

## **Supporting Information**

# **Hierarchical assembly of tungsten spheres and epoxy composites in three-dimensional graphene foam and its enhanced acoustic performance as a backing material**

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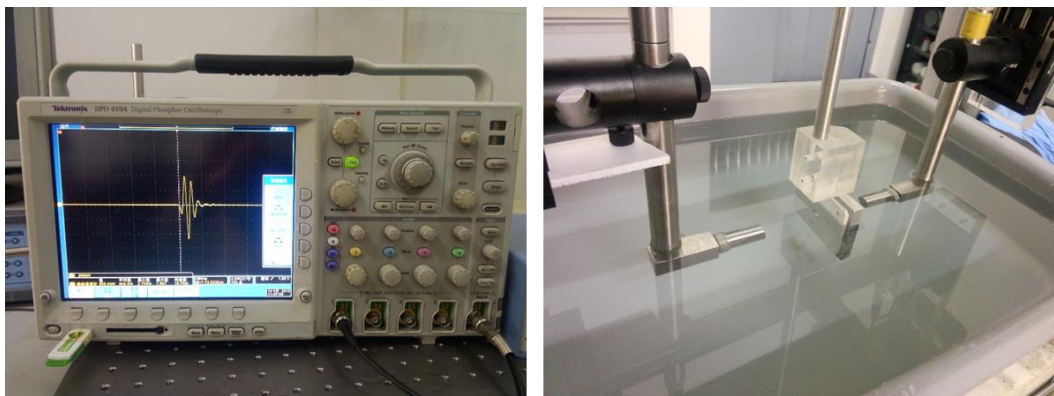
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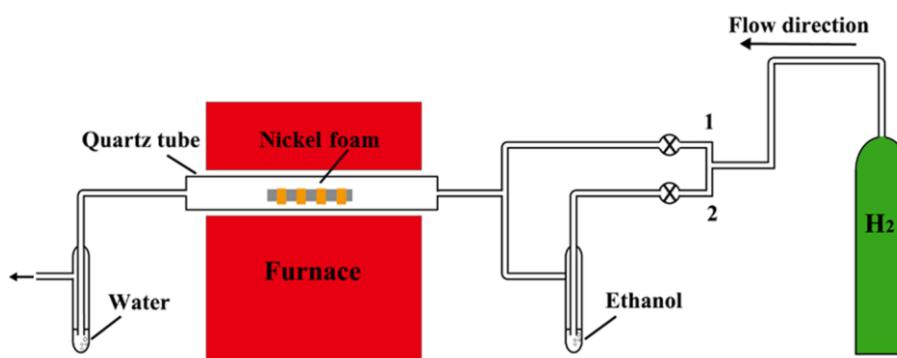
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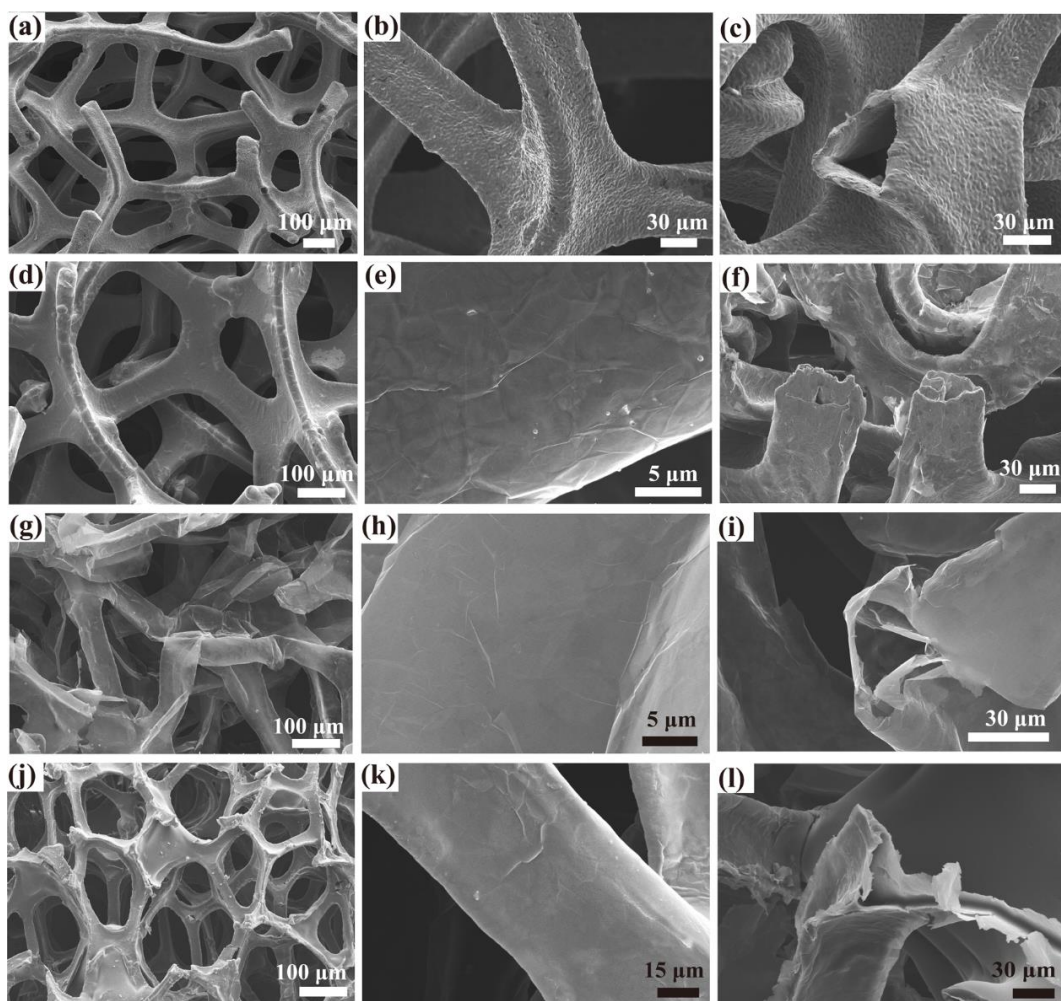
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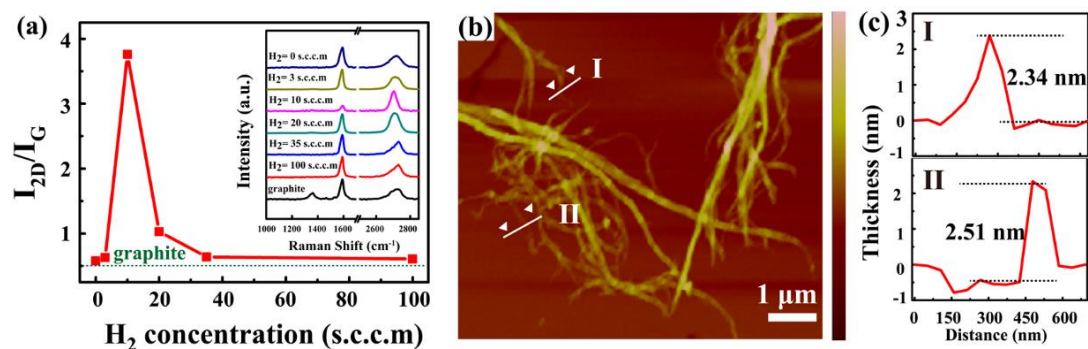
**Figure S1.** Experiments setup for the ultrasonic measurements in present work.



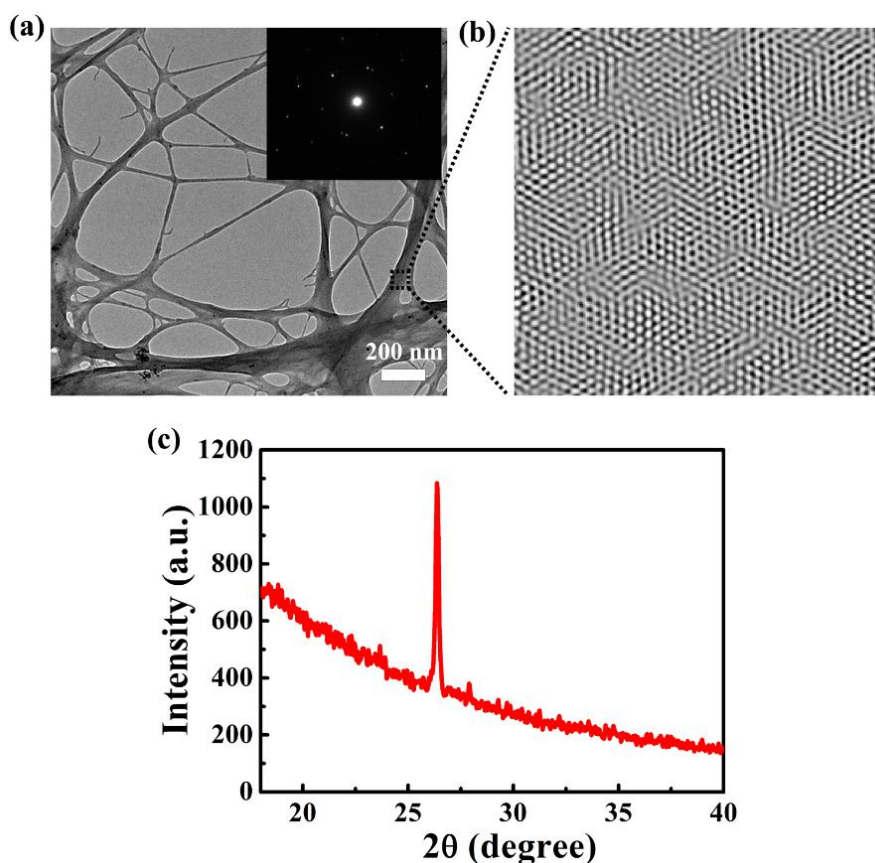
**Scheme S1.** CVD procedure for the preparation of three-dimensional graphene using nickel foam as template. 1 and 2 are two gas valves.



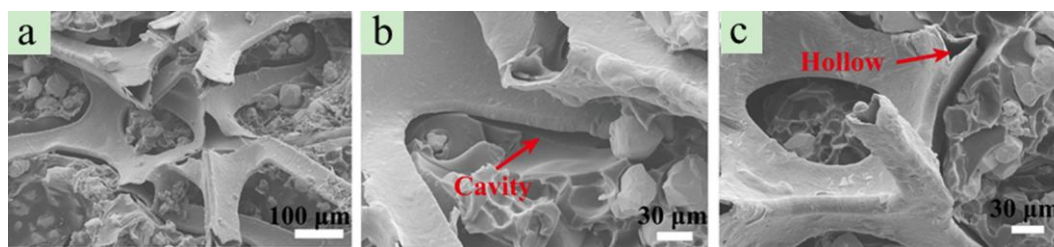
**Figure S2.** a, b, c) SEM images of nickel foam from low to high magnification. d, e, and f) SEM images of nickel foam/graphene from low to high magnification. g, h, and i) SEM images of 3DG from low to high magnification after etching away nickel foam template. j, k, and l) SEM images of 3DG/epoxy from low to high magnification, which is prepared by coating a thin layer of epoxy.



**Figure S3.** a)  $I_{2D}/I_G$  ratio of as-prepare graphene as a function of different amount of  $H_2$  stream. Inset of a is Raman spectra of corresponding samples. b) AFM image of folded graphene after ultrasonication. c) Height profiles in region I and II of b, as indicated by white bars.



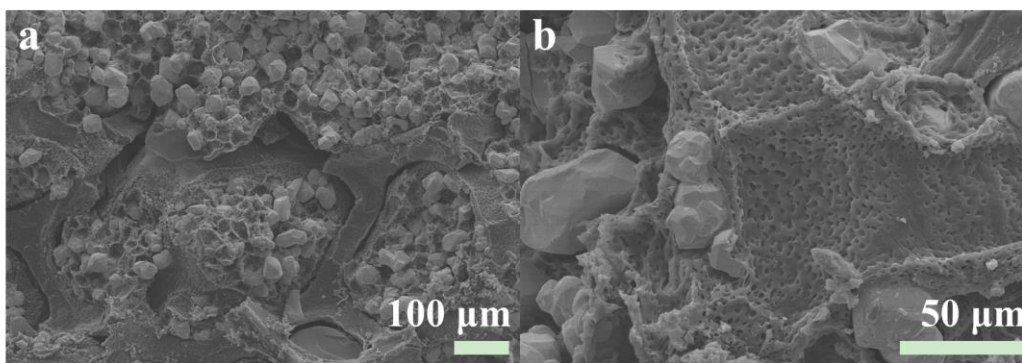
**Figure S4.** a) TEM image of folded graphene after ultrasonication. Inset of a is the corresponding selected area electron diffraction. b) Fourier transform of HRTEM image as marked by dashed square in a. c) XRD spectrum of as-synthesized 3DG.



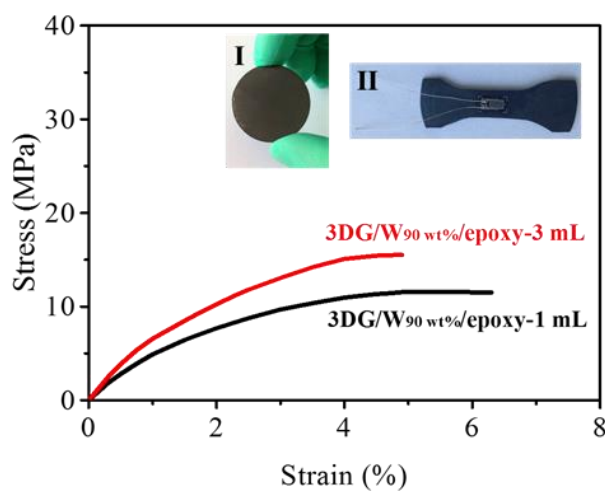
**Figure S5.** a) SEM image of 3DG/W<sub>80 wt%</sub>/epoxy composite film prepared by natural sedimentation. b and c) SEM images showing air cavity and hollow structure of 3DG, respectively.

**Table S1.** Statistical analysis of the acoustic impedance and attenuation of 3DG/W<sub>80 wt%</sub>/epoxy composite films in different runs. The size of W sphere is 20  $\mu\text{m}$ .

Ethanol amount	Acoustic impedance (MRayl)				Acoustic attenuation (dB/cm*MHz)			
	Mean	Standard deviation	Minimum	Maximum	Mean	Standard deviation	Minimum	Maximum
0 mL	10.49	0.22	10.15	10.85	33.8	3.56	28	40
1 mL	9.57	0.68	8.9	10.75	40.16	4.66	32.6	47.5
3 mL	9.47	0.64	8.51	10.25	22.83	3.17	18	29

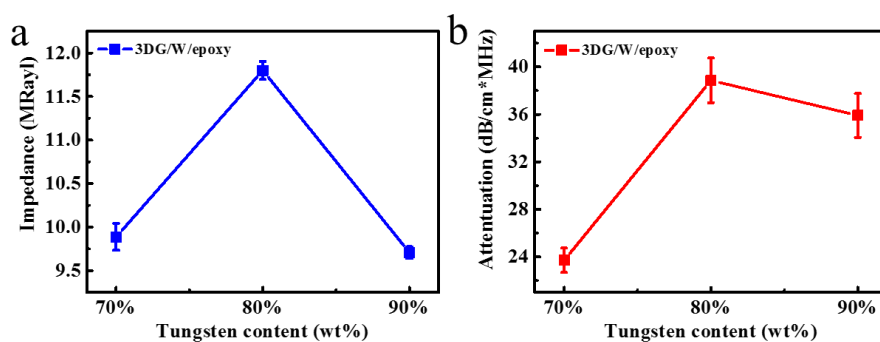


**Figure S6.** a and b) low and high magnification SEM images of 3DG/W<sub>80 wt%</sub>/epoxy composite film prepared by centrifugation-assisted method using 3 mL ethanol.

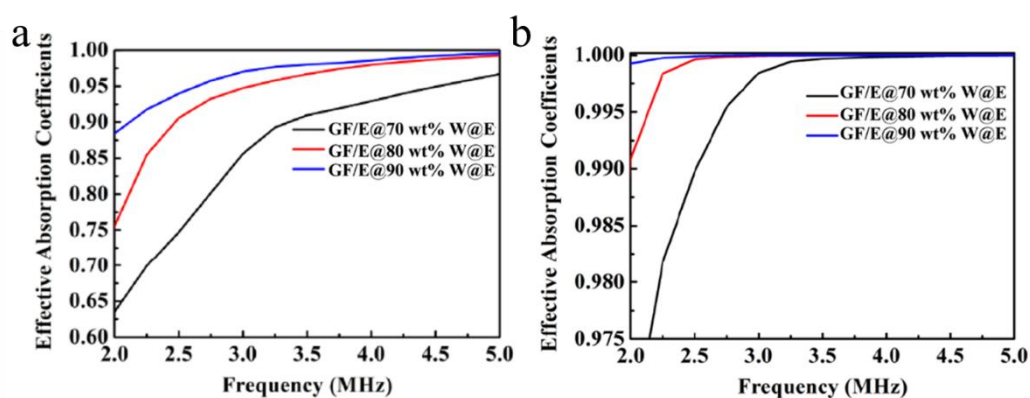


**Figure S7.** Three-point bending stress-strain curves of 3DG/W<sub>80 wt%</sub>/epoxy. The inset I shows the typical 3DG/W<sub>80 wt%</sub>/epoxy film. The inset II shows the dumbbell shape of specimen for stress-strain measurements.

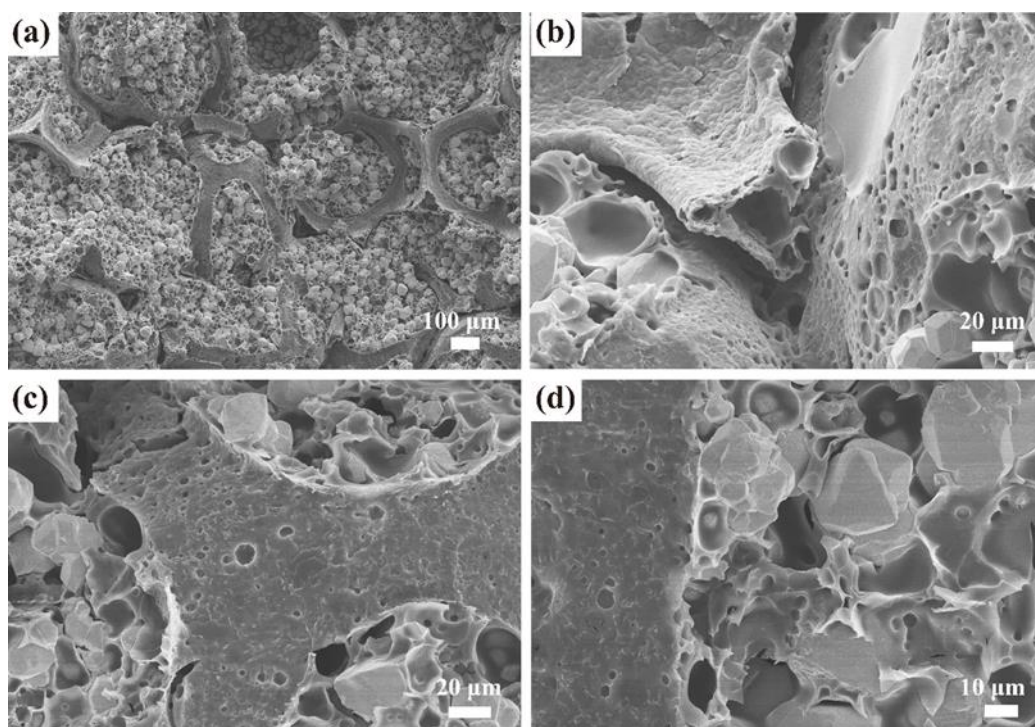




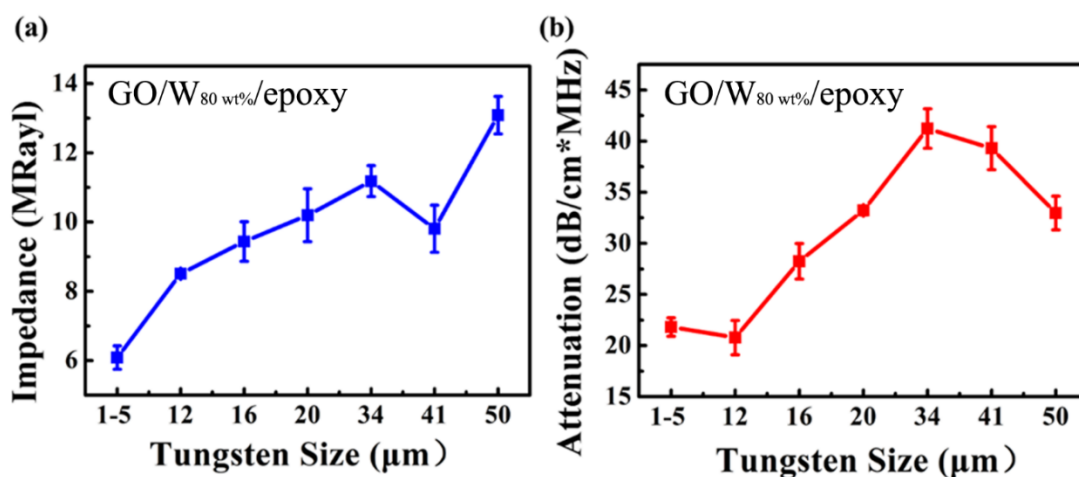
**Figure S8.** a) Acoustic impedance and b) Acoustic attenuation of 3DG/W/epoxy composite films prepared by natural sedimentation as a function of W contents.



**Figure S9.** Absorbance coefficients of 3DG/W/epoxy composite films prepared by natural sedimentation as a function of W contents in a) 3mm and b) 10 mm.



**Figure S10.** SEM images of 3DG/W/epoxy composite films prepared by centrifugation-assisted method from low to high magnification. After centrifugation, the liquid mixture was left to cross-link at 45 °C. Micro-sized air cavities were clear to see on the surface of epoxy coated on graphene, and also can be seen in the mixing phase of W and epoxy in the pores of 3DG.



**Figure S11.** a) Acoustic impedance and b) acoustic attenuation of GO/W80 wt%/epoxy composite films prepared by centrifugation-assisted method. GO content is 3.5 wt% according to our previous work for better acoustic attenuation.