Supporting Information

Facile Fabrication of Three-Dimensional Lightweight RGO/PPy Nanotube/Fe$_3$O$_4$ Aerogel with Excellent Electromagnetic Wave Absorption Properties

Chunmei Zhang$^{a,1}$, Yujie Chen$^{a,1}$, Hua Li$^{a,b,*}$, Ran Tian$^a$, Hezhou Liu$^{a,b}$

$^a$State Key Laboratory of Metal Matrix Composites, School of Materials Science and Engineering, Shanghai Jiao Tong University, Dongchuan Road No. 800, Shanghai 200240, China.

$^b$Collaborative Innovation Center for Advanced Ship and deep-Sea Exploration, Shanghai Jiao Tong University.

*Corresponding author: Hua Li (email: lih@sjtu.edu.cn)

$^1$These authors contributed equally to this work.
List of the Content

1. The mechanical property characterization and discussion of the GPFA composite.

2. The microwave absorption property characterization and discussion of sample GPFA-S, which possesses crushed GPFA powders with the same mass ratio of about 4.08 wt% as sample GPFA.

3. The magnetic property characterization and discussion of the GPFA composite.

4. The microwave absorption property characterization and discussion of sample GPFA-1 and GPFA-2, which were prepared with different amounts of PPy nanotubes.
The mechanical property of the RGO/PPy nanotube/Fe$_3$O$_4$ aerogel (GPFA) was tested as shown in Figure S1. The results exhibit that it can bear at least 153 times larger than its own weight, thus it has good compression strength.

![Figure S1](image)

**Figure S1** The picture of the mechanical test of the GPFA composite

For comparison, we crushed the GPFA into powders and mixed with wax to obtain sample GPFA-S, which possessed the same mass ratio of about 4.08 wt% as the sample GPFA. The EM wave properties of sample GPFA-S were tested and the results are shown in Figure S2. As shown in Figure S2(a), for sample GPFA-S, the values of $\varepsilon'$ and $\varepsilon''$ decrease with increasing frequency from 5.98 to 3.33, and 2.33 to 1.36 respectively in the range 2.0-18.0 GHz. As reported previously, low conductivity would lead to low permittivity due to the free electron theory, thus the conductivity of sample GPFA-S is decreased compared with GPFA. Moreover, the $\mu'$ and $\mu''$
values of GPFA-S are also lower than GPFA. The loss tangent indicates the inherent dissipation of EM energy for material, and higher values of loss tangent indicate that more EM energy will be consumed. As shown in Figure S2(b), the values of the loss
tangent of samples with smashed GPFA are much lower compared with 3D GPFA. Thus, the samples with smashed GPFA should have much weaker EM wave absorption abilities than the 3D GPFA composite. As shown in Figure S2(c), the maximum RL value of GPFA-S are -15.0 dB at 8.55 GHz with a thickness of 4.5 mm, and the bandwidth of RL below -10 dB are 3.0 GHz in the range of 7.45-10.45 GHz, which is significantly decreased compared with sample GPFA. By using 3D ultralight GPFA as a unique integrated filler rather than dispersing crushed fillers into the matrix, we can prevent RGO sheets from aggregation such that the effective conductive interconnections could form at relatively lower loading. Thus, 3D GPFA could be an ideal candidate for absorbing EM waves.

Figure S2 (a) Real part ($\varepsilon_r'$) and imaginary part ($\varepsilon_r''$) of the complex relative permittivity, real part ($\mu_r'$) and imaginary part ($\mu_r''$) of the complex relative permeability of GPFA-S in the range 2-18 GHz; (b) The loss tangent of GPFA-S; and (c) The reflection loss curves of sample GPFA-S at different thicknesses from 2 to 5 mm in the frequency range of 2 - 18 GHz
The magnetic property of the GPFA composite was tested and the results are shown in Figure S3. As shown in Figure S3(a), the fabricated GPFA can be attracted to the magnet, exhibiting that the composite aerogel possesses good ferromagnetic performance.

The magnetization curve of GPFA is shown in Figure S3(b), and the magnetic field versus moment (M – H) measurements of the sample was performed at room temperature (300 K). It can be found that the saturation magnetic moment is 22.1 emu/g for GPFA, and no remanence and coercivity are observed for the GPFA composite, which indicates that the magnetic GPFA is superparamagnetic. In principle, the superparamagnetic contribution could lead to the improvement of the microwave absorption as described in previous reports. The introduction of Fe₃O₄ nanoparticles can generate some magnetic loss, which can dissipate some incident EM wave energy, and besides, it is also beneficial for better impedance matching between dielectric loss and magnetic loss of the GPFA (Figure 8(c)), which will lead to better EM wave absorption properties.

![Figure S3](image)

**Figure S3** (a) Photograph showing that the GPFA was adsorbed onto a magnet easily, (b) Magnetization curve of the GPFA composite measured at room temperature.
Figure S4 The reflection loss curves of sample (a) GPFA-1, (b) GPFA and (c) GPFA-2 at different thicknesses from 2 to 5 mm in the frequency range of 2 - 18 GHz
Samples with different amounts of PPy nanotubes were prepared and their EM wave absorption properties are compared with GPFA as shown in Figure S4. The fabrication process is the same as sample GPFA and the PPy nanotube amounts are 90 mg for sample GPFA-1 and 150 mg for GPFA-2 respectively. The reflection loss curves of sample GPFA-1, GPFA and GPFA-2 at different thicknesses from 2 to 5 mm in the frequency range of 2 - 18 GHz are shown in Figure S4. It can be found that the maximum RL values of GPFA-1, GPFA and GPFA-2 are -34 dB at 10.1 GHz with a thickness of 3.5 mm, -49.2 dB at 11.8 GHz with a thickness of 3 mm, and -39.9 dB at 5.55 GHz with a thickness of 5 mm, and moreover the bandwidth of RL below -10 dB are 4.65, 6.1, and 2.15 GHz respectively in the range of 8.3-12.95, 9.8-15.9, and 4.65-6.8 GHz respectively. The results exhibit that the sample GPFA exhibits the best EM wave absorption performance.

As the equations (1) and (2) described in the paper, reflection loss is significantly influenced by relative complex permittivity ($\varepsilon_r$) and permeability ($\mu_r$) of test sample. To investigate the possible EM wave absorption mechanism of the above samples, we measured the relative complex permittivity and permeability of the samples, and the variation curves of complex permittivity real parts ($\varepsilon'$) and imaginary parts ($\varepsilon''$), complex permeability real parts ($\mu'$) and imaginary parts ($\mu''$) are show in Figure S5. The real part of the complex permittivity ($\varepsilon'$) represents the storage ability of the EM energy. As shown in Figure S5(a), for sample GPFA-1, GPFA, and GPFA-2, the values of $\varepsilon'$ decrease with increasing frequency from 7.83 to 3.66, 8.66 to 4.02, and 9.90-4.76 respectively in the range 2.0-18.0 GHz. It can be found that the $\varepsilon'$ values increase with increasing contents of highly conductive PPy nanotubes, and the values of the imaginary parts of complex permittivity exhibit almost the same trend as the real parts. Moreover, as shown in Figure S5(b), the values of $\mu'$ and $\mu''$ are similar for all samples and are small when compared with complex permittivity. With the increasing content of PPy nanotubes, the EM wave absorption properties of GPFAs are improved at first and then decrease. This can be attributed that more PPy
nanotubes can lead to better conductivity of GPFAs, thus more incident EM waves are reflected rather than absorbed, so the EM wave absorption performance of sample
GPFA-2 is decreased. As shown in Figure S5(c), the sample GPFA shows better impedance matching between the dielectric loss and the magnetic loss than GPFA-1 and GPFA-2, which leads to best EM wave absorption properties.

Figure S5 (a) Real part ($\varepsilon_r'$) and imaginary part ($\varepsilon_r''$) of the complex relative permittivity, (b) Real part ($\mu_r'$) and imaginary part ($\mu_r''$) of the complex relative permeability and (c) The loss tangent of GPFAs with different PPy nanotube contents in the range 2-18 GHz.