Supporting Information

Construction of hollow nickel-magnesium ferrite decorated nitrogen-doped reduced graphene oxide composite aerogel for high-efficient and broadband microwave absorption

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Experimental section

Characterization

Crystal structure was characterized by X-ray diffraction (XRD, LabX XRD-6000, Japan) with Cu-Kα radiation (λ = 0.154 nm) in the scattering range (2θ) of 10.0−80.0° at a scanning rate of 2.0 °/min. Raman spectra were acquired at room temperature by using a laser confocal Raman spectrometer (Renishaw-2000, UK) in the range of 500.0–2000.0 cm^{-1} with an excitation wavelength of 532.0 nm. Fourier transform infrared (FT-IR) spectra were recorded in the wavenumber range of 500.0–4000.0 cm^{-1} using a Nicolet 380 spectrometer (Thermoscientific, USA). The surface chemical composition and elemental valence states were characterized by X-ray photoelectron spectroscopy (XPS, Thermo ESCALAB 250XI, USA). The micromorphology was observed by a field emission scanning electron microscopy (FESEM, Hitachi-Su8020, Japan) and a field emission transmission electron microscopy (FETEM, FEI-TF20, USA) equipped with the energy dispersive spectrometer (EDS) device.

Electromagnetic parameters including the relative complex permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$) and permeability ($\mu_r = \mu' - j\mu''$) were acquired using the vector network analyzer (VNA, Keysight E5080B, USA) using the coaxial-line method in the frequency range of 2.0–18.0 GHz. The specimens were prepared by uniformly blending the samples with paraffin wax at a filling ratio of 10.0 wt.%, and then pressed the mixtures into a toroidal-shaped ring with an outer diameter of 7.0 mm, an inner diameter of 3.04 mm and a thickness of 2.0 mm. It should be mentioned that the actual power level in dBm units of the incident electromagnetic radiation was used for the measurement of electromagnetic parameters.

The microwave absorption performance of absorbers was evaluated by the reflection loss (RL), which could be calculated by the following equations according to the transmission line theory:[1,2]

$$RL(\mathrm{dB}) = 20\lg\left|\frac{Z_{\mathrm{in}} - Z_0}{Z_{\mathrm{in}} + Z_0}\right| \quad \text{(S1)}$$

$$Z_{\mathrm{in}} = Z_0\sqrt{\frac{\mu_{\mathrm{r}}}{\varepsilon_{\mathrm{r}}}}\tanh\left[j\left(\frac{2\pi fd}{c}\right)\sqrt{\mu_{\mathrm{r}}\varepsilon_{\mathrm{r}}}\right] \quad \text{(S2)}$$

Herein, Z_0 represents the impedance of air, Z_{in} signifies the input impedance of the sample, ε_{r} is the relative complex permittivity, μ_{r} is the relative complex permeability, d is the thickness of the absorber, c is the velocity of light in free space and f is the frequency. Generally, the microwave absorbers with $RL \leq -10.0$ dB are considered to be suitable for practical applications.[1,2]

The computer simulation technology (CST) Studio Suite 2020 was used to simulate the radar cross section (RCS) under far-field response of a microwave absorber. A double layer cuboid model was established on the x-o-y plane which was using perfect electronic conductor (PEC) and composite aerogels samples as the substrate and the coverings, respectively. The frequency of 13.04 GHz was selected as the field detection frequency. Theta and phi were the scattering directions in spherical coordinates, respectively. The directional far-field response was calculated using the RCS of the simulated samples which could be defined by the following formula:[3,4]

$$\sigma\left(\mathrm{dBm}^2\right) = 10\log\left(\frac{4\pi S}{\lambda^2}\left|\frac{E_{\mathrm{s}}}{E_{\mathrm{i}}}\right|\right)^2 \quad \text{(S3)}$$

Where S denotes the area of the simulation model, λ is the wavelength of incident waves. E_{s} and E_{i} refer to the electric field intensity of scattered waves and incident waves at the receiving position, severally.

Results and discussion

The weight and size of cylindrical NRGO-based composite aerogels were measured, and their density was then calculated. From Table S1, the bulk density (ρ) of S1, S2 and S3 are 11.5 mg·cm^{-3}, 9.7 mg·cm^{-3} and 11.3 mg·cm^{-3}, separately. The results manifest that the attained composite aerogels present the extremely low bulk density. Therefore, the prepared NRGO-based composite aerogels could be potential candidates as lightweight microwave absorbers.

Table S1 Typical physical parameters of S1–S3.

Samples	Weight (g)	Height (cm)	Diameter (cm)	Volume (cm^3)	Density (g/cm^3)
S1	0.0808	2.40	1.93	7.02	0.0115
S2	0.0739	2.54	1.95	7.59	0.0097
S3	0.0889	2.68	1.93	7.84	0.0113

As depicted in Fig. S1, the statistical average sizes of $NiFe_2O_4$, $Ni_{0.5}Mg_{0.5}Fe_2O_4$ and $MgFe_2O_4$ microspheres are about 257.1 nm, 263.4 nm and 345.1 nm, respectively.

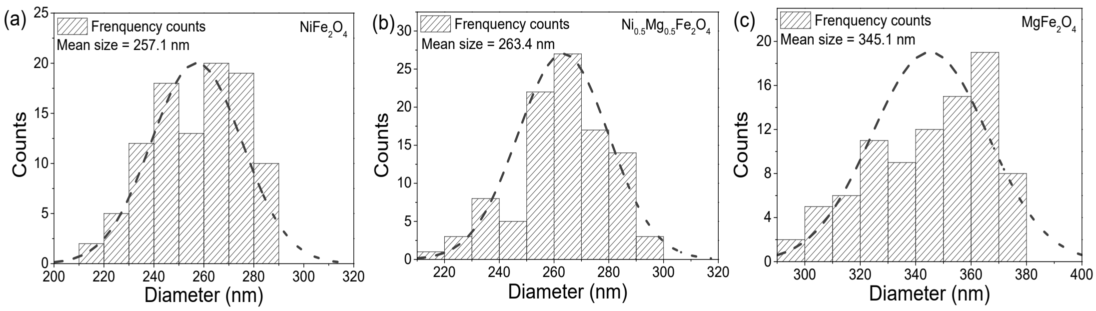

Fig. S1 Histograms of particle size distribution for (a) $NiFe_2O_4$, (b) $Ni_{0.5}Mg_{0.5}Fe_2O_4$ and (c) $MgFe_2O_4$ microspheres.

The relationship between the matching thickness (t_m) and the absorption peak frequency (f_m) can be explained by the quarter-wavelength theory:[5,6]

$$t_m = \frac{n\lambda}{4} = \frac{nc}{4f_m\sqrt{|\varepsilon_r\mu_r|}}(n = 1,3,5,...) \quad \text{(S4)}$$

When t_m and f_m satisfy the above formula, phase cancellation occurs, thus attenuating the incident microwave.[5] As shown in Fig. S2(a), with the increase of t_m, the *RL* peaks of NRGO/hollow $Ni_{0.5}Mg_{0.5}Fe_2O_4$ composite aerogel (S2) shift to the low frequency region. Fig. S2(b) shows a typical $t_m \sim f_m$ curve. Impressively, all t_m^{exp} points are located on the fitted $\lambda/4$ curve. Therefore, the correlation between t_m and f_m is basically determined by the $\lambda/4$ model. As can be seen from Fig. S2(a) and (c), S2 has the strongest *RL* value with the optimal impedance matching. When the thickness is 3.6 mm, it has a minimum *RL* of -56.8 dB at 8.4 GHz, and the corresponding optimal impedance matching value is about 1.0.

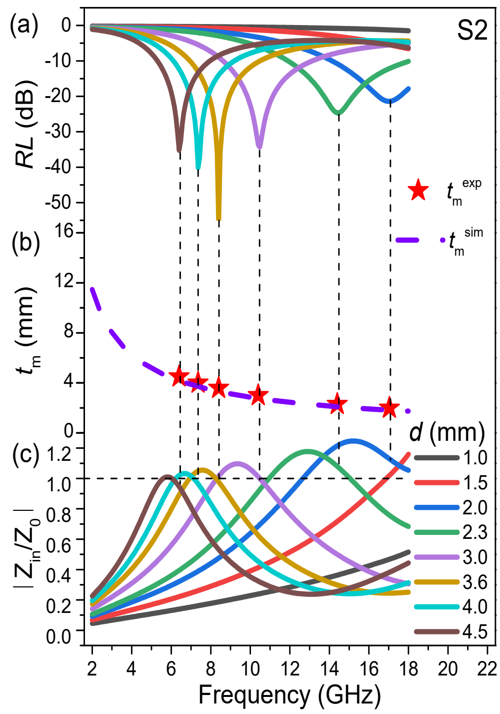

Fig. S2 (a) Frequency-dependent *RL*, (b) simulation of $t_m \sim f_m$ under the $\lambda/4$ model and (c) $|Z_{in}/Z_0|$ *versus* f curves of S2.

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