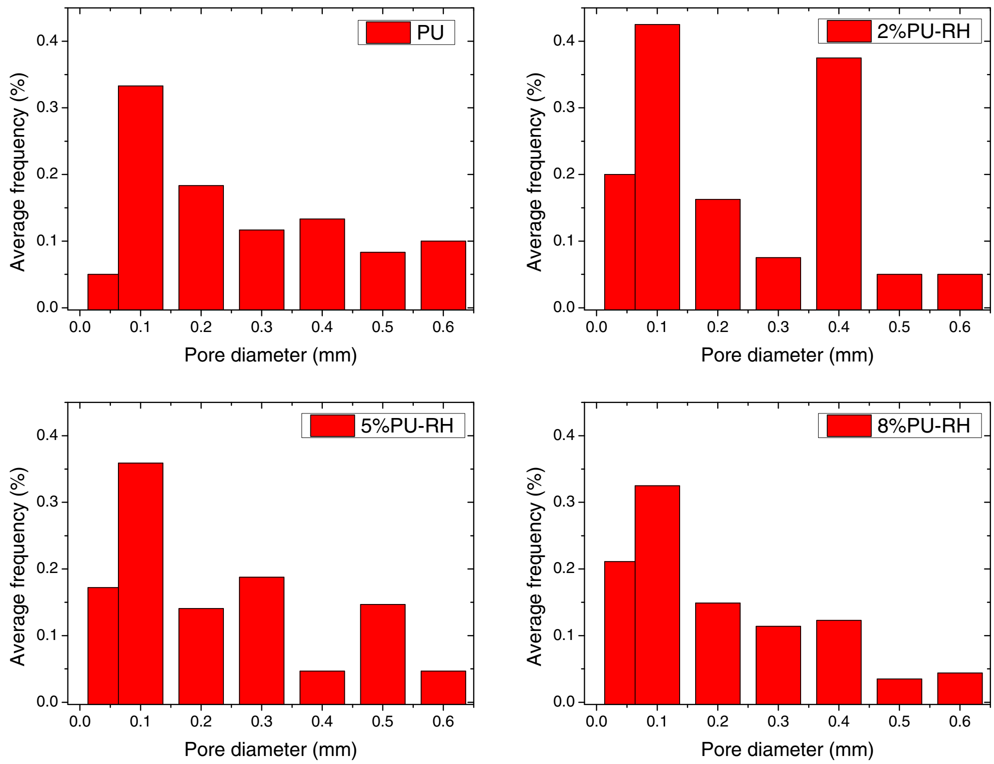


FIG. 9. Pore diameter distributions of PU and PU-RH composites. [Color figure can be viewed at wileyonlinelibrary.com]

bubbles are also squeezed, which caused some bubbles to merge into larger holes. This exacerbates the uneven of pore size distribution of PU-RH composites. Another obvious difference is that the skeleton between the pores of PU-RH are covered with small holes compared with PU foam.

Pore Diameter Distributions

In order to better analyze the specific effect of RH addition on the pore size of PU foam, it is necessary to give the pore size distributions of PU and PU-RH composites.

Figure 9 shows the pore diameter distribution of each sample obtained with the statistical analysis method. Each foam was repeated with three different samples to obtain the average value. As it can be seen from the figure, the maximum pore size distribution is 0.1 mm for all the PU and PU-RH composites. The distribution of PU foam is relatively uniform. Once RH was added into PU foam, there were a large number of tiny holes appeared on the frame between the pores, which may be helpful to the sound absorption performance of PU porous materials.

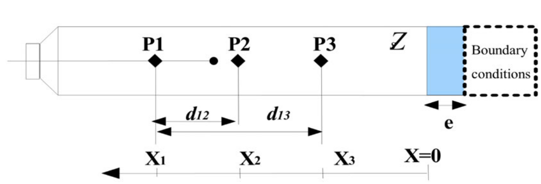


FIG. 10. Measurement of sound velocity in impedance tube (where, the three points P_1,P_2,P_3 correspond to the horizontal axis X_1,X_2, and X_3. The sound pressures p_1 and $p_2$that measured at the points P_1 and P_2, is dominant in the estimation of the components of the echo and the incident wave, and can be used to deduce the theoretical sound pressure value of pointp_3). [Color figure can be viewed at wileyonlinelibrary.com]

Measurement of the Sound Velocity and Air Density

The characteristic parameters of different PU-RH composites were tested. In order to control the influence of sound speed accurately and make sure the experimental results are repeatable, two methods are used here, method 1 is ideal gas state equation method and the method 2 is dual microphone method.

For method 1, the sound speed$c_0$is obtained based on ideal gas law:

$$c_0 = \sqrt{\gamma r T_0} \tag{16}$$

where, $\gamma$is the ideal gas constant, this method depends only on ambient temperature.

In case of method 2, we did not consider the change of temperature, but to obtain the change of acoustic pressure in the impedance tube, and it is more accurate. As shown in Fig. 10, the value of sound velocity is deduced by measuring the change of sound pressure at three different positions in the impedance tube.

$$p_3 \sin(k_0 x) = p_2 \sin(k_0 y) - p_1 \sin(k_0 (y - x)) \tag{17}$$

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TABLE 1. Sound velocity and air density for PU-RH composites.

Sample	Air density (g/cm^3)	Sound speed (m/s method 1)	Sound speed (m/s method 2)	Sound velocity difference (m/s)
Contrast	1.1708	342.6977	345.9000	3.2023
2% RH	1.1813	341.4926	346.1000	4.6074
5% RH	1.1595	344.5027	346.6000	2.0973
8% RH	1.1587	344.4554	347.9000	3.4446

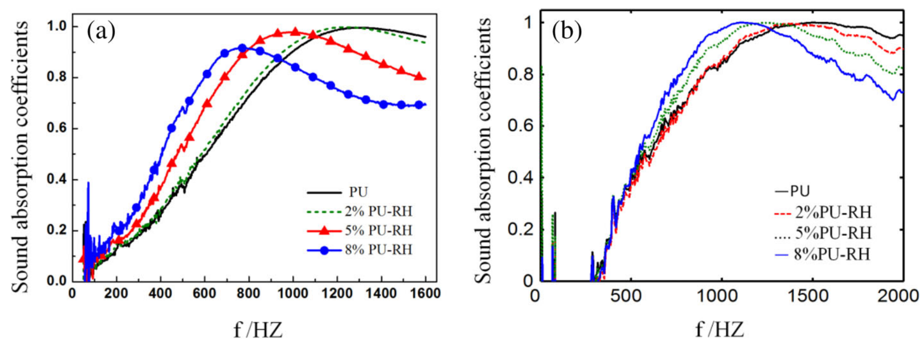

FIG. 11. Contrast of sound absorption coefficients of PU-RH composites (a is for impedance tube manufactured by BSWA TECH, b is self-made). [Color figure can be viewed at wileyonlinelibrary.com]

where, $k_0 = \omega/c_0$is the propagation constant of air. x and y were obtained by x_1,$x_2$and x_3, respectively.

$$\begin{cases} x = x_1 - x_2 \\ y = x_1 - x_3 \end{cases} \tag{18}$$

The values of sound speed and air density with two methods are shown in Table 1. The results showed that the difference with sound speed of two methods is relatively small, reveals that the air in the laboratory is close to the ideal gas. In order to test accurately and consistent with the previous two test results, double microphone method was chosen here.

Sound Absorption Coefficients of Different PU-RH Composites

Sound absorption coefficients of PU-RH composites tested with impedance tube manufactured by BSWA TECH and self-made equipment are shown in Fig. 11 (a is for impedance tube manufactured by BSWA TECH, b is self-made). The results showed that the trend is also consistent, the sound absorption coefficient curve of PU-RH composites is moved to low frequency after adding RH, and the low frequency sound absorption performance of the material can be improved by adding RH, especially between 300 and 1,300 Hz, with the content increase of RH, sound absorption performance is gradually reduced at high frequency, but increased at low frequency. Therefore, the sound absorption properties of PU foam at low frequency can be improved by adding a large amount of RH. Many experiments at domestic and foreign show that it is very difficult to improve the sound absorption performance of the porous materials without increasing its thickness. In this experiment, enhance the sound absorption performance at low frequency was achieved through the addition of RH, which will be an effective way to improve the sound absorption property of PU foam. Above all, PU composites containing 5% RH can provide the most satisfactory sound absorption performance.

Effect of RH on the Characteristic Parameters of PU Foam

The values of the characteristic parameters of each sample are shown in Table 2. The flow resistance of PU foams in this set is small; it indicates that RH has a great influence on the pore size and pore size distribution of PU foam.

TABLE 2. The characteristic parameters of different PU-RH composites.

Sample	Flow resistance (rayls/m)	Porosity	Curvature	Viscous shape factor	Thermal shape factor	M'
Contrast	8,707.2	0.9126	1	0.3233	1.6237	1.0051
2%RH	7,567.7	0.8692	1.2	0.3356	0.8787	2.9295
5%RH	8,285.2	0.8733	1.9	0.3662	1.2411	1.0001
8%RH	8,607.5	0.8620	1.6	0.3761	1.2909	1.0000

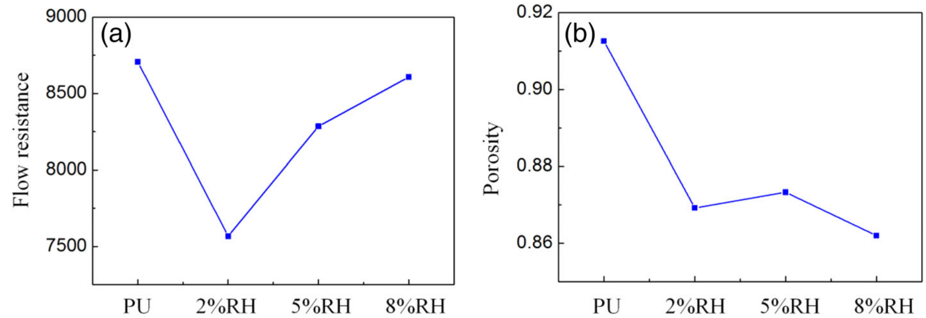

FIG. 12. Effect of RH quantity on flow resistance (a) and porosity (b) of PU foam. [Color figure can be viewed at wileyonlinelibrary.com]

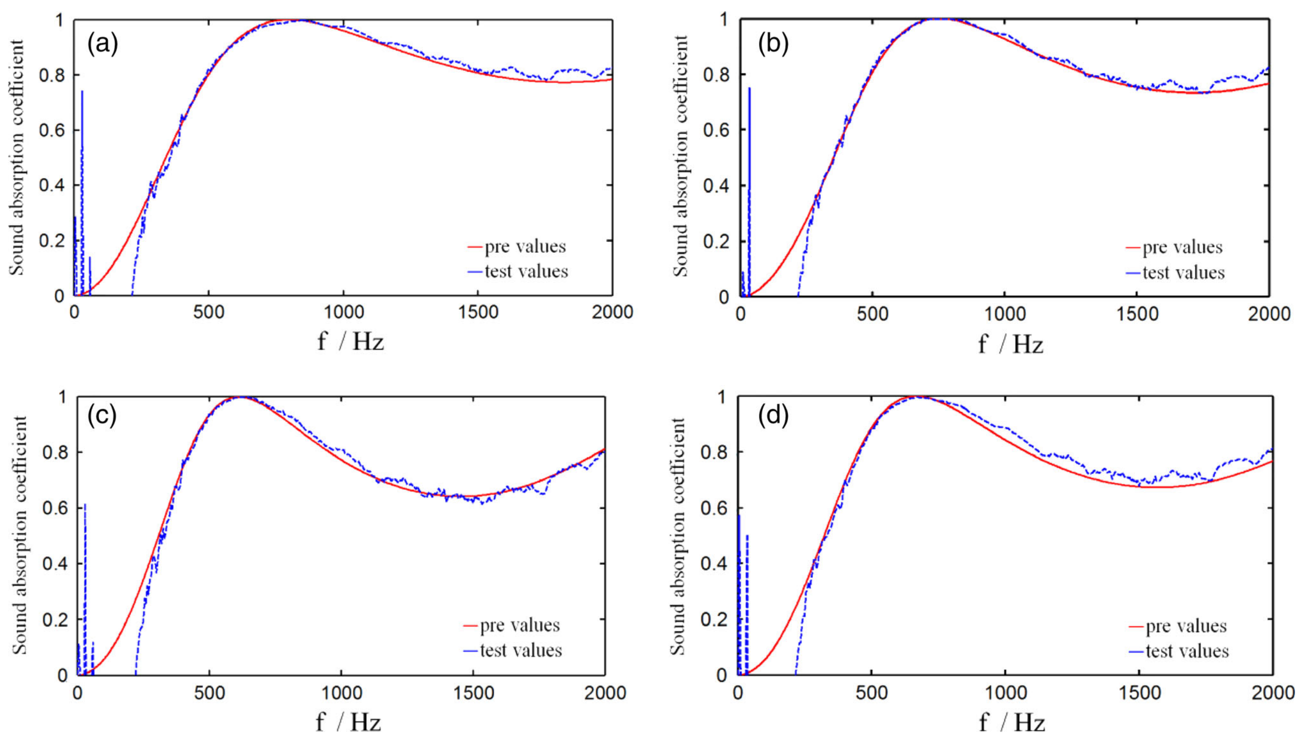

FIG. 13. Comparison of predicted and experimental values of sound absorption coefficients of different PU-RH (a is for comparison sample, b is for 2% RH, c is for 5% RH, and d is for 8% RH). [Color figure can be viewed at wileyonlinelibrary.com]

The flow resistance and porosity of the samples are shown in Fig. 12 (a is for flow resistance, b is for porosity), the results revealed that the flow resistance decreased with the increase of RH, until the minimum value when 2% RH was added. As the quantity of RH continues to increase, the flow resistance increased instead. But flow resistance with 8% RH just has a little different with PU sample. For porosity, with the increase of RH content, the porosity of the samples showed a trend of slow decline, while porosity of 5% RH sample increased slightly compared with 2% RH sample.

Effective Validation

In order to verify the accuracy of the test values, the sound absorption coefficients of the samples with different amount of RH were calculated under the condition of backed with 30 mm cavity, and compared with the test values (a is for comparison sample, b is for 2% RH, c is for 5% RH, d is for 8% RH); the results are shown in Fig. 13. The predicted values fit well with the test values, especially over the frequency of 300 Hz. It indicated that the test method is accurate and reliable, which can be used to predict the characteristic parameters of porous materials.

CONCLUSIONS

This article studied the influence of RH content on the acoustic performance of PU foam. Analyzed the impact of RH on the flow resistance and porosity of PU foam with a self-made rapid test-bed, and also compared the predicted values with experimental values of acoustic characteristic

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parameters. The results showed that adding RH is able to improve the sound absorption performance of PU foam at low frequency, and PU-RH composite with 5% RH showed the best sound absorption performance. The flow resistance of PU with 2% RH was lowest, when the content of RH increase, the resistance continues to increase, but the resistance of PU with 8% RH and without RH has little difference. In case of the porosity, with the content of RH increase, the porosity of samples generally showed the tendency of slow decline, while PU-RH containing 5% RH has a little increase compared with that of 2% RH.

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