Supplementary Materials

A Strategy Based on Composition Control and Structural Design to Prepare 3DOM RFCo Composites with 3D Ordered Macroporous Structure for Enhanced Electromagnetic Wave Absorption

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2. Experimental Section

2.1 Preparation of 3D Ordered PMMA Templates

The homogeneous poly(methyl methacrylate) (PMMA) microspheres with a particle size of 320 nm were prepared by soap-free emulsion polymerization. Under the protection of argon, 480 mL deionized water and 100 mL MMA were added to a five-neck flask and stirred at 80 °C for 2 h. Then, 20 mL deionized water containing 0.5 g of $K_2S_2O_8$ was quickly added to initiate the polymerization reaction of polymethyl methacrylate. Subsequently, the monodisperse PMMA microspheres were obtained after stirring for further 2 h at 80 °C. Finally, the dispersions were placed in Petri dishes and placed at room temperature for 3 months to obtain 3D ordered PMMA templates with a particle size of 320 nm.

2.2 Preparation of 3DOM RFCo and 3DOM RF

First, 0.048 g NaCO₃ was dissolved in 4 mL of formaldehyde. Then, 3 g resorcinol was added to the solution and the light yellow solution was obtained after stirring for 15 min. Next, 0.005 mol Co(NO₃)₂·6H₂O was dissolved in a light yellow solution to Subsequently, a piece of PMMA template with a size of 2 cm \times 2 cm \times 1 cm was immersed in the precursor solution and removed after standing for 50 min. Then, the impregnated PMMA template was dried at 80 °C for 24 h. Finally, 3DOM RFCo-2 was obtained by heating the dried template in a tube furnace at 700 °C for 2 h under argon atmosphere to remove the PMMA microspheres. 3DOM RF, 3DOM RFCo-1 and 3DOM RFCo-3 were prepared by the same experimental procedure, only that the

addition amounts of $Co(NO_3)_2 \cdot 6H_2O$ were set to 0 mol,0.001 mol and 0.01 mol, respectively.

2.3 Characteriation

The samples of 3DOM RF and 3DOM RFCo were characterized by scanning electron microscopy (SEM, Phenom ProX) and transmission electron microscopy (TEM, JEM-2100F). The elemental mappings were detected by a scanning transmission electron microscope unit (STEM, HITACHI S-5500, 200 kV) with the high-angle annular darkfield (HAADF) detector mode. X-ray diffraction (XRD) patterns of 3DOM RF and 3DOM RFCo-2 samples were obtained on a MiniFlex 600 (Rigaku) X-ray diffractometer. X-ray photoelectron spectroscopy (XPS) analysis was performed on 3DOM RFCo-2 samples using a Thermo Scientific diffractometer. The magnetic properties of 3DOM RFCo composites were measured by a Lake Shore 7404 vibrating sample magnetometer (VSM) at room temperature. Nitrogen adsorption and desorption isotherms of 3DOM RF and 3DOM RFCo-2 samples were measured using a Micromeritics TriStar II 3020 instrument. A vector network analyzer (Keysight E5063A) was used to measure the electromagnetic parameters of the sample in the frequency range of 2-18 GHz. The samples and paraffin wax were mixed and heated at a ratio of 3:17, and then the mixture was pressed into a cylindrical ring with an inner diameter of 3.04 mm and an outer diameter of 7.0 mm.

2.4 Finite Element Simulation

The simulation of 3DOM RFCo-2 sample was carried out in COMSOL software using RF module and the corresponding model was built to investigate the electromagnetic energy loss capability of three-dimensional ordered macroporous structure composite. The solution model can be divided into three layers, i.e., the upper and lower perfect matching layers and the middle research object layer. The perfect match layer is used to absorb the reflected and transmitted electromagnetic waves. The studied object is paraffin wax as the base material and 3DOM RFCo-2 composite as the absorbing material. The electromagnetic wave with a power of 1 W (frequency of 8.0 GHz) is emitted from the source port and enters the MA material along the Z-axis, with the electric field direction in the X-axis. Furthermore, the models created are divided into a rectangular model of the epoxy resin base and a model of the 3DOM RFCo-2 composite with three-dimensional ordered macroporous structure. The length, width, and height dimensions of the rectangular model are 10 µm. The pore size is 1 µm in the model of the 3D ordered macroporous structure.

2.5. RCS Simulation

Calculation of 3DOM RFCo-2 composites using Rader cross sections under the Microwave & RF/OPTICAL module of CST. According to the single-layer homogeneous absorber model, the study object was composed of an absorber layer with a thickness of 3.0 mm and an ideal electrical conductor (PEC) backing plate with a thickness of 4.0 mm. The length and width of the two layers are 400 mm * 400 mm respectively. The electromagnetic parameters of the absorber are measured by a vector network analyzer. The measured electromagnetic parameters are imported into CST, and the electromagnetic parameters of the absorber are obtained by fitting with the dispersion model. Open (add space) boundary conditions are set in all directions. The

field monitor type is RCS. The linear polarized plane wave is used, and the electric polarization propagation direction is along the X-axis. The RCS values can be calculated by using the equation,

$$\sigma(dBm^2) = 10\log\left[\frac{4\pi S}{\lambda^2} \left|\frac{E_S}{E_i}\right|^2\right] \#(S1)$$

where, S is the area of the target object simulation model, λ is the wavelength of the electromagnetic wave, E_s is the electric field intensity of transmitting waves, and E_i is the electric field intensity of receiving waves.



Fig. S1. SEM images of (a) 3DOM RFCo-1 and (b) 3DOM RFCo-3.



Fig. S2. (a) 3D RL map and (b) corresponding 2D contour projection map of 3DOM RF.

Table S1. Comparison of microwave absorption properties of 3DOM RFCo-2 composites with those of representative porous MA materials reported in recent literature.

| MAMs | EAB | Thickness | Ratio | Refs. |
|--|-------|-----------|-------|-----------|
| | (GHz) | (mm) | (wt%) | |
| Co-C/MWCNTs | 4.3 | 1.8 | 25% | 1 |
| Fe ₃ C@Fe/C | 5.1 | 1.5 | 30% | 2 |
| HPCNs-3 | 5.17 | 1.6 | 25% | 3 |
| CoP@HNC | 4.96 | 1.72 | 15% | 4 |
| Ni-SA/HPCF | 5.0 | 2.0 | 10% | 5 |
| NRGO/hollow CoFe ₂ O ₄ | 5.2 | 1.8 | 15% | 6 |
| 3DOM RFC0-2 | 5.28 | 1.8 | 15% | This work |



Fig. S3. The Cole-cole curve for 3DOM RF.



Fig. S4. (a) CST simulation model. (b) 3D radar wave scattering signal. (c) RCS simulated curves of PEC and 3DOM RF samples at different scanning angles.

References

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