

Effect of Surface-Modified Silica Nanoparticles on Cellular Structure and Sound Absorption Behavior of Polyurethane Foams

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Abstract

This study examined the effects of surface-modified silica nanoparticles on the cellular structure and sound absorption of polyurethane foams. Unmodified silica nanoparticles may show poor dispersion and weak compatibility with the polymer matrix. Therefore, the silica nanoparticles were modified with GPTMS and APTES silane coupling agents.

A total of 26 foam samples were prepared. These samples included pure polyurethane foam, foams with unmodified silica nanoparticles, and foams with surface-modified silica nanoparticles. FT-IR analysis confirmed successful surface modification. The observed acoustic trends were consistent with changes in nanoparticle-matrix interaction and foam structure after surface modification. The sound absorption coefficient was measured using an impedance tube according to ISO 10534-2.

The observed changes in cellular morphology **suggest** that surface modification affected the foaming process. The modified foams showed more uniform and interconnected cellular structures. These structural changes improved sound absorption, mainly at mid and high frequencies. Among the modified systems, the APTES-treated foams showed the most consistent improvement.

These results show that nanoparticle surface chemistry plays an important role in controlling the structure and sound absorption of polyurethane foams.

1. Introduction

Noise exposure is a common problem in buildings and workplaces. Sound-absorbing materials are often used to reduce noise and improve acoustic comfort (1–5). Polyurethane foams are widely used for this purpose because they have high porosity, low density, and good acoustic performance (6–8). Their acoustic performance is mainly determined by their cellular structure and viscoelastic behavior. Vibration of cell walls and the dissipation of viscous energy within the porous structure are the primary ways that flexible polyurethane foams absorb sound. Material characteristics like density, viscoelastic response, and pore structure affect their acoustic behavior (9). Micro- and nano-scale fillers have been added to the polymer matrix of polyurethane foams to enhance their sound absorption capabilities (9). These fillers can modify the cellular structure and viscoelastic behavior of the foam, which may improve acoustic energy dissipation (10, 11). Previous studies have shown that the acoustic performance of nanoparticle-filled polyurethane foams is influenced not only by filler content, but also by filler dispersion and interfacial bonding. Poor nanoparticle dispersion can create local stiffness variations and reduce acoustic energy dissipation within the foam (10–12). In cellular polymers, the final performance is closely related to foam morphology, including cell size distribution, cell wall thickness, and pore connectivity. Accordingly, controlling nanoparticle-matrix interaction is a key factor for achieving stable and efficient sound absorption behavior.

The performance of silica-filled polymer composites strongly depends on the chemical structure of the silica surface. Silica nanoparticles contain a large number of silanol groups, which promote particle

agglomeration through hydrogen bonding (13). As a result, weak interfacial interaction and non-uniform dispersion may occur inside the polymer matrix. Despite extensive studies on silica-filled polyurethane foams, limited attention has been paid to the influence of nanoparticle surface chemistry on sound absorption over different frequency ranges. Most previous studies have focused on filler loading, whereas the effects of surface modification on foam morphology and acoustic performance remain insufficiently understood.

Therefore, this study examined the effects of silica surface modification on the cellular structure and sound absorption of polyurethane foams. Silica nanoparticles were modified with GPTMS and APTES silane coupling agents. The modified and unmodified nanoparticles were added to polyurethane foam at different concentrations. The samples were then compared in terms of surface chemistry, cellular morphology, and sound absorption at low, mid, and high frequency ranges. The main aim was to determine whether nanoparticle surface modification could influence foam morphology and improve the sound absorption behavior of polyurethane foams.

2. Methods

2.1 Materials Used

Hydrophilic silica nanoparticles (20–30 nm, > 99% purity, specific surface area: 150 m²/g) were supplied by Mokarrar Industrial Group. Polyol HR-338 and MDI-based isocyanate (8001) were obtained from Mokarrar Engineering Materials Company. GPTMS and APTES were used as silane coupling agents for surface modification of silica nanoparticles. Toluene and ethanol were used as solvents, and triethylamine (TEA) was used as the catalyst.

2.2 Sample Preparation

To remove moisture from the silica particles, they were placed in a vacuum oven at 70°C for 12 hours. Then, they were mixed with 80 mL of toluene on a magnetic stirrer for 10 minutes to ensure proper blending. The samples were then subjected to ultrasonic waves using a Bandelin HD 3200 ultrasonic device from Germany, equipped with a KE-76 probe at 70% power for 3 minutes. A 0.038 g amount of triethylamine was added as a catalyst. Next, 3 mL of GPTMS silane was added dropwise to the solution, which was then stirred on a magnetic stirrer for 30 minutes. The necessary materials were added to the stirring solution, and the reaction was carried out at 80°C for 24 hours under mechanical stirring. To separate the modified nanoparticles and remove unreacted silanes, the obtained mixture was washed three times with ethanol using a centrifuge at 9000 rpm for 30 minutes per cycle. After centrifugation, the sample was left to dry in an open environment for 24 hours, followed by further drying in a vacuum oven at 60°C for another 24 hours.

The same surface modification procedure was applied to APTES-modified silica nanoparticles by replacing GPTMS with the same volume of APTES under identical reaction conditions.

In the next step, the desired mold for polyurethane foam fabrication was made of Teflon using the machining process. The main components of polyurethane foam included polyol, isocyanate, a catalyst, and a surfactant, with a silicone-based surfactant used in this process. Initially, for the production of pure polyurethane foam, isocyanate was added to polyol, and the resulting mixture was stirred for 35 seconds at 2500 rpm using an electric mixer. The mixture was then left undisturbed at room temperature for 10 minutes to allow the foam formation process to complete. After this stage, the formed foam was removed from the mold and cut to specified dimensions using a laser cutting device, with diameters of 3 cm and 6 cm and a thickness of 2 cm.

For nanocomposite foams, the required amount of modified or unmodified silica nanoparticles was first dispersed in the polyol component. The mixture was mechanically stirred for 1 min before adding the isocyanate. Foam preparation was then carried out using the same procedure as that used for pure polyurethane foam.

2.3 Design, Analysis, and Optimization

In this study, the design and analysis of data related to the effect of combined modified nanosilica on the sound absorption coefficient were performed using Design Expert 11 software. The experiments were conducted randomly to minimize systematic errors. After conducting the experiments based on the Central Composite Design (CCD) experimental plan, analysis of variance (ANOVA) was used to examine relationships between parameters, describe response variables, and predict optimal conditions. ANOVA was employed to evaluate the combined effect of nanosilica modified with glycidoxypolytrimethoxysilane (GPTMS) and nanosilica modified with 3-aminopropyltriethoxysilane (APTES) on the sound absorption coefficient. A polyol-to-isocyanate weight ratio of 40:60 was considered optimal in terms of foam formation quality and structural stability after formation, and this ratio was used in the study.

A set of experimental runs was designed to cover different contents of unmodified and surface-modified silica nanoparticles.

2.4 Determination of Sound Absorption Coefficient

The sound absorption coefficient of the samples was measured using an impedance tube device in accordance with ISO 10534-2 standards (14, 15). Sound absorption was measured using a BSWA SW477 impedance tube according to ISO 10534-2. In this device, sound waves were generated by a sound source inside the tube. Then, by measuring the acoustic pressures at two fixed locations using microphones mounted on the tube wall and calculating the complex acoustic transfer function, the sound absorption in normal incidence and the impedance ratios of the absorbing material were determined. The impedance tube system was equipped with a large-diameter tube (6 cm) for measuring the sound absorption coefficient at low frequencies (63–1600 Hz) and a small-diameter tube (3 cm) for measuring the sound absorption coefficient at high frequencies (1600–6300 Hz).

2.5 Determination of Apparent Density and Surface Porosity

The apparent density of each foam sample was determined using the geometric volume method according to ASTM D1622. The samples had a cylindrical shape, and their volume was calculated from the measured diameter and thickness. Apparent density was calculated by dividing the sample mass by its geometric volume.

Surface porosity was estimated from SEM images using image-processing analysis in MATLAB. The images were converted to binary images using a global thresholding method. The surface porosity was calculated from the ratio of pore pixels to the total number of pixels in the analyzed image.

2.6 SEM Characterization

The cellular structure of the polyurethane foams was examined using a SU3500 scanning electron microscope. Images were obtained from the cross-sections of selected foam samples. Several fields of view were recorded for each sample. Images at the same magnification were used for visual comparison. The representative images were obtained at $\times 20$ magnification with a 2.00 mm scale bar. These images were used to compare cell shape, cell-size distribution, and pore connectivity among the selected formulations. The SEM images were used to evaluate foam morphology. They were not used as direct evidence of nanoparticle dispersion. Representative SEM images were selected from several fields of view for each sample.

3. Results

A total of 26 polyurethane foam formulations were prepared, including pure polyurethane foam, foams containing unmodified silica nanoparticles, and foams containing GPTMS-, APTES-, and combined surface-modified silica nanoparticles. Representative physical and acoustic properties of selected formulations are summarized in Table 2. The acoustic response varied with both nanoparticle surface treatment and formulation. The optimum sample depended on the frequency range. Sample 13 showed the highest average low-frequency absorption, Sample 14 showed the highest average mid-frequency absorption, and Sample 21 showed the highest average high-frequency absorption. Differences in apparent density and SEM-derived surface porosity were also observed among the formulations, indicating that nanoparticle surface modification influenced the cellular structure of the foams. Figure 1 shows the FTIR spectra of unmodified and surface-modified silica nanoparticles. New characteristic peaks observed after modification confirm successful chemical bonding of GPTMS and APTES on the silica surface.

Figure 2 presents representative SEM images of selected polyurethane foams. The pure polyurethane foam showed a heterogeneous cellular structure with a broad distribution of cell sizes. The foam containing unmodified silica nanoparticles also exhibited several large and irregular cells. In contrast, the foams prepared with surface-modified silica nanoparticles showed a more uniform cellular structure and

a more homogeneous distribution of pores. The combined modified systems also exhibited interconnected open-cell structures with fewer oversized cavities. These observations suggest that surface modification influenced foam morphology, which may contribute to the differences observed in sound absorption performance. The SEM images describe changes in foam morphology but do not directly demonstrate nanoparticle dispersion within the polymer matrix.

The experimental design and formulation details of the prepared samples are summarized in Table 1.

Table 1

Experimental design and formulation details of polyurethane foam samples containing unmodified and surface-modified silica nanoparticles.

Experimental Run	Sample Code	Unmodified Nano-Silica (%)	GPTMS-Modified Nano-Silica (%)	APTES-Modified Nano-Silica (%)	Thickness (cm)	Diameter (cm)
1	Pure PU	0	0	0	2	3 & 6
2	PU with Unmodified Nano	1	0	0	2	3 & 6
3	PU with Unmodified Nano	2	0	0	2	3 & 6
4	PU with Unmodified Nano	3	0	0	2	3 & 6
5	PU with Unmodified Nano	4	0	0	2	3 & 6
6	PU with Modified Nano GPTMS	0	1	0	2	3 & 6
7	PU with Modified Nano GPTMS	0	2	0	2	3 & 6
8	PU with Modified Nano GPTMS	0	3	0	2	3 & 6
9	PU with Modified Nano GPTMS	0	4	0	2	3 & 6
10	PU with Modified Nano APTES	0	0	1	2	3 & 6
11	PU with Modified Nano APTES	0	0	2	2	3 & 6
12	PU with Modified Nano APTES	0	0	3	2	3 & 6

Experimental Run	Sample Code	Unmodified Nano-Silica (%)	GPTMS-Modified Nano-Silica (%)	APTES-Modified Nano-Silica (%)	Thickness (cm)	Diameter (cm)
13	PU with Modified Nano APTES	0	0	4	2	3 & 6
14	PU with Combined Modified Nano	0	1	3	2	3 & 6
15	PU with Combined Modified Nano	0	3	1	2	3 & 6
16	PU with Combined Modified Nano	0	3	0	2	3 & 6
17	PU with Combined Modified Nano	0	2	2	2	3 & 6
18	PU with Combined Modified Nano	0	0	2	2	3 & 6
19	PU with Combined Modified Nano	0	2	2	2	3 & 6
20	PU with Combined Modified Nano	0	2	4	2	3 & 6
21	PU with Combined Modified Nano	0	1	1	2	3 & 6
22	PU with Combined Modified Nano	0	2	2	2	3 & 6
23	PU with Combined Modified Nano	0	4	2	2	3 & 6

Experimental Run	Sample Code	Unmodified Nano-Silica (%)	GPTMS-Modified Nano-Silica (%)	APTES-Modified Nano-Silica (%)	Thickness (cm)	Diameter (cm)
24	PU with Combined Modified Nano	0	2	2	2	3 & 6
25	PU with Combined Modified Nano	0	3	3	2	3 & 6
26	PU with Combined Modified Nano	0	2	2	2	3 & 6

3.1 Sound Absorption Coefficient

The sound absorption coefficients of the prepared polyurethane foams were measured over the frequency range of 100–6300 Hz. The results are presented according to the type of silica nanoparticle surface treatment to facilitate comparison among the different formulations. The results of the sound absorption coefficient measurements for the samples, obtained using the impedance tube method in the frequency range of 100 to 6300 Hz, are presented in Table 2. Additionally, the average absorption coefficients measured in three frequency ranges, low frequency (100 to 500 Hz), mid-frequency (500 to 1600 Hz), and high frequency (1600 to 6300 Hz) are reported separately in Table 2.

Table 2

Representative physical and acoustic properties of polyurethane foams filled with unmodified and surface-modified silica nanoparticles.

Representative formulation	Apparent density (kg/m ³)	SEM-derived surface porosity (%)	Avg. low-frequency absorption	Avg. mid-frequency absorption	Avg. high-frequency absorption	NRC
Pure PU	43	40.44	0.065	0.157	0.562	0.156
PU + 1 wt% unmodified silica	46	47.85	0.055	0.168	0.549	0.171
PU + 2 wt% GPTMS-modified silica	49	42.52	0.064	0.167	0.554	0.156
PU + 1 wt% APTES-modified silica	54	40.95	0.064	0.212	0.558	0.191

Table 2 summarizes the representative physical and acoustic properties of selected polyurethane foam formulations. Compared with the pure polyurethane foam, surface-modified silica nanoparticles generally improved the acoustic performance. Among the investigated formulations, the foam containing 1 wt% APTES-modified silica showed the highest average mid-frequency sound absorption coefficient and the highest NRC. GPTMS-modified silica also improved the acoustic response compared with unmodified silica. These results indicate that nanoparticle surface modification had a greater influence on acoustic performance than the addition of unmodified silica alone.

Figure 3 compares the sound absorption coefficients of pure polyurethane foam and foams containing different contents of unmodified silica nanoparticles. All samples showed relatively low sound absorption at low frequencies, followed by a gradual increase with increasing frequency. The maximum sound absorption occurred in the mid-frequency range, after which the absorption coefficient gradually decreased at higher frequencies. Among the unmodified silica formulations, the foam containing 1 wt% silica exhibited the highest sound absorption over most of the measured frequency range. Increasing the silica content beyond 1 wt% did not result in further improvement and, in some frequency ranges, reduced the acoustic performance. These results indicate that the addition of unmodified silica alone is not sufficient to consistently improve the sound absorption behavior of polyurethane foams.

1. The sound absorption coefficient of polyurethane foam with modified nanoparticles

Figure 4 shows the results for foams with GPTMS-modified silica and APTES-modified silica. Sample 7 contained 2 wt% GPTMS-modified silica. It showed the highest average high-frequency absorption among the GPTMS samples. The value was 0.554. Sample 13 contained 4 wt% APTES-modified silica. It showed the highest average low-frequency absorption. The value was 0.085. Sample 10 contained 1 wt% APTES-modified silica. It showed the highest average mid-frequency absorption among the APTES samples. The value was 0.212. The absorption of the APTES samples increased up to about 4000 Hz. It then decreased at higher frequencies.

Figure 5 compares several foams with combined GPTMS- and APTES-modified silica. The foam with 1 wt% GPTMS and 3 wt% APTES usually showed higher absorption than the foam with 3 wt% GPTMS and 1 wt% APTES. Their values were close at 50–500 Hz and 2500–3000 Hz. The foam with 2 wt% GPTMS showed higher absorption than the foam with 2 wt% APTES at frequencies below 2000 Hz. However, the foam with 2 wt% APTES showed higher absorption at 2000–3000 Hz. It also kept a more stable trend at 4000–6300 Hz. The foams with 4 wt% GPTMS and 2 wt% APTES and with 3 wt% GPTMS and 3 wt% APTES showed similar trends below 1600 Hz. The foam with 4 wt% GPTMS and 2 wt% APTES showed the highest absorption at 1600–3600 Hz.

Additional statistical and optimization results are provided in the Supplementary Material.

4. Discussion

The effect of unmodified silica depended on both filler content and frequency. The sample with 1 wt% unmodified silica showed a small increase in mid-frequency absorption. However, higher silica contents did not produce a consistent improvement. Some samples even showed lower absorption than pure polyurethane foam. Pirani et al. also reported that silica content can affect the acoustic response of polyurethane foam through changes in cellular morphology (16). These findings show that filler content alone is not enough to improve sound absorption.

The lower absorption observed at higher loadings of unmodified silica may be related to particle agglomeration and poor compatibility with the polymer matrix. These effects can disturb cell formation and create local changes in stiffness. As a result, the foam structure may become less effective in dissipating acoustic energy. This may explain why increasing the unmodified silica content above 1 wt% did not produce a consistent improvement in sound absorption.

In contrast, surface-modified silica produced a better acoustic response than unmodified silica in several formulations. The improvement was more evident at mid and high frequencies. Silane treatment may reduce the strong interaction between silica particles and improve their compatibility with the polyurethane matrix. This may contribute to the formation of a more uniform foam structure during foaming. Babaei et al. also reported that silane treatment changes silica surface properties and improves compatibility with polyurethane systems (17).

Among the modified samples, the APTES-treated foams showed the most consistent improvement in sound absorption. This may be related to the amino groups of APTES and their interaction with the polyurethane matrix. Better compatibility may help produce a more stable cellular structure. It may also increase internal friction during sound propagation. Wenbo et al. also showed that pore structure and density can strongly affect the sound absorption of polyurethane foams (18).

The influence of porosity and foam morphology is also important for sound absorption. The SEM observations showed noticeable changes in the cellular structure after the addition of surface-modified silica nanoparticles. The modified foams generally exhibited a more uniform pore structure than the unmodified foams. A more homogeneous cellular structure may provide longer pathways for sound propagation and increase viscous and thermal energy dissipation within the foam. These changes may contribute to the improved acoustic performance observed in the modified samples (19–21).

However, increasing the nanoparticle content did not always improve sound absorption. Higher filler contents may influence the foaming process and reduce the uniformity of the cellular structure. As a result, the acoustic performance may not continue to improve with increasing nanoparticle loading. Mohammadi et al. (2021) reported similar observations, showing that excessive filler content can reduce the acoustic efficiency of polyurethane foams (22). Therefore, an optimum nanoparticle content appears to be necessary for achieving improved sound absorption.

In combined modified systems, increasing the proportion of APTES enhanced sound absorption, while higher GPTMS content showed less improvement. These results suggest that nanoparticle surface

chemistry played an important role in influencing the acoustic behavior of polyurethane foams. Although density showed some correlation with sound reduction behavior, the relationship was not strictly linear. Similar discussions on density effects were reported by Nandanwar et al. (2017), although their findings differed due to differences in material systems (23).

Overall, the improved sound absorption of the surface-modified polyurethane foams can be explained by changes in foam morphology after nanoparticle surface treatment. The modified foams generally showed a more uniform cellular structure, which may provide longer pathways for sound propagation and increase viscous and thermal energy dissipation within the porous network. Similar mechanisms have been reported in previous studies on polyurethane acoustic foams and surface-modified polymer nanocomposites (24–26). Therefore, the results suggest that nanoparticle surface chemistry is an important factor influencing the acoustic behavior of polyurethane foams.

5. Conclusion

This study investigated the effect of surface-modified silica nanoparticles on the cellular structure and sound absorption of polyurethane foams. Compared with pure polyurethane foam and foams containing unmodified silica, surface-modified silica nanoparticles improved the acoustic performance, particularly at mid and high frequencies. Among the investigated formulations, APTES-modified silica showed the most consistent improvement in sound absorption.

FT-IR analysis confirmed the successful surface modification of silica nanoparticles. SEM observations showed noticeable changes in foam morphology after surface treatment. These structural changes were consistent with the improved acoustic performance of the modified foams. The findings suggest that nanoparticle surface chemistry plays an important role in controlling the cellular structure and acoustic behavior of polyurethane foams.

Overall, surface modification of silica nanoparticles provides an effective approach for improving the sound absorption performance of polyurethane foams. The results of this study may support the future development of lightweight polymer-based acoustic materials for engineering and noise control applications.

Declarations

Ethical Approval

This study was approved by the Ethics Committee of Zanjan University of Medical Sciences under the ethical approval code IR.ZUMS.REC.1401.035. Since this research does not involve human or animal subjects, ethical approval was obtained to ensure compliance with research integrity and safety guidelines.

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Conflicts of Interest

The authors declare that they have no conflict of interest.

Data and Code Availability

The datasets generated and analyzed during the current study are available from the corresponding author upon reasonable request.

AI Statement

During the preparation of this work, the authors used ChatGPT to assist with the translation and improve the language of the manuscript, as the authors are non-native English speakers. The authors reviewed and edited the generated text as necessary and accept full responsibility for the content of the published article.

Authors' Contributions

Roohalah Hajizadeh: Conceptualization, Resources, Funding Acquisition, Supervision, Validation.

Amirhossein Montazeri: Methodology, Writing – Original Draft, Writing – Review & Editing, Supervision, Data Curation.

Mehrana Esnaasharieh: Conceptualization, Methodology, Investigation, Writing – Original Draft, Writing – Review & Editing.

Mohammad Hossein Beheshti*: Funding Acquisition, Software, Resources, Project administration.

Ruhollah Jamshidi*: Formal analysis, Conceptualization, Data Curation, Investigation, Visualization

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Figures

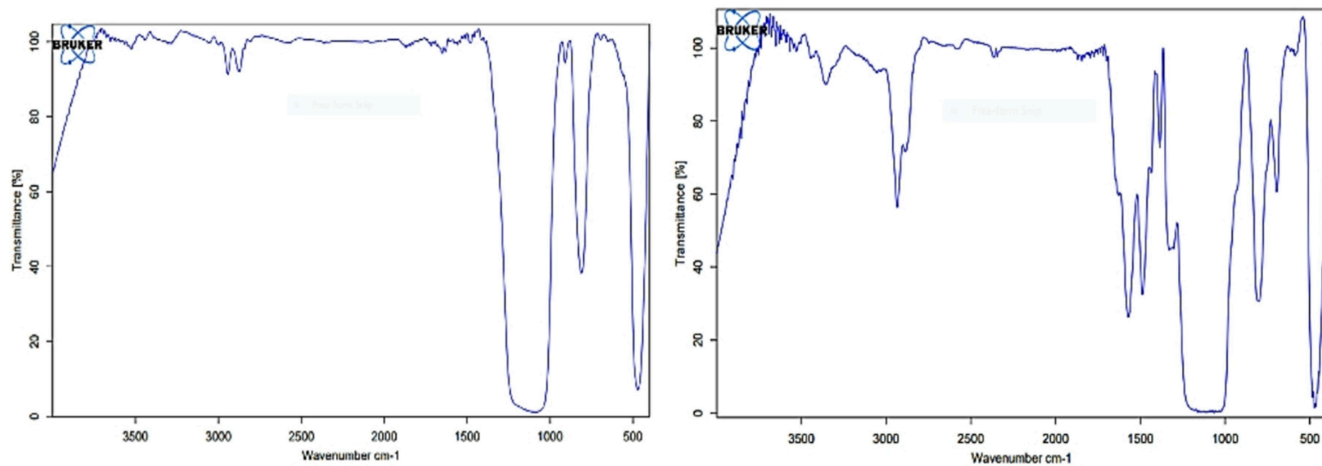


Figure 1

FT-IR spectrum of silica particles modified with the silane agents GPTMS and APTES

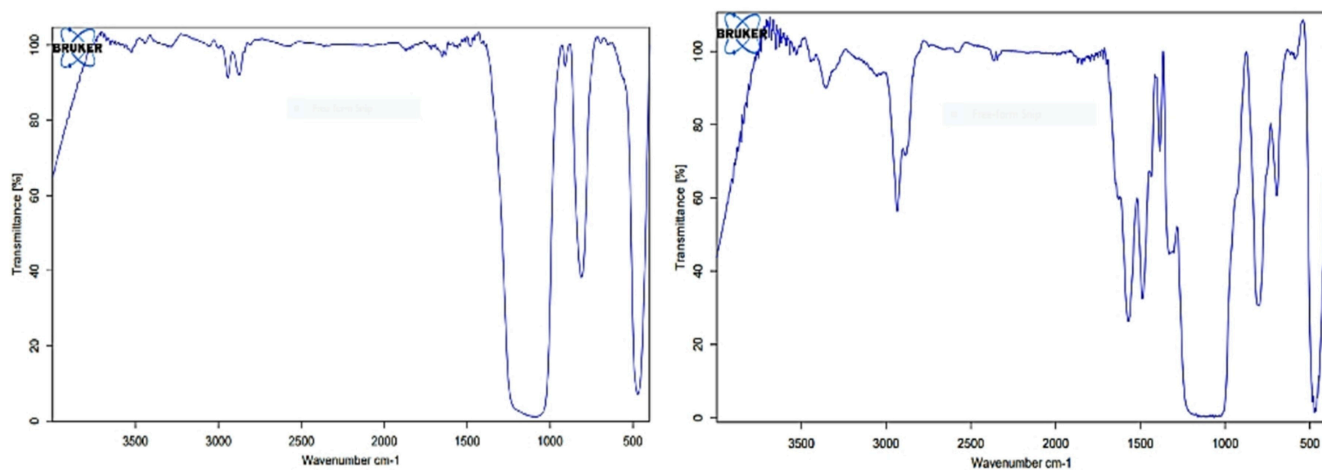


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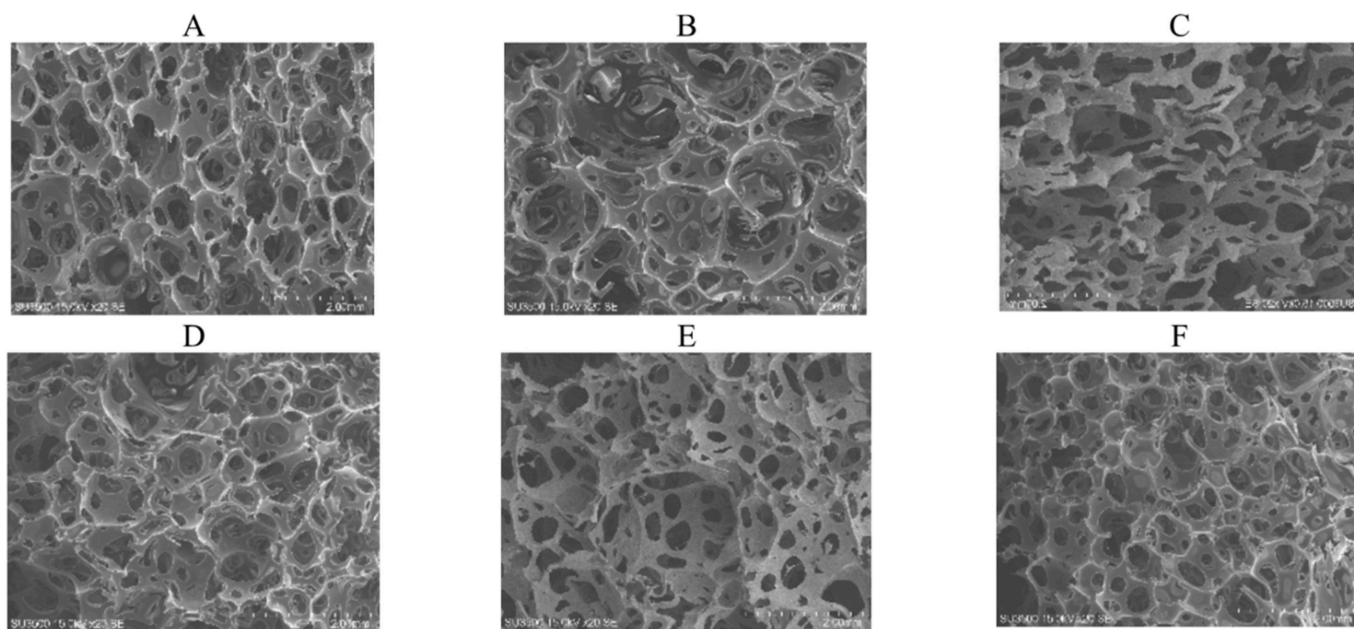


Figure 2

Representative SEM images of selected polyurethane foams obtained at the same magnification: (A) pure polyurethane foam, (B) foam containing 1 wt% unmodified silica, (C) foam containing 2 wt% GPTMS-modified silica, (D) foam containing 1 wt% APTES-modified silica, (E) foam containing 1 wt% GPTMS-modified silica and 3 wt% APTES-modified silica, and (F) foam containing 1 wt% GPTMS-modified silica and 1 wt% APTES-modified silica.

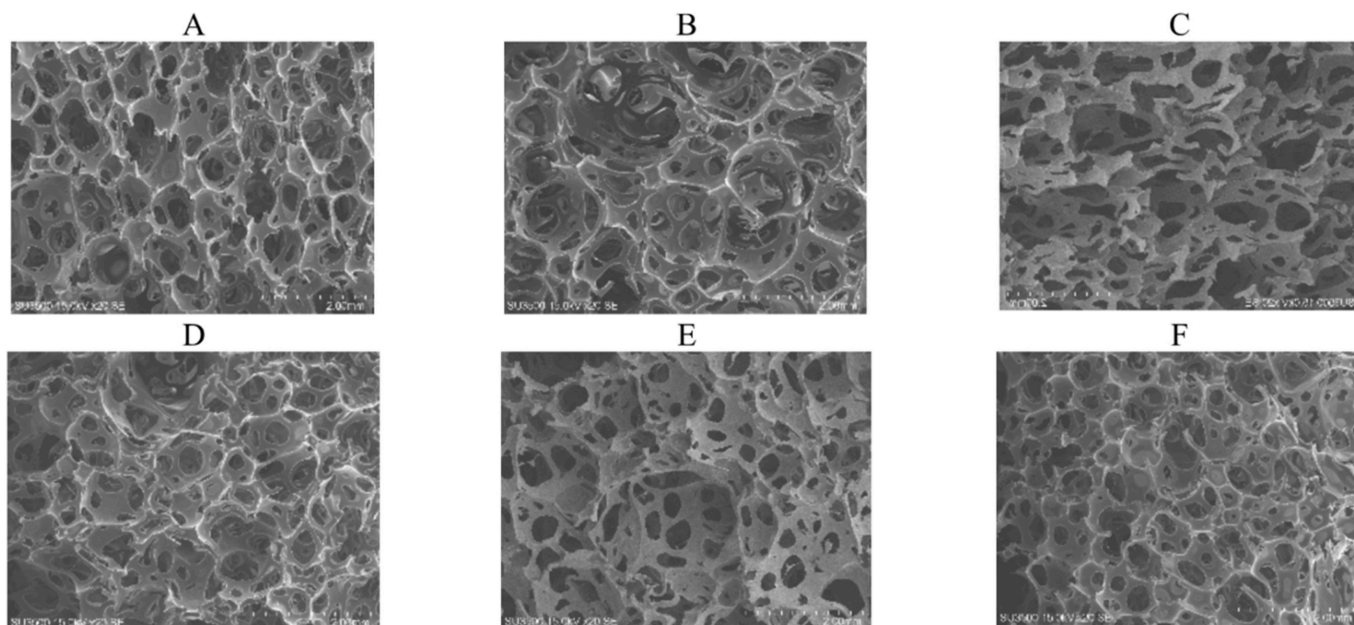


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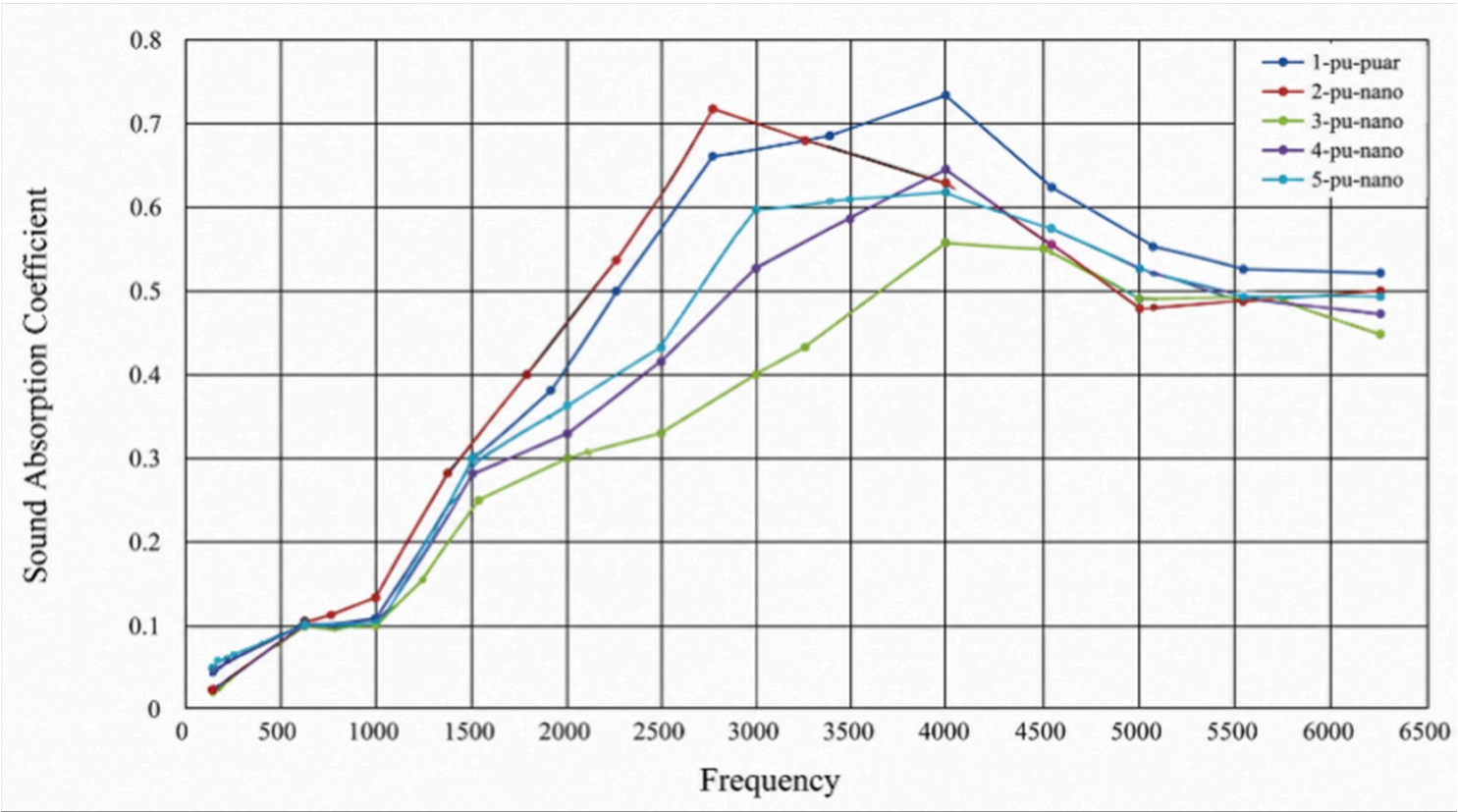


Figure 3

Sound absorption coefficients of pure polyurethane foam and foams containing 1, 2, 3, and 4 wt% unmodified silica nanoparticles

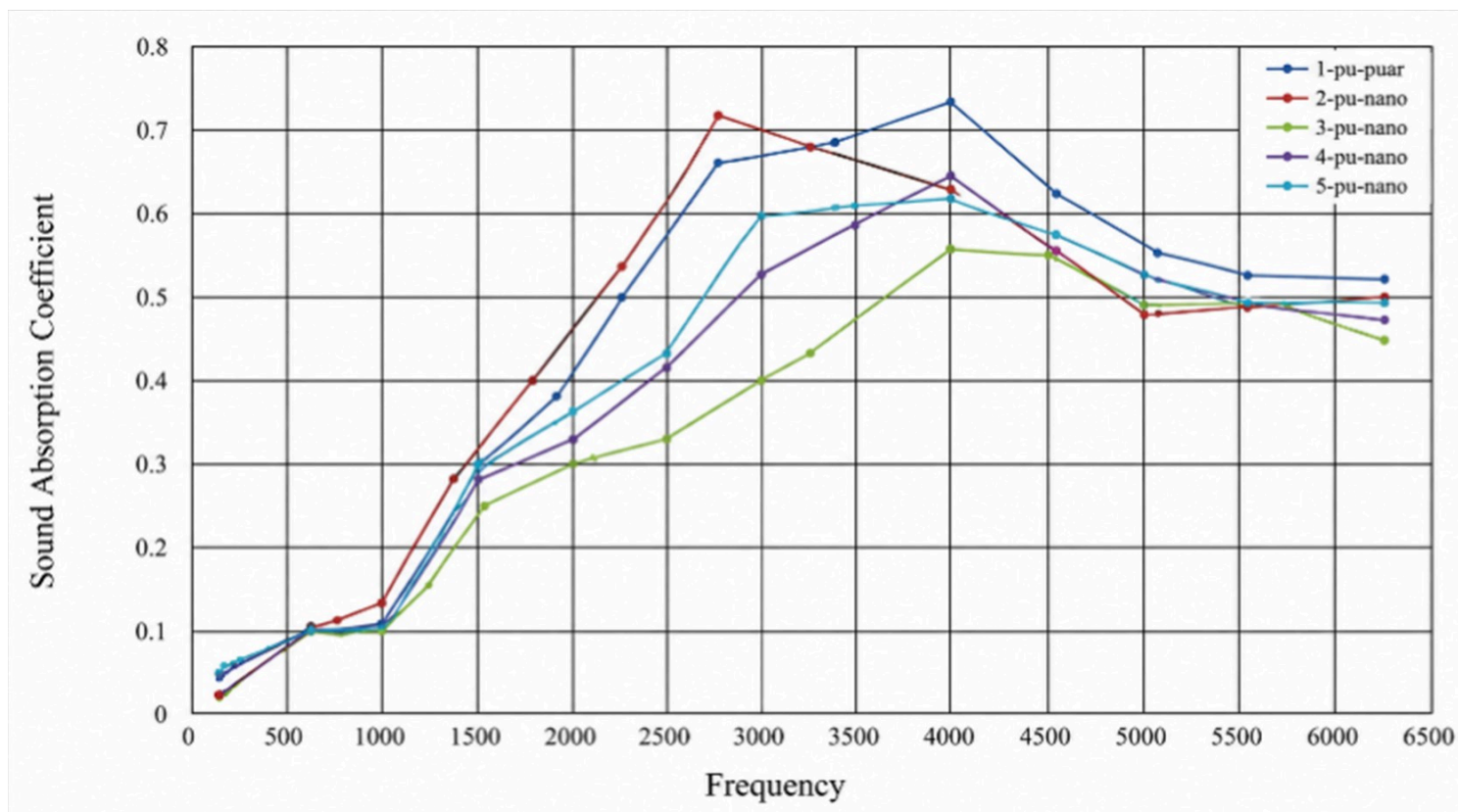


Figure 3

Sound absorption coefficients of pure polyurethane foam and foams containing 1, 2, 3, and 4 wt% unmodified silica nanoparticles

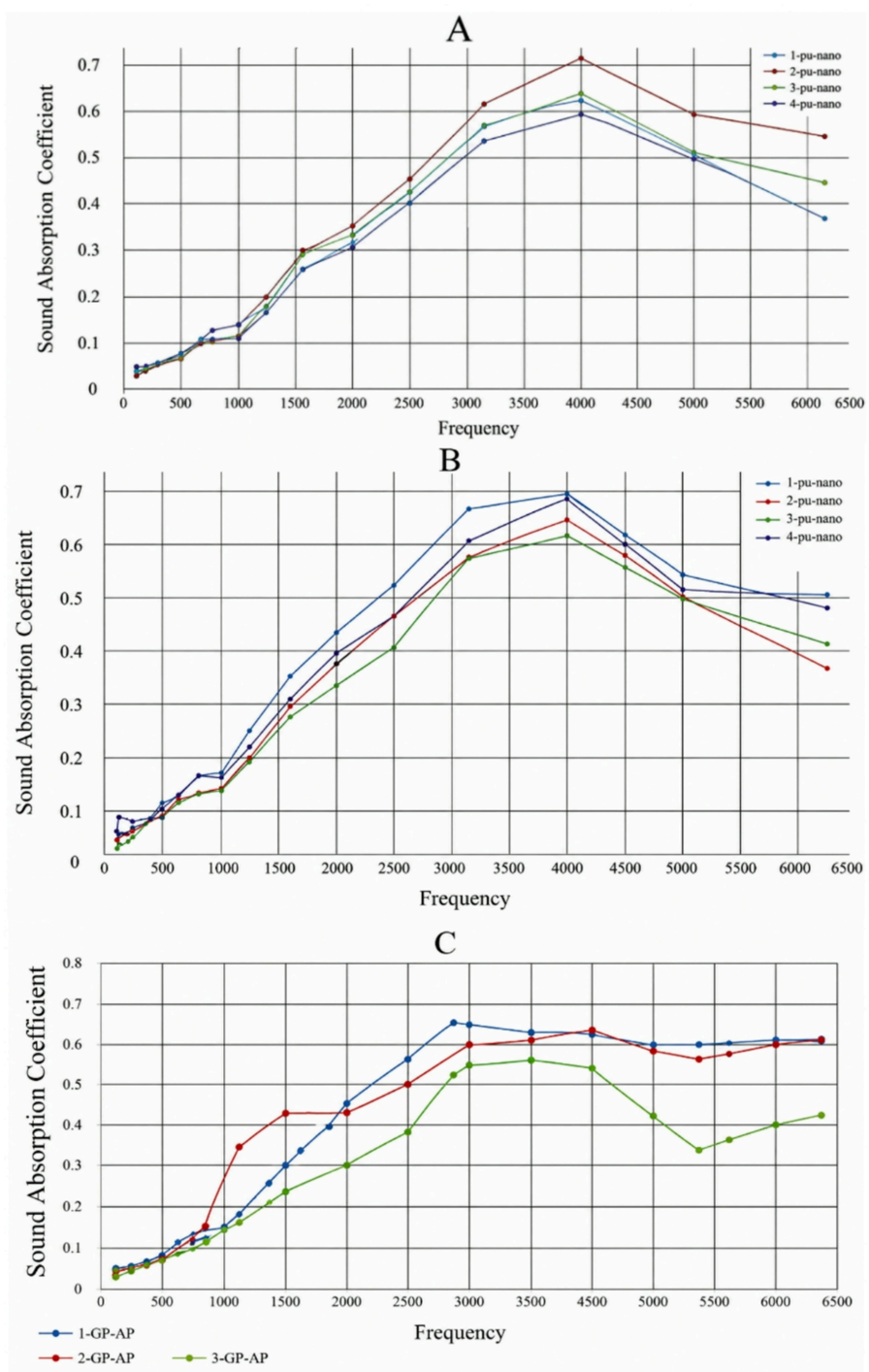


Figure 4

Sound absorption coefficients of polyurethane foams containing GPTMS-modified silica (A), APTES-modified silica (B), and combined GPTMS/APTES-modified silica (C)

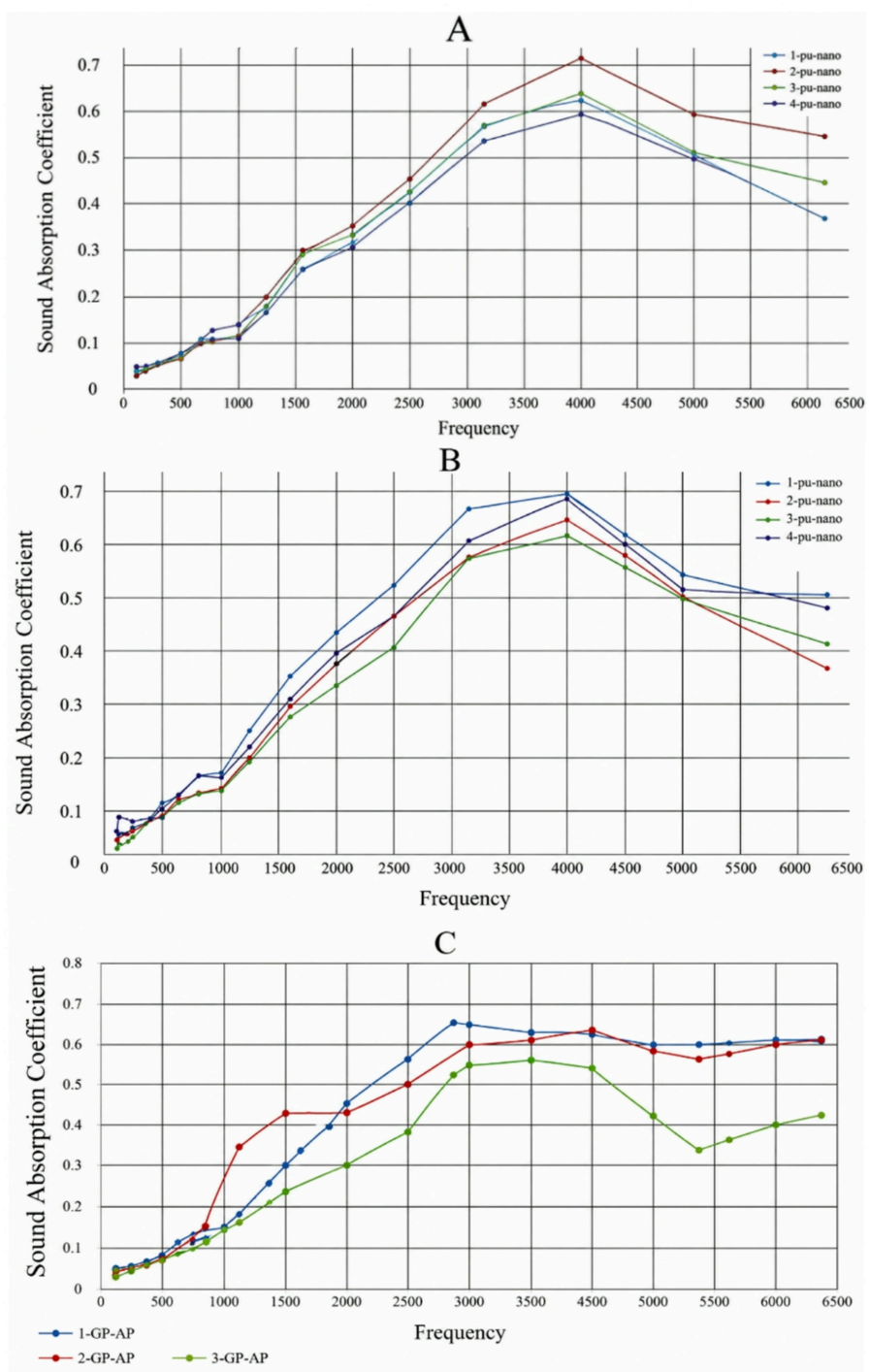


Figure 4

Sound absorption coefficients of polyurethane foams containing GPTMS-modified silica (A), APTES-modified silica (B), and combined GPTMS/APTES-modified silica (C)

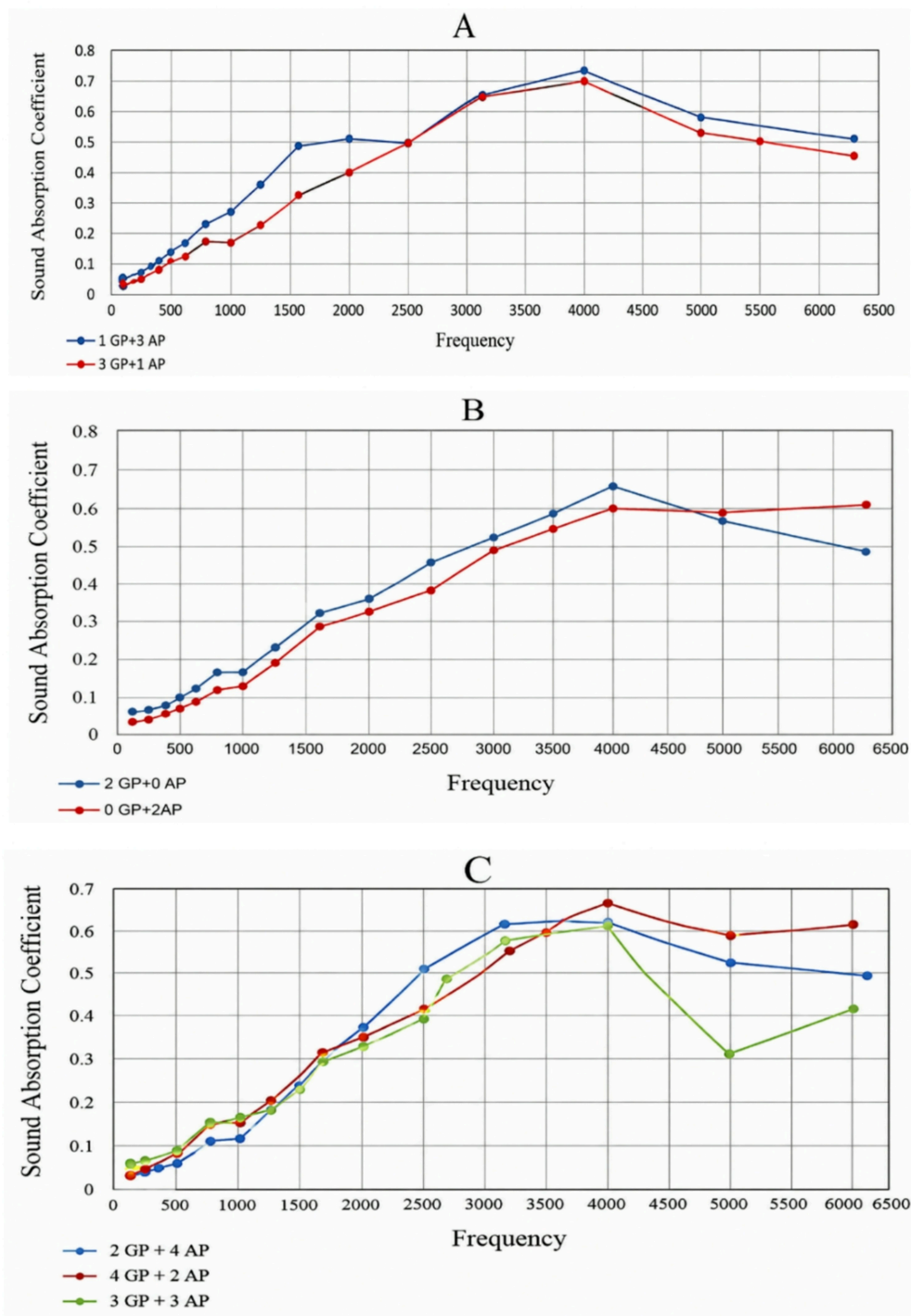


Figure 5

Comparison of the sound absorption coefficients of polyurethane foams containing different combinations of GPTMS- and APTES-modified silica nanoparticles: (A) 1 wt% GPTMS + 3 wt% APTES and 3 wt% GPTMS + 1 wt% APTES; (B) 2 wt% GPTMS and 2 wt% APTES; and (C) combined formulations containing 2 wt% GPTMS + 4 wt% APTES, 4 wt% GPTMS + 2 wt% APTES, and 3 wt% GPTMS + 3 wt% APTES.

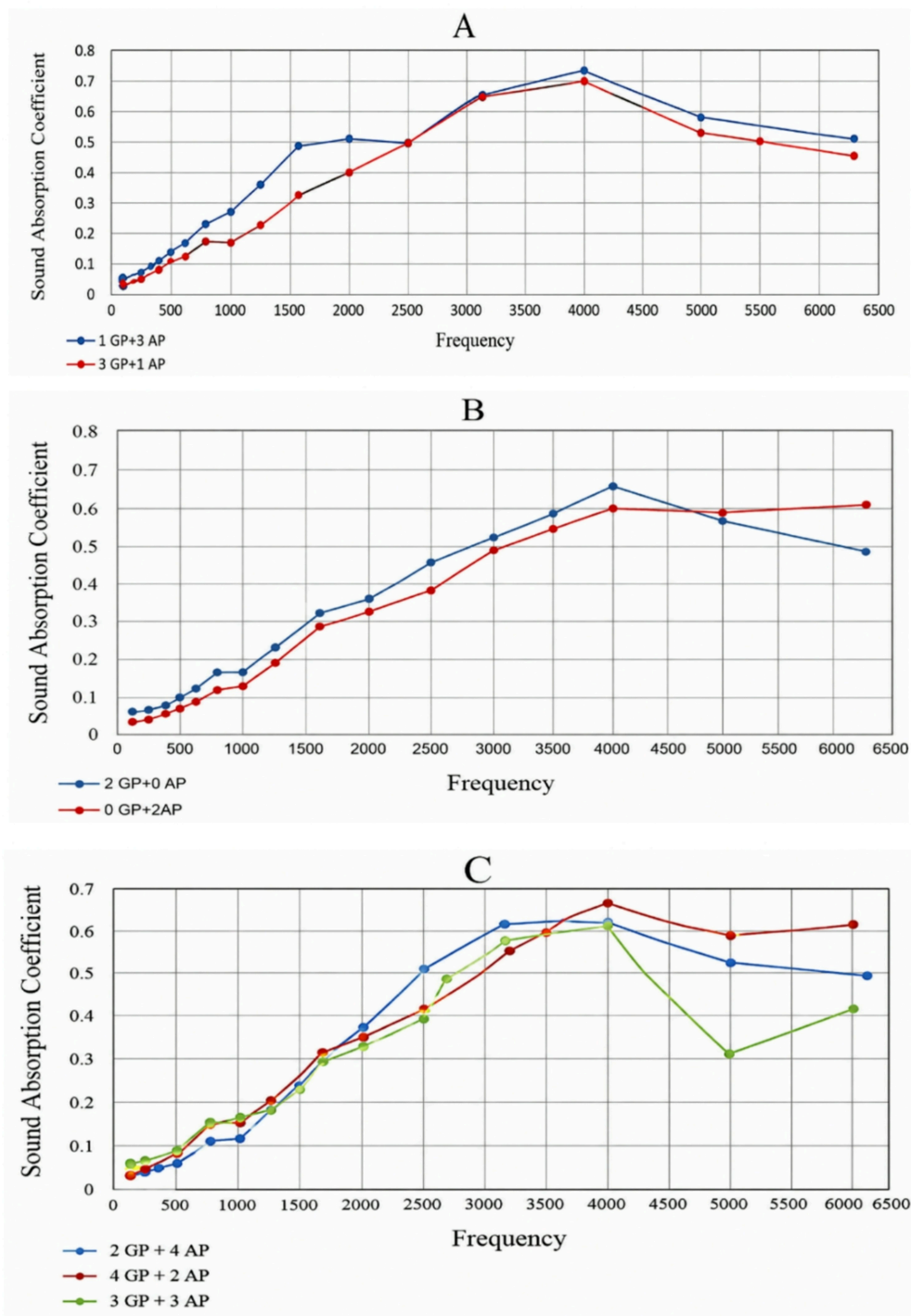


Figure 5

Comparison of the sound absorption coefficients of polyurethane foams containing different combinations of GPTMS- and APTES-modified silica nanoparticles: (A) 1 wt% GPTMS + 3 wt% APTES and 3 wt% GPTMS + 1 wt% APTES; (B) 2 wt% GPTMS and 2 wt% APTES; and (C) combined formulations containing 2 wt% GPTMS + 4 wt% APTES, 4 wt% GPTMS + 2 wt% APTES, and 3 wt% GPTMS + 3 wt% APTES.