Directly freeze-drying porous graphene aerogel as acoustic-absorbing material

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Abstract. Noise pollution, which is no less harmful than air, water and soil pollution, is becoming an environmental shortcoming that cannot be ignored. Scientists are also seeking various solutions to work on reducing noise pollution. In this work, porous graphene aerogel (FD-GA) and porous graphene oxide aerogel (FD-GOA) prepared by foaming combined with freeze-drying are reported as acoustic-absorbing materials. The effects of thickness and degree of compression on the acoustic-absorbing performance of FD-GA and FD-GOA in the 1000-6000 Hz range, where human ear hearing is sensitive, were investigated, respectively. FD-GA was found to have significantly higher acoustic absorption performance with a peak absorption of 96.7%. The present work provides a simple and straightforward strategy for the synthesis of the acoustic-absorbing material. Key words: freeze-drying, graphene aerogel, acoustic-absorbing

1. Introduction

Noise can cause both auditory and non-auditory health effects. Noise exposure can cause annoyance and even induce hypertension and cardiovascular disease\cite{1}. Currently, the use of porous materials to control noise is the most common method. Various natural acoustic-absorbing materials (such as pineapple-leaf fibres\cite{2}, kenaf\cite{3}, coir fibre\cite{4}) and synthetic acoustic-absorbing materials (such as typically polyurethane and its composite materials\cite{5,6}) have been developed.

In recent years, due to the excellent physical properties of graphene, graphene-based composite porous acoustic-absorbing materials have gradually appeared in the field of noise control. For example, several incorporated graphene oxide (GO) into melamine foam by dipping to build a graphene oxide-based flake network in the foam skeleton\cite{7}. This composite structure showed a 60% improvement in the absorption band from 128 Hz to 4000 Hz. Similarly, Liu et al. separated the self-assembled graphene oxide film connected with the melamine network by introducing functionalized carbon nanotubes, which increased the airflow resistance and tortuosity, and also improved the acoustic absorption performance\cite{8}. In addition to incorporating graphene into melamine, composites of graphene and polyurethane foam have also been developed. Jung-Hwan et al. reported an antagonistic graphene oxide-polyurethane hybrid aerogel with directional pores, which exhibited high acoustic absorption capacity at low frequencies\cite{9}.
So far, there are very few reports on the use of simple three-dimensional graphene as acoustic-absorbing materials. In order to give full play to the physical advantages of graphene, we have adopted a method that does not combine with other materials, and directly prepares FD-GA as acoustic-absorbing material through a foaming process and a freeze-drying method. Taking FD-GOA as a reference, this article focuses on the annealing process and the influence of the thickness and compression of FD-GA on its acoustic absorption performance through the impedance tube method.

2. The preparation of FD-GOA and FD-GA

Figure 1a shows a schematic diagram of the preparation process of FD-GOA and FD-GA. The GO solution was prepared by modified Hummers methods. Measure 100 mL of 12 mg•mL\(^{-1}\) GO solution and add 8 g of 20% alkyl glycoside as the foaming agent. The mixture was stirred at high speed for 5 minutes, changing from black to yellow. Put it in a vacuum freezer for 3 days until it is completely dry. Finally, the FD-GOA is annealed at 200 °C for 4 hours in a muffle furnace to obtain the FD-GA.

Figure 1b shows the physical image of the FD-GOA, and the diameter of the foamed container is 100 mm. After laser cutting, a cylindrical FD-GO with a diameter of 30 mm is obtained for further compression. After the FD-GOA is compressed, it protects and rebounds without collapsing or cracking. In this article, we defined the ratio of the sample height before (\(h_{\text{before}}\)) and after (\(h_{\text{after}}\)) compression as the compression factors (\(n_c\)):

\[
\frac{n_c}{h_{\text{before}}} = \frac{h_{\text{after}}}{h_{\text{before}}}
\]

We prepared FD-GOA and FD-GA with compression factors \(n_c\) of 1, 2, and 3. In addition, FD-GA with thicknesses of 10 mm, 20 mm, and 40 mm are also prepared.

Figure 1. The preparation of FD-GOA and FD-GA. a) Schematic diagram of the preparation of FD-GOA and FD-GA. b) The physical photos of FD-GOA and compression process.

3. Microscopic characterization of FD-GA

Figure 2 shows the low and high magnification SEM images of FD-GA with different degrees of compression. It can be seen from Figure 2a that the original FD-GA is composed of numerous interconnected honeycomb-shaped cavities. The skeleton of the cavity is graphene nanosheets interwoven with each other. As can be seen from Figure 2b, these cavities are all with small holes. The existence of these small holes makes these cavities interconnected. When the sound wave comes in, it will enter the next cavity mainly through these small holes after being consumed. It can be seen from the high magnification SEM image that the cavity size of the initial FD-GA is 100-300 μm. Figure 2c is a low magnification SEM image of a FD-GA with a compression factor \(n_c\) of 2. After compression in the FD-GOA stage, the cavity of the FD-GA is directionally compressed. The entire field of view is no longer a clearly visible honeycomb structure. By observing the high magnification SEM image, the honeycomb structure of the FD-GA with a compression factor \(n_c\) of 2, obviously tends to be flat, and the cavity wall is slightly curled, as shown in Figure 2d. In addition, the cavity wall of the FD-GA is slightly damaged. Figure 2e is a low-magnification SEM of the FD-GA with a compression factor \(n_c\).
of 3. There are no obvious cavity units in the field of view. Figure 2f shows a high-magnification SEM image of the FD-GA. At this time, the graphene wall inside the FD-GA is severely twisted and cracked. Although the graphene nanowall inside has undergone drastic changes, there is no obvious crack in the overall degree of appearance.

Figure 2. SEM image of the FD-GA with different compression factor $n_c$ of 1, 2, and 3. a) High and b) low magnification SEM images of the FD-GA with the compression factor $n_c$ of 1. c) High and d) low magnification SEM images of the FD-GA with the compression factor $n_c$ of 2. e) High and f) low magnification SEM images of the FD-GA with the compression factor $n_c$ of 3.

4. Acoustic absorption properties of FD-GOA and FD-GA

The acoustic absorption performance of FD-GOA and FD-GA here were tested by impedance tube method. Figure 3a shows the acoustic absorption curves of FD-GOA and FD-GA with the thickness of 10 mm. After the annealing process, the acoustic absorption performance of the FD-GOA has been greatly improved in the range of 1000-6000 Hz. Interestingly, the FD-GA has an obvious absorption peak at 4828 Hz, and the maximum acoustic absorption coefficient (AAC) is 96.7%. The excellent acoustic absorption performance of FD-GA is related to its porous structure. When sound waves enter the FD-GA, it causes the air in the cavity to vibrate, converting acoustic energy into mechanical energy. On the other hand, the sound wave generates friction with the graphene nanowall during the propagation process to convert part of the sound energy into heat energy. Of course, the numerous interconnected cavities allow the sound energy to be greatly absorbed. Figure 3b shows the acoustic absorption curves of the FD-GA with different thickness. It has been reported before that the thickness will increase the acoustic absorption performance of lower frequencies, and at the same time the peak acoustic absorption will shift to lower frequencies. The acoustic absorption curve of FD-GA also exhibits similar properties. The thickness of FD-GA increased from 10 mm to 40 mm, and the acoustic absorption peak position also moved from 4828 Hz to 2082 Hz. In addition, the peak width at the
acoustic absorption peak also gradually narrowed. As the thickness of the FD-GA increases, the acoustic absorption performance at lower frequencies has been significantly improved. Therefore, increasing the thickness of the FD-GA is beneficial to improve the acoustic absorption performance of the FD-GA.

Figure 3c shows the acoustic absorption curves of FD-GOA and FD-GA when the compression factor is 2, when the thickness is 10 mm. The acoustic absorption curve of the compressed FD-GA also has an obvious absorption peak, which is related to the increase in tortuosity. The absorption peak of FD-GOA appears at 2102 Hz, while the absorption peak of FD-GA appears at 2947 Hz. Interestingly, the trends of the acoustic absorption curves of the two are similar. Figure 3d shows the acoustic absorption curves of FD-GOA and FD-GA when the compression factor is 3, when the thickness is 10 mm. The absorption curves of both FD-GOA and FD-GA have two absorption peaks, in which the two absorption peaks in the low frequency region are located very close to each other at 1986 Hz and 2093 Hz, while the peaks are basically the same, 98.7% and 97.9%, respectively. The only difference is the narrower peak width of the FD-GOA absorption peak at the lower frequency position. The location of the absorption peaks in the high frequency region differs greatly between the two, with the absorption peak of FD-GA located at 4527 Hz with a peak of 81.8% and the absorption peak of FD-GOA located at 5264 Hz with a peak of 87.6%.

Figure 3. Acoustic absorption properties of FD-GOA and FD-GA. a) Acoustic absorption properties of FD-GOA and FD-GA with the compression factor $n_c$ of 1 and the thickness of 10 mm. b) Acoustic absorption properties of FD-GA with the thickness of 10 mm, 20 mm, and 40 mm. c) Acoustic absorption properties of FD-GOA and FD-GA with the compression factor $n_c$ of 2 and the thickness of 10 mm. c) Acoustic absorption properties of FD-GOA and FD-GA with the compression factor $n_c$ of 3 and the thickness of 10 mm.

5. Conclusion

In this work, porous FD-GOA was prepared by foaming process combined with freeze-drying method, and FD-GA was obtained by further annealing. FD-GOA and FD-GA with different compression factors were obtained by controlled compression of FD-GOA and FD-GA. The acoustic absorption
performance test results showed that the annealing process improved the acoustic absorption performance of FD-GOA. Also, the increase of thickness can effectively improve the acoustic absorption performance of FD-GA at lower frequencies. With the increase of compression, FD-GOA and FD-GA showed similar acoustic absorption properties. However, FD-GOA has poor hydrophobicity and poor elasticity.

Annealing FD-GOA to obtain FD-GA exactly compensates for these defects, making it naturally a new carbonaceous acoustic material with good application prospects.

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