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Acoustic performance of flexible polyurethane composite foams filled with melamine particles

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Abstract−PU foams are widely used in the automobile industry not only for their lightweight properties, but also as sound-absorbing and seat cushioning materials. The effects of particles on the sound absorption properties of PU composite foams were investigated. PU foams containing 3 wt% melamine particles showed excellent sound absorption properties. However, when the particle content exceeded 3 wt%, particle agglomeration occurred, leading to reduced sound absorption. A dispersant was added during the fabrication process to improve the interfacial compatibility between the PU matrix and melamine particles, resulting in a well-developed cavity and pore structure. The sound absorption characteristics of the PU composite foams were significantly improved in the frequency range below 2,000 Hz. Our study highlights the importance of adjusting the concentration of particles and optimizing the cavity and pore structure of the material for achieving better sound absorption properties in PU composite foams.

Keywords: Polyurethane Foam, Composites, Dispersant, Melamine Particles, Morphology, Sound Absorption

INTRODUCTION

Automobile noise can have a significant impact on passenger comfort during driving [1,2]. Consequently, the automobile industry has conducted extensive research on various sound-absorbing materials to reduce noise. Noise generated within automobiles can be classified as either structure-borne noise (30-500 Hz) or airborne noise (500-8,000 Hz), based on its transmission path [3,4]. Porous materials, including polyurethane (PU) foam, glass wool, glass fiber felts, and mineral fiber composites, are widely used for noise absorption in the automobile industry [5-7]. Among these, PU foams are popular as sound-absorbing materials for the interior of automobiles due to their light weight, moldability, and high sound absorption [8,9].

In PU foam sound absorption, air molecules vibrate and rub against the pore wall, converting sound energy into heat energy through resonance [10-13]. Therefore, the sound absorption properties of PU foams are closely related to their cellular morphology. The sound absorption performance of PU foams can be improved by adjusting their formulation, such as changing the concentration of polyol, catalyst, crosslinker, water, isocyanate, and surfactant [2,3,9,11-14]. Additionally, the addition of various particles can greatly impact the cavity size and pore types in PU foams due to changes in the interfacial compatibility between the particle and PU matrix [15-20]. While melamine particles are widely used as flame retardants in PU foam [21,22], few studies have investigated their effects on sound absorption. Dispersants are used to evenly disperse solid powders in polyol.

In this study, we report on the effects of melamine particles and dispersants on the sound absorption and morphology of PU foams. First, we examined the sound absorption coefficient of PU foam by varying the concentration of melamine particles. We added a constant amount of dispersant and changed the concentration of melamine particles to determine the optimal amount for the highest sound absorption coefficient. We investigated the effects of melamine particles and dispersant on the reaction rate using Fourier-transform infrared spectroscopy (FTIR) and analyzed the cellular morphology of PU foams, including cavity and pore size and pore types, using scanning electron microscopy (SEM). Finally, we investigated the sound absorption coefficient, acoustic activity, and noise reduction coefficient of the PU foams using an impedance tube.

EXPERIMENTAL

1. Materials

Polyether polyol with the trade name of PPG-6000 manufactured by Kumho Petrochemical was used as a starting material for the synthesis of PU foams. PPG-6000 has an OH value of 28±2 and a molecular weight of 6,000 g/mol with an average functionality of 3. The aromatic isocyanate used was KW 5029/1C-B from BASF, which has an %NCO of 35±0.5 and a viscosity of 290 cps at 25 °C, containing 78% of 4,4′-methylene bis(phenyl isocyanate), 5% of benzene 1,1 methylene bis(4-isocyanato) homopolymer, and 17% of toluene diisocyanate. The PU foaming reaction was catalyzed by DABCO 33LV, which is a blend of 33% triethylenediamine and 67% dipropylene glycol from Air Products and Chemicals (USA). The blowing catalyst used was DABCO BL11, which is an acid-blocked version of 70% bis(2-dimethylaminoethyl) ether diluted with 30% dipropylene glycol, also from Air Products and Chemicals. De-ionized water was used as a blowing agent. For cross-link-

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ing, diethanolamine with a molecular weight of 105.14 g/mol from Sigma-Aldrich in the USA was used. The silicone surfactant used to stabilize the cell distribution during the foaming reactions was L-3002 from Momentive (USA). To fabricate PU composite foams, melamine particles with a trade name of M2659 and a size of 430± 15 μm from Sigma-Aldrich in the USA were used. PU PE-40 dispersant from Evonik in Germany was applied to achieve homogeneous distribution of the melamine particles in the polyol mixture during the pre-mixing stage.

2. Synthesis

Flexible PU foams were synthesized using a two-component system foaming process. First, the polyol system, which included polyol, gelling and blowing catalyst, crosslinker, deionized water, silicone surfactant, dispersant, and melamine particles, was weighed and stirred at 1,700 rpm for 10 min in a 1 L paper cup. The particle content ranged from 0 wt% to 5 wt%, and dispersant was also used in formulations. Afterward, the pre-weighed isocyanate was added to the polyol system and stirred at 6,000 rpm for 8 sec. The detailed formulations for fabricating the PU foams are presented in Table 1. The PU mixtures were promptly poured into an aluminum mold with dimensions of 200 mm×200 mm×50 mm and cured at 60 °C for 20 min. Finally, the foams were removed from the mold and stored at room temperature under a relative humidity of 50±10% for three days before cutting the foams. All foams were prepared after removing 10 mm of the surface skin.

3. Sample Characterization

3-1. Fourier-transform Infrared Spectroscopy (FTIR)

To analyze the reaction rates of the addition of dispersant and melamine particles, we utilized a Fourier transform infrared spectrometer (FTIR Frontier, PerkinElmer Inc.) equipped with an attenuated total reflectance (ATR) accessory. This allowed us to compare NCO conversion during the polymerization process. We obtained FTIR spectra from four scans at a resolution of 4 cm^{-1}, and analyzed the functional group conversion using time-based software. The reaction mixtures were scanned using the ATR accessory with an 8 sec time interval for 60 min, during which the samples were placed on the diamond crystal. After baseline corrections, the NCO conversion was calculated using Eq. (1):

$$\text{NCO conversion} = 1 - \frac{I_t}{I_0}, \quad (1)$$

where I_0 and I_t are normalized peak heights for the NCO absorption band at the beginning of the reaction and time t [23]. The free NCO stretching band at 2,272 cm^{-1} was normalized by the intensity at 2,970 cm^{-1} of the internal standard (CH stretching band), which shows invariable peak intensity during the polymerization [24-26].

Table 1. Formulation details for manufacturing the PU composite foams including melamine particles without and with dispersant

	Material	Content (g)
Polyol system	Polyol (PPG-6000)	100
	Gelling catalyst (33LV)	0.72
	Blowing catalyst (BL11)	0.08
	Cross-linker (DEA)	0.60
	Blowing agent (H_2O)	4.00
	Surfactant (L-3002)	1.32
	Melamine particles (M2659)	0.00 1.00 2.00 3.00 4.00 5.00
	Dispersant (PE40)	0.00, 1.00
Isocyanate* (KW 5029/1C-B)		61.38

* NCO Index: 1.0

3-2. Morphology

We used a scanning electron microscope (SEM, SNE-3000M, SEC Co. Ltd., at 15 kV) to examine the cell morphology of the PU matrix, including cavities and interconnecting pores, as well as the distribution of melamine particles. Before imaging, the SEM samples were coated with a thin layer of gold sputter (MCM 100, SEC Co. Ltd.). The images were analyzed using Image-Pro Plus software (Media Cybernetics) to determine cavity and pore sizes, as well as the ratios of different pore types (open, partially open, closed). For each sample, we analyzed 15 images.

3-3. Sound Absorption Property

We utilized two impedance tubes (SW420 and SW470, BSWA) equipped with two 1/4-inch microphones (MPA416, BSWA) to measure the sound absorption coefficients of the PU composite foams. For these measurements, all samples had a thickness of 20 mm, while sample diameters of 30 mm and 100 mm were used for high (1,000-6,300 Hz) and low frequency (63-1,600 Hz) measurements, respectively. To obtain a single range plot, we combined the sound absorption coefficients from low, mid, and high frequencies using VA-Lab software (BSWA).

RESULTS AND DISCUSSION

1. Morphology

The sound absorption properties of flexible polyurethane foams with a porous structure are closely linked to their morphology, which affects the interaction of sound waves with cavities and pores, leading to the dissipation of energy as thermal energy. To improve the compatibility between the PU matrix and particle, we employed a dispersant and investigated its effect on the PU reaction kinetics. Fig. 1 displays the NCO conversion of PU composite foams, including 0 wt% and 3 wt% melamine particles, both with and without dispersant. The conversion rate was higher at 0 wt% melamine particles since the addition of melamine particles increased initial viscosity and hindered the drainage flow, resulting in a less smooth PU forming reaction [26]. To mitigate the decrease in NCO conversion rate due to melamine particle addition, we compared the reaction rates of PU composite foams with and without dispersant. The dis-

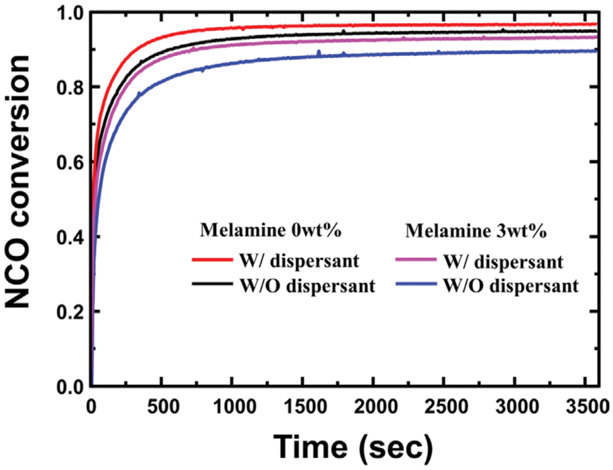

Fig. 1. Initial NCO conversion of the PU composite foams including melamine particles without and with dispersant.

persant improves dispersity via a three-step process, in which it adheres to the surface of melamine particles to enhance wetting and compatibility with the polyol system (step 1), generates friction on the particle surface to disperse the particles (step 2), and prevents re-agglomeration of particles by generating static electricity or steric hindrance on the surface of the particles (step 3) [27,28]. The NCO conversion rate of the PU composite foams containing 0 wt% and 3 wt% melamine particles with dispersant was higher than that without dispersant because the dispersant contains non-reactive amines that adsorb onto the particles, improving dispersion stability and decreasing viscosity to increase drainage flow, thus increasing NCO conversion rate. The formation of cavities and pores during the initial reaction rate is influenced not only by the PU matrix but also by CO_2 gas movements [9,11]. The reaction rate of the foam varies based on the addition of the dispersant and melamine particles (Fig. 1), and it further results in different foam morphology.

SEM images of the PU composite foams with varying contents of melamine particles (0, 3, and 5 wt%) are presented in Fig. 2(a), both without and with the use of a dispersant. To obtain average cavity and pore sizes, we utilized Image-pro Plus software for analysis, and the resulting measurements are displayed in Fig. 2(b) and 2(c). The cavity size of the PU composite foams decreased as the amount of melamine particles increased, consistent with previous studies [19,29,30]. Particles can serve as nucleating agents that initiate cavity formation, with an increase in the number of nucleation sites leading to more small-sized cavities. In addition, increasing particle content also raises the viscosity of the system, making it harder for the PU matrix to flow and inhibiting cavity growth. Consequently, the average cavity size decreases with increasing particle content. At 3 wt% particle content, however, the decreasing tendency is reversed, likely due to particle agglomeration. Aggregated particles can reduce the available nucleation sites, thereby increasing the cavity size. High particle content may have a negative impact on morphol-

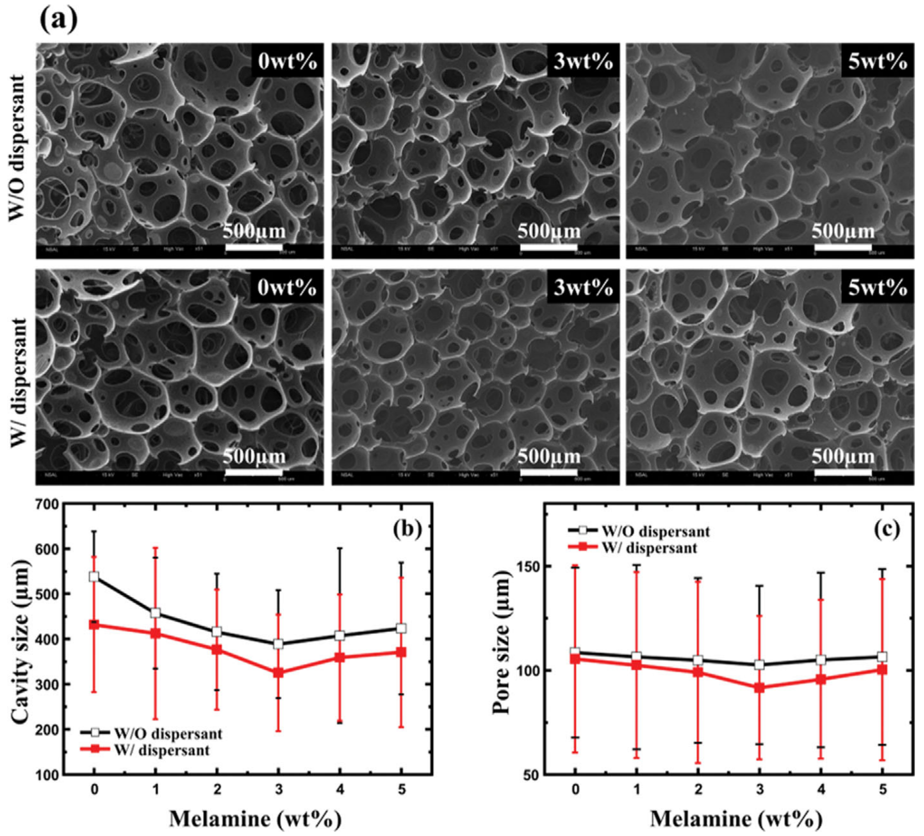

Fig. 2. (a) Typical SEM images of the PU composite foams including melamine particles without and with dispersant and their average cavity (b) and pore (c) sizes.

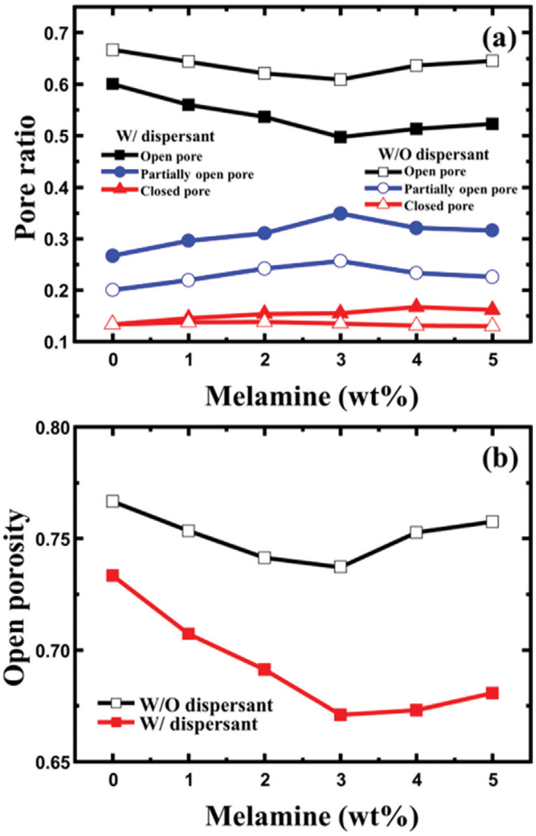

Fig. 3. **(a) Ratios of open, partially open, and closed pores of the PU composite foams including melamine particles without and with dispersant and (b) their open porosities.**

ogy development because of the agglomeration phenomena [31]. Furthermore, the cavity and pore sizes of the PU composite foams with melamine particles and dispersant were smaller compared to those without dispersant. The dispersant reduces the viscosity of the system, thereby accelerating the urethane formation reaction between isocyanate and polyol. This reaction strengthens the cavity strut, suppressing cavity growth.

Fig. 3(a) presents the relative ratios of different pore types (open, partially open, and closed) in the PU composite foams filled with melamine particles, with and without a dispersant. The ratios of open and partially open pores exhibited significant differences depending on the particle content and the presence of a dispersant. In comparison to the non-particle case, the addition of melamine particles decreased the open pore ratio while increasing the partially open pore ratio, likely due to the increased viscosity of the PU matrix and reduced drainage flow [3,16,32], which delayed pore opening. However, after the 3 wt% particle content, the trend is reversed, with aggregated melamine particles poorly compatible with the PU matrix (see Fig. 4(a)), leading to cell collapse and increasing open pore formation during CO_2 gas expansion. Furthermore, the addition of a dispersant further reduced the open pore ratio and increased the partially open pore ratio compared to the non-dispersant case. This can be attributed to the increased compatibility between the PU matrix and the surface of the particles. Additionally, the inclusion of the dispersant increased the NCO conversion rate (as shown in Fig. 1), causing the NCO group to react faster and resulting in stronger cell walls, leading to an increase in the partially open and closed pore ratios. However, the opposite trend is observed at 3 wt% particle content due to the low amount of dispersant relative to the particle content. Therefore, to achieve a desirable morphology that is closely related to sound absorption properties, it is crucial to apply an appropriate amount of dispersant to a specific amount of particle when manufacturing PU composite foams. Fig. 3(b) presents the open porosity of PU composite foams filled with melamine particles with and without dispersant.

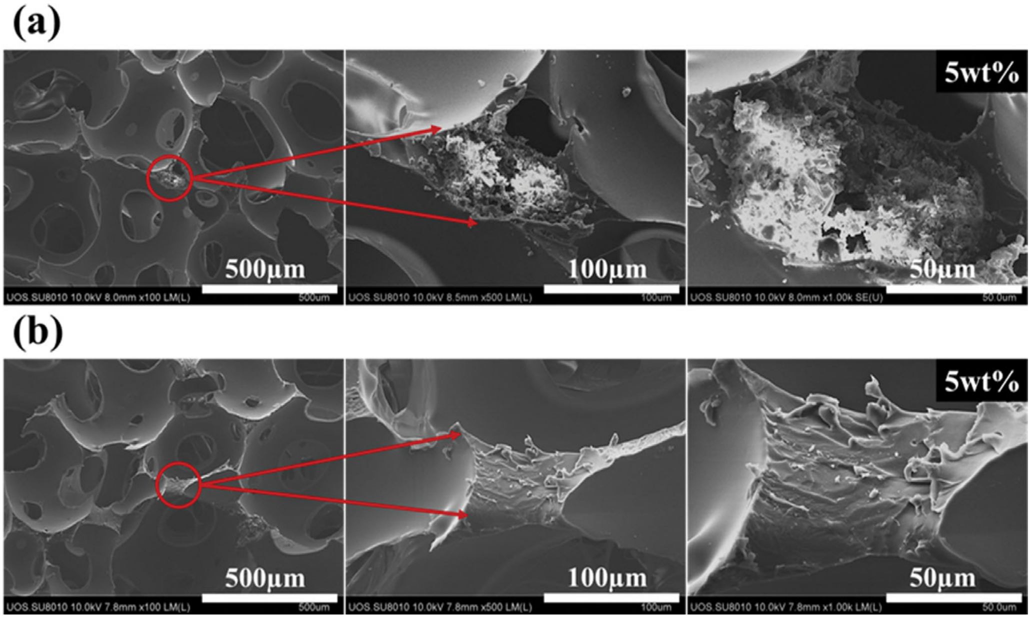

Fig. 4. **Typical SEM images of the PU composite foams including 5 wt% melamine particles without (a) and with (b) dispersant. Melamine particles are severely agglomerated in the wall without dispersant, but they are well dispersed in PU matrix with the dispersant.**

The open porosity was determined using Eq. (2): [15]

$$\text{Open porosity}=(N_o+N_p/2)/(N_o+N_p+N_c) \qquad (2)$$

where N_o, N_p, and N_c are the numbers of open, partially open, and closed pores. The open porosity exhibits a similar trend to the open pore ratio. The addition of melamine particles reduces the open porosity, and this effect is more pronounced in the case of melamine particles with dispersant. This is especially important for sound absorption characteristics at low frequencies because it increases the possibility of sound waves colliding with cavity struts, leading to scattering or transmission.

Fig. 4 depicts representative SEM images of the PU composite foams containing 5 wt% melamine particles with and without dispersant. Fig. 4(a) shows the aggregation of particles on the polymer matrix in the foams without dispersant. As explained in Fig. 3, excessive particle content can lead to poor compatibility between the particle surface and the PU matrix, resulting in an increase in open pore formation. Therefore, the open pore ratio of the PU composite foams containing more than 3 wt% of particles increased again. On the other hand, Fig. 4(b) illustrates that the addition of dispersant results in well-dispersed melamine particles in the PU matrix, leading to excellent interfacial compatibility and a further increase in the partially open pore ratio for particle content lower than 3 wt% (Fig. 3(a)). However, excessive amounts of dispersant can reduce the physical properties of the foams. Therefore, it is recommended to apply optimal amounts of particles and dispersant in the manufacture of PU composite foams to achieve the best performance.

2. Sound Absorption Properties

The sound absorption coefficient (α) is a measure of the amount of incident sound energy absorbed by a material. Porous materials absorb sound through three main mechanisms. First, air molecules vibrate with the pore walls, converting sound energy into heat energy. Second, the air in the pore is compressed and released, resulting in energy consumption during the energy conversion process. Third, sound energy is converted into heat energy through resonance with the pore walls [2,10,11]. The morphology of the material plays a crucial role in determining its sound absorption properties. To achieve optimal sound absorption performance, the material must have a large number of pores that are connected in an appropriate size for the propagation of sound waves, and there should be continuous channels between the inner pores and the outer surface [10].

Fig. 5(a) depicts the sound absorption coefficient of PU composite foams with varying melamine particle content without dispersant. The maximum peak value of α increases from 0.88 to 0.98 as the particle content increases. However, it decreases again when the particle content exceeds 3 wt%, which can be attributed to the small cavity size (Fig. 2) and the highest partially open pore ratio (Fig. 3) at 3 wt% melamine particle content. The small cavity size increases the air flow resistivity, increasing the residence time of

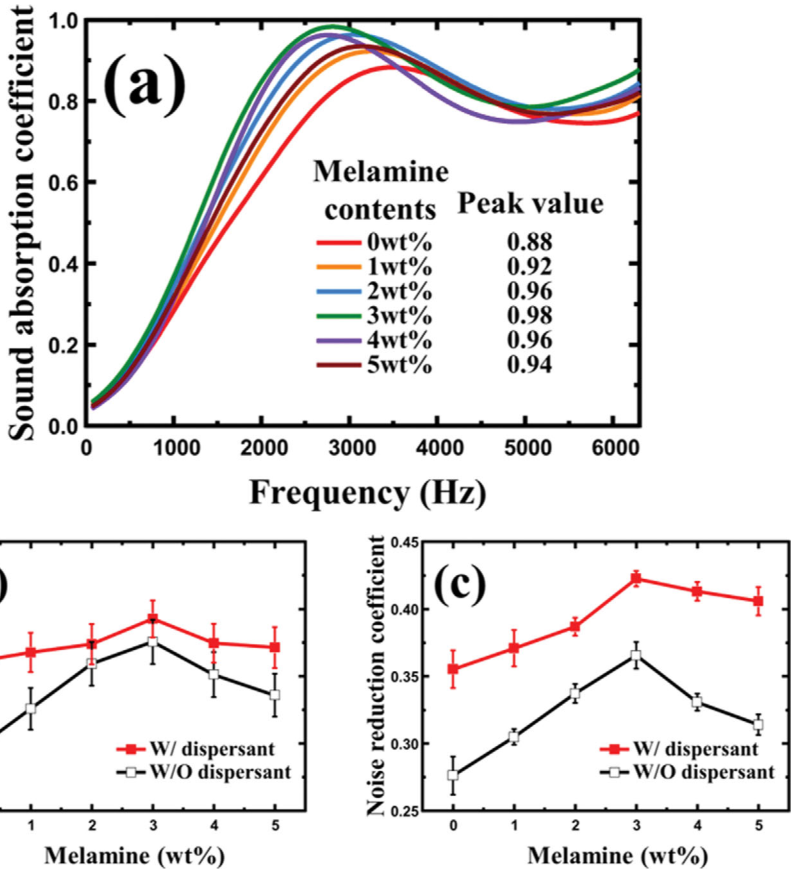

Fig. 5. (a) Sound absorption coefficient from the PU composite foams manufactured without dispersant, acoustic activity (b) and noise reduction coefficient (c) from the PU composite foams including melamine particles without and with dispersant.

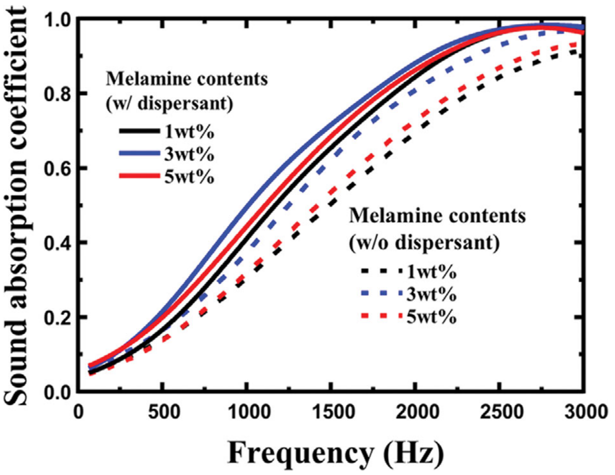

Fig. 6. Sound absorption coefficient of the PU composite foams including melamine particles without and with dispersant for the low frequency region.

sound waves in the foams and probability of collisions with air molecules, thereby improving sound absorption [17-19,33]. In addition, the highest partially open pore ratio promotes collisions with sound waves, resulting in better dissipation of sound energy through viscous friction and heat exchange, leading to higher sound absorption characteristics. Figs. 5(b) and 5(c) display the acoustic activity (AA) and noise reduction coefficient (NRC) of PU composite foams with melamine particles, both with and without dispersant. AA is calculated as the average of the sound absorption coefficient over the entire frequency range, while NRC is evaluated as the average of the sound absorption coefficient at 250, 500, 1,000, and 2,000 Hz [3,19,34,35]. The PU composite foams including 3 wt% melamine particles with dispersant show the highest performance in terms of both AA and NRC. Consequently, the particle not only reduces the cavity size but also increases the partially open pore ratio, improving sound absorption efficiency. Both factors were higher with the dispersant than without dispersant due to the highest partially open pore ratio and the well-developed cavity and pore structure resulting from reduced particle agglomeration.

Fig. 6 displays the sound absorption coefficients of the PU composite foams containing melamine particles, with and without dispersant, for the low frequency region. The mechanism of sound absorption differs based on the method of sound wave propagation, which changes depending on the frequency region. High frequencies have shorter wavelengths and stronger directionality, leading mainly to reflection. In contrast, low frequencies transmit more than they reflect due to their longer wavelengths and strong diffraction properties [36-38]. For porous structures, the highest open pore ratio is advantageous for sound absorption in the high frequency region, while the highest partially open pore ratio enhances sound absorption in the low frequency region through wall transmission. The PU composite foams with dispersant exhibit a higher partially open pore ratio than those without, indicating that transmission through the partially open pores is more effective in sound absorption than reflection through open pores in low frequency areas (as depicted in Fig. 3).

CONCLUSIONS

We investigated the morphology and sound absorption properties of PU composite foams containing melamine particles with and without a dispersant. Results showed that adding 3 wt% melamine particles improved sound absorption, with small cavity sizes and the highest partially open pore ratio. However, at higher particle content, particle aggregation caused defects in the PU matrix and reduced sound absorption. To address this issue, we added a dispersant to the PU composite foams, which improved interfacial compatibility between the PU matrix and melamine particles. As a result, the PU composite foams containing melamine particles with dispersant showed a higher partially open pore ratio than those without dispersant. This was due to increased wettability of the particles, which delayed open pore formation. As a result, the addition of the dispersant greatly improved NRC by effectively absorbing sound through transmission of partially open pores in the low frequency region.

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