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Sound Absorption Behavior of Polyurethane Foam Composites with Different Ethylene Propylene Diene Monomer Particles

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Considering the environmental pollution caused by waste rubber, some measures should be taken to improve the utilization rate of waste rubber. In this study, the effect of Ethylene Propylene Diene Monomer (EPDM) particles in the polyurethane (PU) foams on sound absorption behavior is investigated for improving sound environment within vehicles and reducing the environment pollution. EPDM particles of different contents and hardness are used as fillers for producing foams with different pore morphologies and sound absorption properties. The results show that adding EPDM to foam would produce smaller pores, higher density and bigger air-flow resistivity. Simultaneously, there are better sound absorption properties of the PU foam composites in the medium frequency region, and the better value can be obtained at the lower frequency with the content of EPDM increasing. The hardness of EPDM also shows better influence on sound absorption properties, especially in the medium frequency region. It means the foam pore morphologies have influence on sound absorption properties.

Keywords: polyurethane foam composites; contents of EPDM; hardness of EPDM; sound absorption coefficient.

1. Introduction

Noise pollution is a critical problem of daily life and getting worse with the development of society. The vehicle noise consists of structural noise and airborne noise, which not only affects the environment but also threatens drivers and passengers health (Sung *et al.*, 2007; Zwinselman, Laux, 1989). In order to solve this problem, the materials are being developed with high sound absorption efficiency.

Polyurethane (PU) foams have been regarded as effective acoustic package materials in automobile industry due to the effective sound damping, low density and easy production (Soto *et al.*, 2017). Study has been verified that acoustic properties of PU foams can be understood through the non-acoustic parameters such as porosity, density and air-flow resistivity (Wassilieff, 2003). Gwon *et al.* (2016a) indicated that the interconnecting open pores of foams play a crucial role in controlling not merely mechanical properties but also acoustic properties. Park *et al.* (2017a) demonstrated the relationship between cell openness and sound absorption behaviors of PU foam. The results show the PU foam with better cell openness gets greater sound absorption performances. Yao *et al.* (2016) indicated that porous materials have excellent sound absorption properties due to the irregular pore morphologies. Berardi and Iannace (2015; 2017) found both the air-flow resistivity and density have significant effect on sound absorption performances of fiber porous materials. Tao *et al.* (2016) found the average sound absorption coefficient of PU foam composites is reduced due to the large holes in the foam. However, pure PU foams show unsatisfactory sound absorption behavior in the low frequency region due to the special pore morphologies. This deficiency of pure PU foams can be modified by adding functional particles such as fibers, granular spheres and rubber particles (Saetung *et al.*, 2010).

Recently, numerous studies have been carried out to develop new materials to improve acoustic properties of PU foams (Soto *et al.*, 2017; Wassilieff, 1996). Cushman (1998) found great sound absorption properties can be obtained with additional acoustic impedance fillers to polymer. Gayathri *et al.* (2013) found the sound absorption coefficient of PU foam is increased at higher content of the fillers employed. Ekici *et al.* (2012) found the sound absorption properties of PU foam have a significant improvement with addition of tea-leaf fibers. Çelebi, Küçük (2012) and Chen, Jiang (2018) found that adding natural fibers of different contents or sizes to PU foams resulted in significant impacts on sound absorption performances. Bahrambeygi *et al.* (2013) observed that both nanoparticles and multi-wall carbon nanotubes gave rise to the considerable improvement on sound absorption behavior of PU foams. Hong *et al.* (2007) found that PU foams with addition of recycled rubber particles had excellent acoustic performances. Zhao *et al.* (2010) and Yang *et al.* (2004) found that wood-waste tire rubber or straw-waste tire composite panel possesses better soundproof effects. Saetung *et al.* (2010) observed that PU foams filled with hydroxytelechelic natural rubber of different molecular weights or epoxide levels exhibited excellent acoustic absorption characteristics. They also studied the physic-mechanical and biological properties of the obtained polyisoprene based PU materials. Gayathri *et al.* (2013) and Maderuelo-Sanz *et al.* (2013) also investigated the sound absorption performances of PU foams filled with recycled rubber.

At present, waste rubber causes black pollution that is more difficult to handle than white pollution. The direct burning of waste rubber not only pollutes the environment but also wastes rubber resources. However, recent researches on rubber particles show that rubber can be employed as filler for noise absorption improvement. As far as we know, there are no studies about the acoustic performances of PU foams filled with Ethylene Propylene Diene Monomer (EPDM) of different contents or hardness which is always used as sound insulation material. In this study, EPDM of different contents or hardness are added to PU foams separately to investigate the acoustic properties through the non-acoustic parameters. The aim of present study is to develop the PU foam composites with high sound absorption efficiency and to improve the utilization rate of waste EPDM. Morphological measurements for pore size and interconnection are implemented by scanning electron microscope and image analysis software. Sound absorption coefficient is also tested by an impedance tube in order to understand the relationship between the pore morphology and the acoustic absorbing performance (Gwon *et al.*, 2016b).

2. Experiment

2.1. Materials

The polyether polyols include 330N (OH-value: 33–36 mg KOH/g) and 3630 (OH-value: 33–37 mg KOH/g). Modified isocyanate (MDI, diphenylmethane 4,4-diisocyanate) is used as matrix material to synthesize PU foam. Catalyst A1 (mixture of 70% 2-dimethylaminoethyl ether and 30% dipropylene glycol) and catalyst A33 (solution of 33% triethylenediamine) are both chosen as the amine catalysts for gelling reaction. Silicone oil is used as surfactant. Triethanolamine (TEA) is also employed as a catalyst for gelling reaction. Deionized water is used as blowing agents to produce CO_2 gases and amine functionalities. The EPDM of the same size is purchased directly from the seller and it has three different hardness levels: 65 HA, 70 HA and 85 HA. It is cylinder with a diameter of 4 mm and a thickness of 3 mm.

2.2. Sample preparation

PU foam can be synthesized by one step polymerization, semi-prepolymer and prepolymer processes. The one step polymerization process is selected to prepare the PU foam in this paper because it is the simplest method than others. The formulation of pure PU foam is shown in Table 1. The proportion of each material is compared with the total mass of the polyols. For preparing PU foam composites, the 65 HA EPDM is weighed 2 g, 4 g, 6 g, 8 g and the others are weighed 4 g, respectively. The materials except for MDI and EPDM are weighed in a paper cup and pre-mixed at 2000 rpm for 2 min using a mechanical mixer equipped with 2 impellers. Then the various EPDM are added to the mixtures respectively and stirred for 2 min to mix it uniformly. After that, MDI is added to this mixture. The whole components are stirred again for 15 seconds, and then the mixture is poured rapidly into the mold before foaming occurred. The curing process is performed in a drying oven for 30 min at 50°C. Then the foams are removed from the mold and saved at room temperature for 24 h before being cut for testing.

Table 1. Pure PU foam formulation.

Raw material	Content [g]
Polyols (330N, 3630)	330 N = 60, 3630 = 40
Modified isocyanate (MDI)	30
Amine catalysts (A1, A33)	A1 = 0.05, A33 = 1.0
Catalyst (TEA)	3
Surfactant (silicone oil)	1.8
Blowing agent (deionized water)	3

2.3. Characterizations

2.3.1. Pore morphologies

A scanning electron microscope (SEM, ZEISS EVO18) is used to obtain pore morphologies (pore size and interconnection) at 10 kV. The samples are cut from the middle of the foams with a sharp knife, and then the cross-sections are sputter coated with gold before scanning. In order to ensure the normal operation of the SEM, the vacuum degree in the lens barrel is maintained at $1.33 \times 10^{-3} \sim 1.33 \times 10^{-2}$ Pa. The SEM images are used to analyze the average pore size and interconnection.

2.3.2. Air-flow resistivity

The air-flow resistivity of foam is defined as a resistance for per unit thickness. Thus, the resistance of different foams to fluids can be directly compared without considering the effects of thickness (Sung, Kim, 2017). Its unit is $\mathrm{Pa \cdot s/m^2}$, expression as following:

$$r_0 = \frac{S \cdot \Delta P}{U \cdot T}, \tag{1}$$

where ΔP is the air pressure difference across the sample with respect to the atmosphere, Pa, S is the cross-section area of the test sample, $\mathrm{m^2}$, U is the velocity of air flow through the sample, $\mathrm{m^3/s}$, T is the thickness of the test sample, m.

The continuous flow process is used to measure the air-flow resistivity of foams. It meets the requirements of the standard ASTM C522-03 (2009). Five measurements are performed for each sample, and then the mean air-flow resistivity is obtained.

2.3.3. Density

Cylindrical test samples (100 mm in diameter and 35 mm in thickness) are cut from the middle of the foams in perpendicular direction to the foam growth with a hot wire cutter. The sample is weighed, and then the density is calculated to the given specimen volume.

2.3.4. Acoustic property

The sound absorption coefficient is defined as the ratio of the acoustic energy absorbed by a sample to the incident acoustic energy as a function of frequency (Gwon et al., 2016b). Meanwhile, the average sound absorption coefficient is widely used to evaluate the sound absorption ability in engineering practice. It is calculated with Eq. (2)

$$\alpha_a = \frac{\alpha_{125} + \alpha_{250} + \alpha_{500} + \alpha_{1000} + \alpha_{2000} + \alpha_{4000}}{6}, \tag{2}$$

where α_a is the average sound absorption coefficient. $\alpha_{125} \sim \alpha_{4000}$ represent the sound absorption coefficients at 125 Hz, 250 Hz, 500 Hz, 1000 Hz, 2000 Hz, and 4000 Hz, respectively.

Sound absorption coefficient test is performed with a two-microphone impedance tube device SCS90AT. It is based on transfer function method in accordance to ISO 10534-2 (1998) and ASTM E1050-12 (2012) international standards. Cylindrical 35 mm thickness samples with 100 mm and 28 mm in diameters are tested for the frequency ranges of 100–1500 Hz and 500–6300 Hz, respectively (Al-Rahman et al., 2012; SCS90AT Manual, 2013).

3. Results and discussions

3.1. Pore morphologies and characteristics

The acoustic performances of PU foam composites are closely related to the pore characteristics such as pore size and interconnection. Study shows the pores isolated with other adjacent pores also called closed pores which allow some level of sound absorption, but only open pores guarantee a continuous tunnel with the external surface of the material which allows higher sound absorption properties (Arenas, Crocker, 2010). When sound waves enter the pores of the material, the cell walls provide the sound with multi-refraction which increases the collision frequencies of sound waves with air and solid walls. Thus, the sound energy will be more transformed to heat energy to dissipated (Huang et al., 2015). Thus, the sound absorption materials should have more pores to allow the sound enter the material and for dissipation. Therefore, morphologies analysis for the pores is the first concern to understand the sound absorption capacity of PU foam composites (Gwon et al., 2016a). EPDM particles of different contents or hardness are used to investigate the diverse pore structures of PU foam composites.

3.1.1. Content effects

Figure 1 shows foam pore morphologies with addition of EPDM of five types of contents. As seen in the figures that almost all the pores of PU foam composites are connected by the edges or cell walls, and the pores are smaller when the contents of EPDM are increased. The edge is formed between the interconnecting pores of the foam which thickness is closely related to the density and the stiffness of the cell walls. However, the edge thickness of the foam is different when the content of EPDM is changed. As shown in Fig. 1c and 1d, there is an obvious deformation of pore morphology near to the EPDM particles, and pore interconnection decreased with the increasing contents of EPDM. However, the pores are more uniform and tight when the contents are increased. It is possibly attributed to that EPDM is not reacting with the reactants and restricting the gelling reaction.

In a certain range, the effects of foam density and average pore size on the sound absorption performances of foams are contradictory. The foam with large density and small pores has good sound absorp-

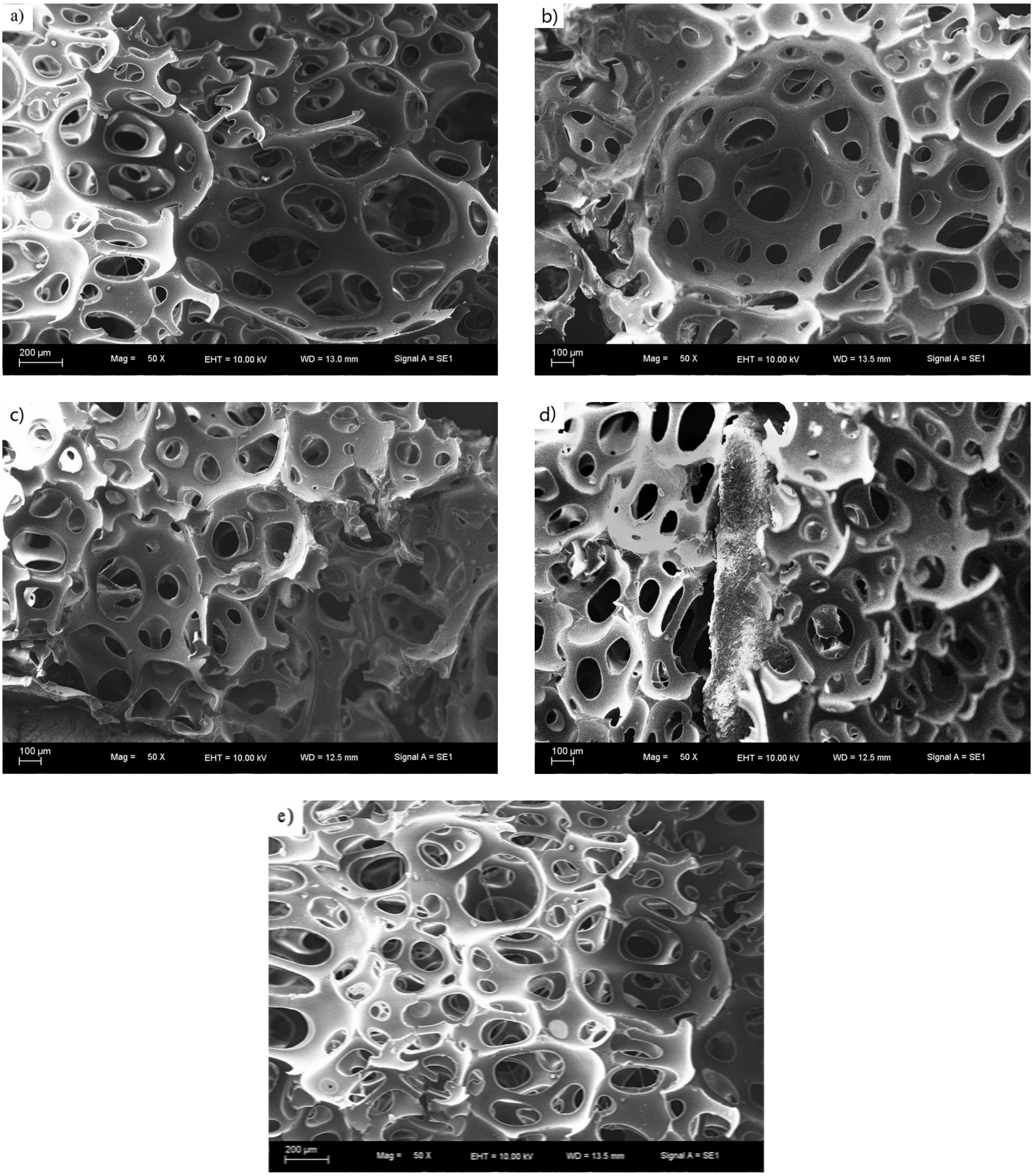

Fig. 1. SEM images of PU foams filled with EPDM of different contents under magnification 50: a) PU foam filled with 2 g EPDM, b) PU foam filled with 4 g EPDM, c) PU foam filled with 6 g EPDM, d) PU foam filled with 8 g EPDM, e) pure PU foam.

tion performances. It can be observed in Fig. 2 that the foam density is enhanced when the contents of EPDM is increased. The PU foam filled with 8 g EPDM gets the maximum density, and the pure PU foam gets the smallest value. This may be the high content of additives fill up more voids inside the foams, and the density of EPDM is higher than pure PU foam. Figure 3 shows the average pore size of the foams with varying contents of EPDM. The average pore size is obtained from 10 different pores for each SEM image except

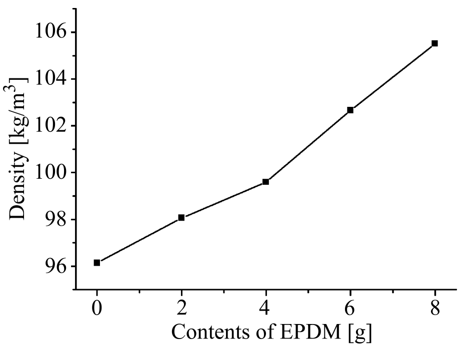

Fig. 2. Changes in foam density with addition of EPDM of different contents.

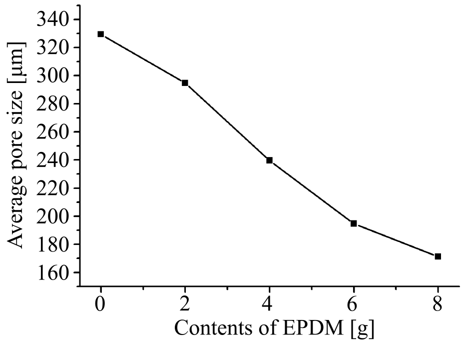

Fig. 3. Average pore size of PU foams which filled with EPDM of different contents.

some special pores. The average pore size shows significant decrease with the contents of EPDM increasing. The smallest value is 172 μm when filled with 8 g EPDM which is almost the half of the pure PU foam. As shown in Fig. 1, it can be explained that EPDM is not a reactant, and it is hindering the reactants connecting with each other.

The air-flow resistivity is also used to assess the acoustic performances. A low air-flow resistivity indicates little resistance to air streaming through the material, whereas a too high value indicates that the air streaming is obstructed and lead to low sound absorption properties in the low frequency region (Sung, Kim, 2017; Park *et al.*, 2017). Figure 4 is the results about air-flow resistivity of PU foams filled with EPDM of different contents. It can be seen that the curve gets raised with the content of EPDM increasing. PU foam filled with 8 g EPDM gets the maximum air-flow resistivity which would increase the resistance for sound waves propagating in the materials. However, the air-flow resistivity is related to surface impedance of material. The great sound absorption properties occur when surface impedance of material is equal to air impedance. This means that low and high air-flow resistivity can lead to low sound absorption performances (Jian *et al.*, 2006). It also can be observed through Berardi and Iannace (2015; 2017) study that the air-flow resistivity of the dense kenaf is bigger than other fibers. However, the sound absorption coefficient is not the best especially in the medium frequency. Therefore, the sound absorption properties of the foams would be also influenced with the contents of EPDM.

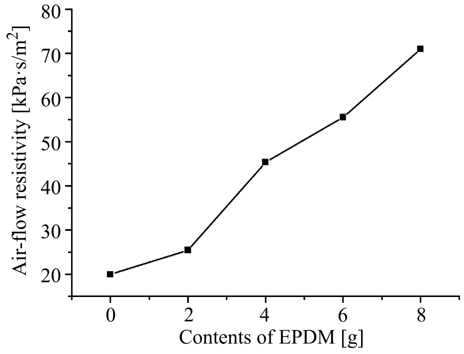

Fig. 4. Air-flow Resistivity of PU foams which filled with EPDM of different contents.

3.1.2. Hardness effects

Figure 5 shows foam pore morphologies with addition of EPDM of four kinds of hardness. Figure 5a represents pure PU foam, because the hardness of EPDM is 0. All the images show the PU foam composites are composed of great deal of pores. It can be seen that pore size and interconnection have clearly distinction when EPDM of different hardness is added into the foam, especially in Fig. 5c and 5d. As shown in Fig. 5c, the pores are smaller, and it is uniformly distributed in the foam. It may be explained that the hard EPDM is difficult to compress during the nucleation process, and the pores interconnection is blocked between the cells. In other words, the same foam produced in a common space or in some separated spaces would get different pore sizes and interconnection ratio.

Figure 6 shows the foam density changing together with the hardness of EPDM. It is pure PU foam when the hardness of EPDM is 0 HA. It can be seen that the curve in 0–70 HA gets raised. However, in the 70–85 HA range, the curve gets decreased. The foam density gets maximum value when the hardness of EPDM is 70 HA, and the smallest value is obtained for the pure PU foam. It may be explained by the higher density of EPDM compared to foam. Figure 7 shows the change tendency of the average pore size with different PU foam composites. It is obvious that the minimum average pore size is obtained when the hardness of EPDM is 70 HA, which would be benefit for sound absorption. It can be explained that pores expansion is blocked by the hard EPDM.

Figure 8 shows the air-flow resistivity of PU foams which filled with EPDM of different hardness. It is obvious that the air-flow resistivity grows significantly when the hardness is increased. It gets the largest value when the hardness is 85 HA. Meanwhile, it can be ob-

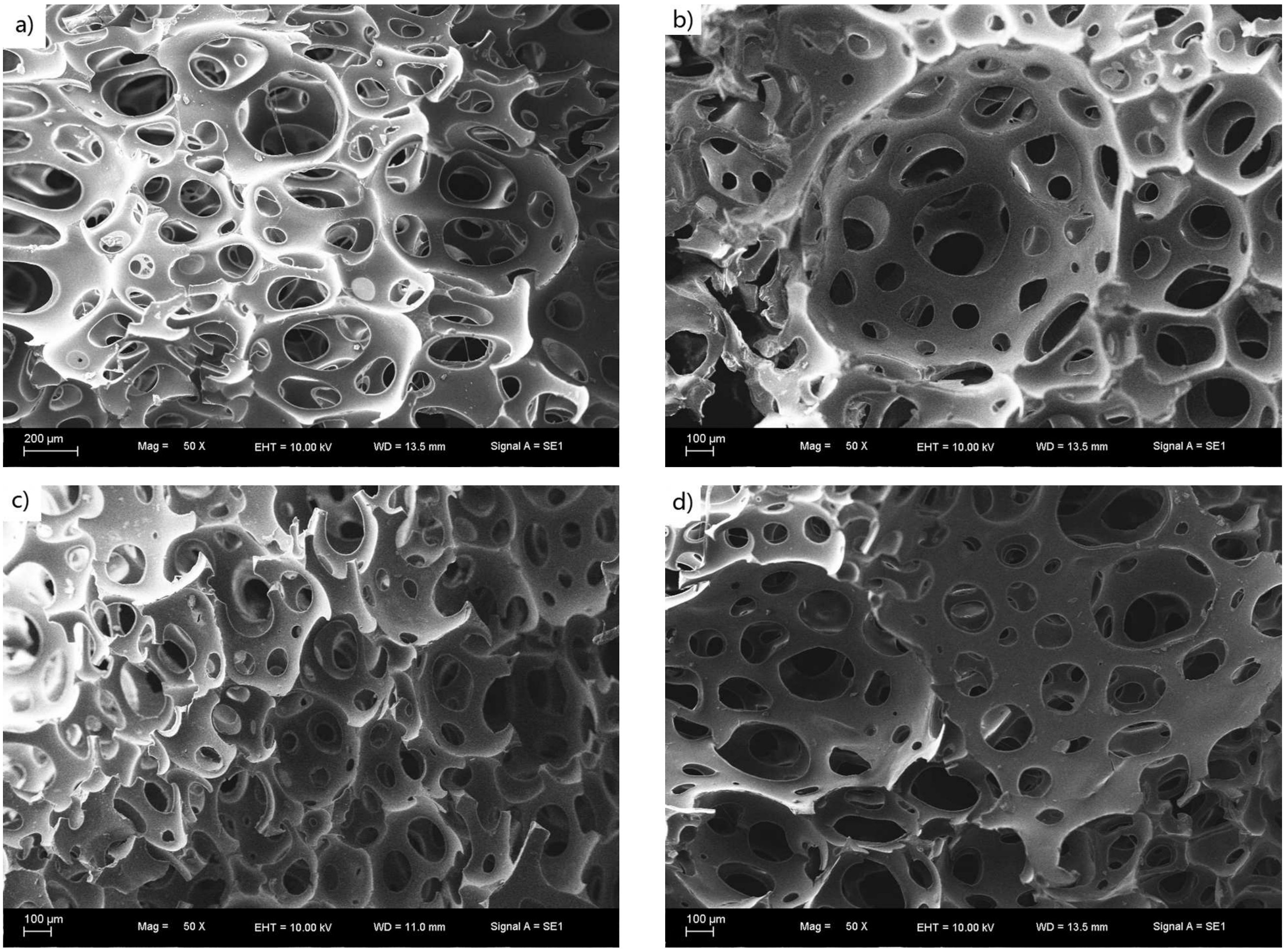

Fig. 5. SEM images of PU foams filled with EPDM of different hardness under magnification 50: a) hardness of EPDM is 0 HA, b) hardness of EPDM is 65 HA, c) hardness of EPDM is 70 HA, d) hardness of EPDM is 85 HA.

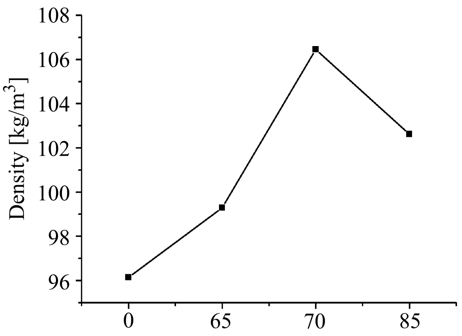

Fig. 6. Changes in foam density with addition of EPDM of different hardness.

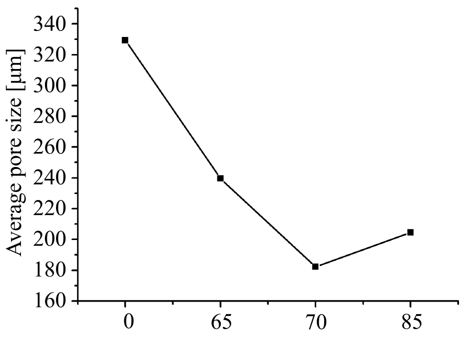

Fig. 7. Average pore size of PU foams which filled with EPDM of different hardness.

tained that the growth ratio of air-flow resistivity is larger in the range from 65 to 70 HA than in the other ranges. This may be the air-flow resistivity is influenced by the density and pore size. In Fig. 6 and Fig. 7, PU foam filled with 70 HA EPDM possesses high density, small pore size and numerous uniform pores which are advantageous in enhancing sound absorption performances of the foams.

3.2. Sound absorption property

The sound absorption coefficient curves of the PU foam composites with EPDM of various contents or hardness are shown in Fig. 9. Generally, the sound absorption capacity of porous materials is related to the interior pore size and interconnection. The sound waves lead to the vibration of cell walls and air inside

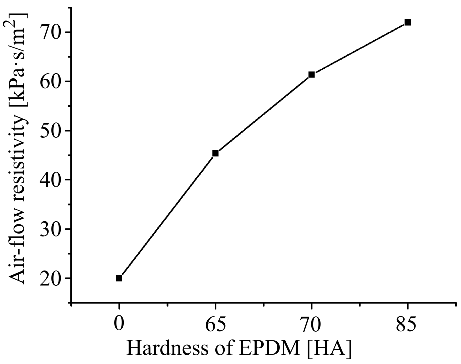

Fig. 8. Air-flow Resistivity of PU foams which filled with EPDM of different hardness.

pores. Then, the sound energy dissipates or converts to the heat through vibration damping of the cell walls and viscidity damping of the air. Meanwhile, the cell walls are forced to stretch and bend due to the pressure and disturbance of sound waves, which would make the sound energy converted into kinetic energy (Lee *et al.*, 2012). Therefore, the sound absorption properties can be improved by changing the sound damping which are related to the interior pore morphologies. As Subsec. 3.1 has discussed, the sound absorption performances of PU foam composites change together with the foam density, pore morphologies and air-flow resistivity which are influenced by the additives.

Figure 9a shows the sound absorption coefficient of the five different PU foam composites. It is obvious that the sound absorption coefficient is impacted by the contents of EPDM. It may be the pore morphologies are changed with additional EPDM of different contents. It can be found that at the frequency range of 100–225 Hz, the sound absorption coefficient of all the PU foam composites is smaller than pure PU foam. Meanwhile, the sound absorption coefficient of the foam filled with 8 g EPDM exceeds the pure PU foam at the frequency about 225 Hz, and the foam filled with 2 g EPDM exceeds the pure PU foam at the frequency about 450 Hz. It means the better sound absorption properties are obtained at the lower frequency with the content of EPDM increased. It also can be found that all the curves of the foam composites are different to pure PU foam, and the values are better in the medium frequency region. It may be as Subsec. 3.1.1 discussed, the pure PU foam gets smallest air-flow resistivity and the biggest average pore size in this paper which leads to the sound waves propagating more easily with less energy dissipated.

Figure 9b shows the sound absorption coefficient of PU foams which filled with EPDM of different hardness. It can be found that the sound absorption coefficient curves of the foams have different growth tendency. The PU foam filled with 85 HA EPDM gets bigger sound absorption coefficient than pure PU foam in the frequency range of 160–2500 Hz. It may be the foam filled 85 HA EPDM gets smaller average pore size, higher density and larger air-flow resistivity than

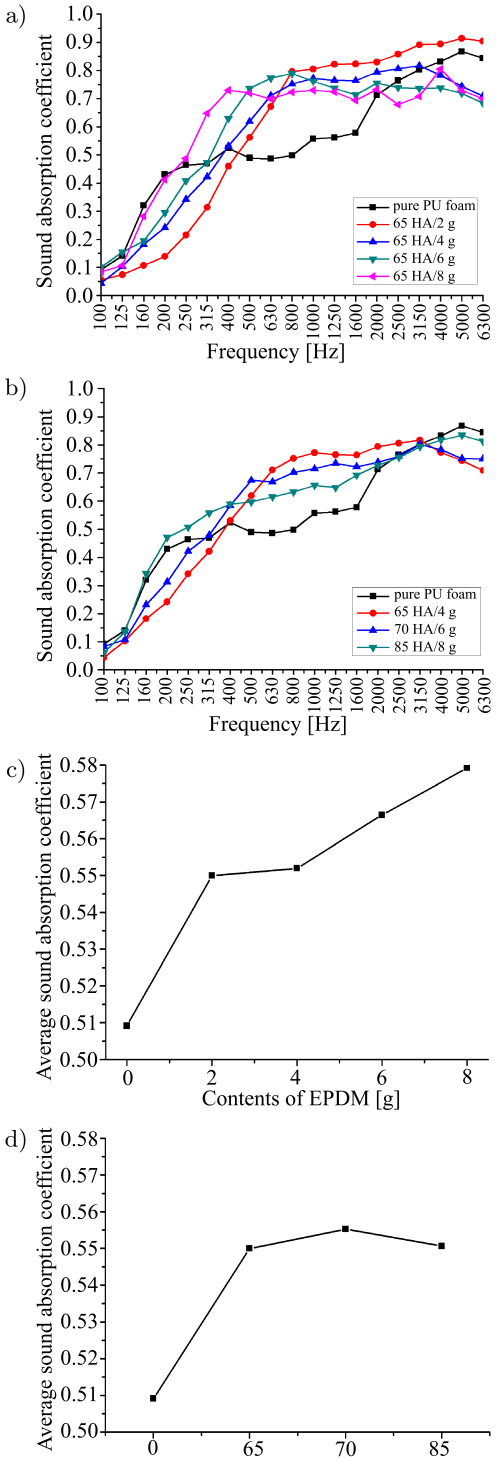

Fig. 9. Sound absorption properties curves of PU foams. (a) sound absorption coefficient of PU foams filled with EPDM of different contents. (b) sound absorption coefficient of PU foams filled with EPDM of different hardness. (c) average sound absorption coefficient of PU foams filled with EPDM of different contents. (d) average sound absorption coefficient of PU foams filled with EPDM of different hardness.

pure PU foam, which would increase the collision frequencies of sound waves with solid walls to dissipate more sound energy. Simultaneously, as Subsec. 3.1.2 discussed that the growth ratio of air-flow resistivity is bigger, and the value is little bigger than the foam filled 65 HA when the hardness of EPDM is 70 HA. It may be the reason that the curves growth trend is similar, and the sound absorption properties of the foam filled 70 HA EPDM gets better frequency bandwidth in the medium frequency region. Figure 9b also indicates the sound absorption properties of the foam composites in the medium frequency region are better than the pure PU foam. Meanwhile, the better sound absorption performances can be obtained at lower frequency with the hardness of EPDM increased.

Figures 9c and 9d show the average sound absorption coefficient of the PU foam filled with EPDM of different contents and hardness. It can be seen that all the average sound absorption coefficients of the foam composites are better than pure PU foam. Figure 9c shows the maximum value is obtained with addition of 8 g EPDM to PU foam. Figure 9d shows the maximum value is obtained when the hardness of EPDM is 70 HA. As Subsec. 3.1 discussed, both the foam composites get the biggest density and smallest average pore size. It means the foam with large density and small pores has good sound absorption performances. These results agree with similar investigation reported by Berardi and Iannace (2015; 2017). Meanwhile, the foam filled with 8 g EPDM gets the maximum air-flow resistivity which increases the resistance to sound waves propagating through the material. Thus, the sound absorption performances of the material are increased. However, as Subsec. 3.1.2 mentioned, the biggest air-flow resistivity is obtained when the PU foam filled with 85 HA EPDM. It may be the average pore size and density is rebounded in the hardness range from 70–85 HA.

4. Conclusions

In this paper, PU foams filled with EPDM of different contents (2 g, 4 g, 6 g, 8 g) and different hardness (65 HA, 70 HA, 85 HA) are prepared by one step polymerization process. The pore morphologies are affected by the EPDM as studied from the SEM images and the curves about the average pore size. The foam density and air-flow resistivity are also impacted by filling different EPDM. Meanwhile, the average sound absorption coefficient is changed with the contents and hardness of EPDM changing. The results also indicate the better sound absorption properties of foams are obtained when there are smaller pore sizes, higher foam density and air-flow resistivity. It means the sound absorption properties of PU foam composites are closely related to pore morphologies of the foams. On the other hand, the sound absorption properties of PU foam composites get better in the medium frequency region, and the better sound absorption properties are obtained at the lower frequency with the content of EPDM increased. The hardness of EPDM also shows same influence on the sound absorption properties. Because PU foam filled with EPDM of suitable contents or hardness can get smaller pores and higher air-flow resistivity. Simultaneously, EPDM is a kind of sound insulation material that can reflect sound waves which would increase the collision frequencies of sound waves with air and solid walls in the foams to dissipate more sound energy. Therefore, EPDM can be added to PU foam to improve the sound absorption performances of the PU foams, but also reduce the environment pollution caused by waste EPDM.

Acknowledgments

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