Electronic Supplementary Information

For

Constructing magnetic/dielectric loss and phonon/electron thermal carriers γ-Al_2O_3-based yolk-shell microspheres to collaboratively advance microwave absorption and heat conduction

Kang Fu,† Qibin Yao,† Lingling Xu,† Wanyi Zhou,† Zijian Wang,† Yujia Yang,† Guoxiu Tong,*† Xiaojuan Wang,† Wenhua Wu†

†*College of Chemistry and Material Sciences, Key Laboratory of the Ministry of Education for Advanced Catalysis Materials, Zhejiang Normal University, Jinhua 321004, China.*

* **Corresponding Authors:** E-mail: tonggx@zjnu.cn (G.X. Tong); Tel.: +86-579-82282269; Fax: +86-579-82282269.

1. Experiment section

1.1. Specimen characterization

The surface morphologies, element contents, and element mapping of the magnetic YSMSs were studied using a scanning electron microscope (ZEISS GeminiSEM 300) operated at 10 kV and an energy-dispersive X-ray spectrometer (EDS Horiba, EX-250) connected to an SEM. A software (named Nano Measurer) was used to measure the diameters of the biggest and smallest microspheres. The microstructures were observed on a transmission electron microscope (TEM, JEM-2100F, 200 kV). In selected area electron diffraction (SAED) patterns, the diameter of diffraction rings (*r*) was measured for determining the crystallite plane, in which the value of 1/*r* corresponds to the interplanar spacing (*d*). Phase and structure analyses were carried out using an X-ray diffractometer (XRD, D/MAX-IIIA, 40 kV, 40 mA, 10 °/min) with a Cu Kα radiation source ($\lambda = 0.15418$ nm). The Fourier transform infrared (FTIR) spectra were measured at 400–4000 cm^{-1} using a Perkin Elmer 100 Model FTIR spectrometer. The graphitization degree of carbon was investigated using a Raman spectrometer (Renishaw, RM10000). The surface chemical states were identified using X-ray photoelectron spectroscopy (XPS, ESCALAB 250). A software (named Avantage) was used to fit the XPS peaks based on the BE Lookup Table for Signals from Elements and Common Chemical Species. The textural characteristics were recorded using an autosorb iQ instrument (Quantachrome, Florida, USA) after the specimens remained at 200 °C for 10 hours to outgas.

1.2. Property measurement

The conductivity of the YSMSs was tested on a model four-point probe (RTS-8) after they were pressed into wafers (Thickness: ca. 1.0 mm; Diameter: 7.5 mm). The permeability ($\mu_r = \mu' - j\mu''$) and permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$) measurements were carried out using a vector network analyzer (Agilent N5230A) with a coaxial line method on a ring-shaped specimen (Thickness: ca. 3.0 mm; Φ_{out} = 7.0 mm; Φ_{in} = 3.04 mm). The specimen was obtained by mixing the YSMSs with molten paraffin at a loading amount of 10~40 wt.% and pressing the mixture into a ring-shaped mold.

For circular silica films (Thickness: 1.0 mm; Diameter: 4.0 cm) formed by evenly mixing the YSMS powders with silicone oil, outgassing, and curing in a mold, their heat conductivity, thermal diffusivity, and thermal capacity were assessed by a Sweden TPS2500 Hot Disk analyzer with a 5501 probe (Heating power: 160 mW; Heating time: 5 s). The 5501 probe was sandwiched between two silicone films and compacted with two insulated polyurethane foam plates to ensure adequate contact between the silicone films and the probe.

Table S1 Detailed synthesis conditions for the γ-Al_2O_3@Fe_3O_4@C and the γ-Al_2O_3@$FeAl_2O_4$@Fe@C YSMSs.

No.	$Al_2(SO_4)_3 \cdot 18H_2O$ (g)	Urea (g)	PVP (g)	Temperature/time (℃)/(h)	γ-AlOOH precursor (g)	$[Fe^{3+}]$ (mol/L)	Al^{3+}/Fe^{3+} molar ratio	*T*oluene (mL)	T_a (℃)	Annealing time (h)
S1	3.9986	1.4400	0.3000	180/4	4.5000	/	/	/	800	3
S2	3.9986	1.4400	0.3000	180/4	4.5000	/	/	2.0	800	3
S3	3.9986	1.4400	0.3000	180/4	4.5000	0.3333	9:1	2.0	800	3
S4	3.9986	1.4400	0.3000	180/4	4.5000	0.7500	8:2	2.0	800	3
S5	3.9986	1.4400	0.3000	180/4	4.5000	1.2857	7:3	2.0	800	3
S6	3.9986	1.4400	0.3000	180/4	4.5000	0.7500	8:2	2.0	500	3
S7	3.9986	1.4400	0.3000	180/4	4.5000	0.7500	8:2	2.0	600	3
S8	3.9986	1.4400	0.3000	180/4	4.5000	0.7500	8:2	2.0	700	3

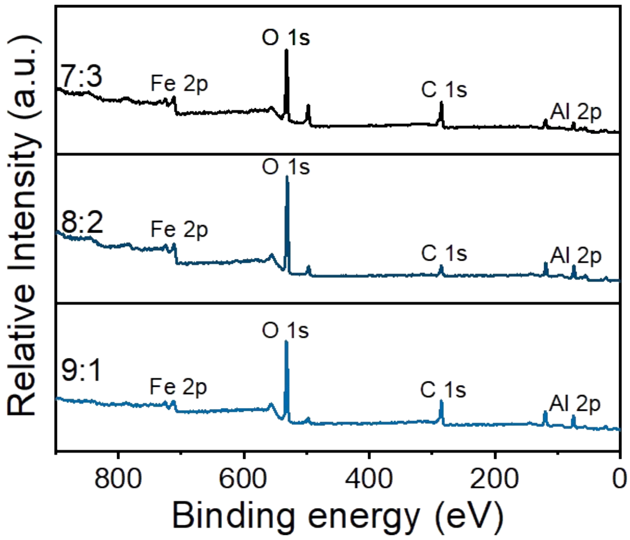

Fig. S1. Survey XPS spectra of the annealed products formed at different Al^{3+}/Fe^{3+} molar ratios.

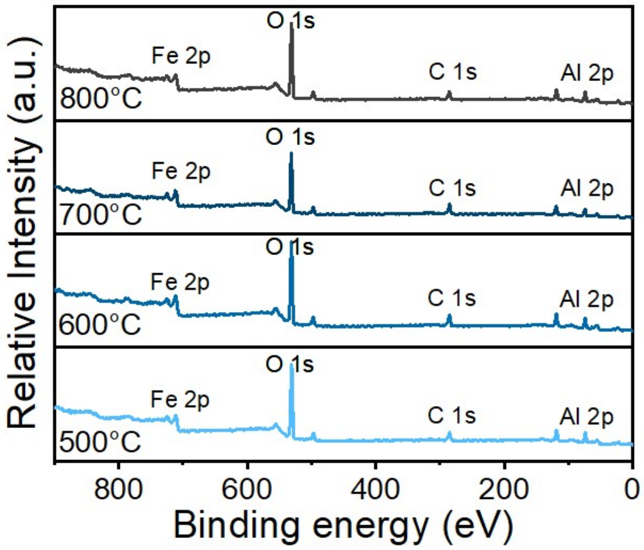

Fig. S2. Survey XPS spectra of the annealed products formed at different T_a.

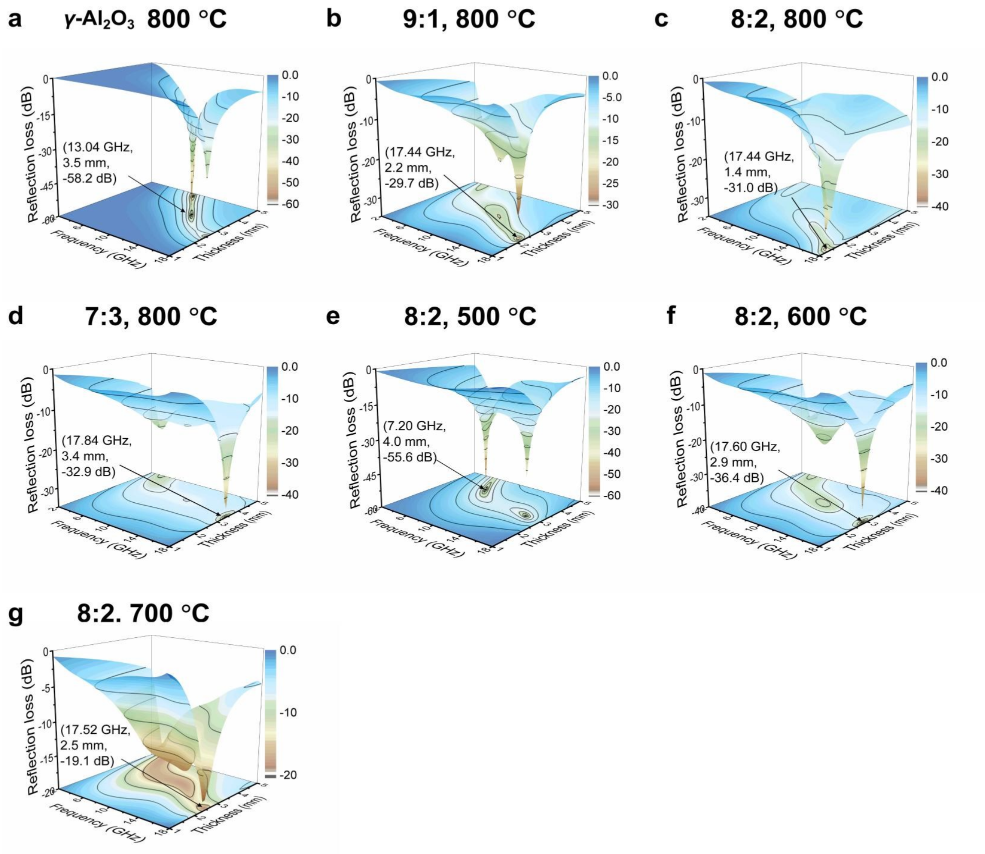

Fig. S3. Frequency characteristics: (a–g) 3D RL plots of the paraffin composites containing γ-Al_2O_3 and γ-Al_2O_3@$FeAl_2O_4$@Fe@C YSMSs obtained at different β and T_a.

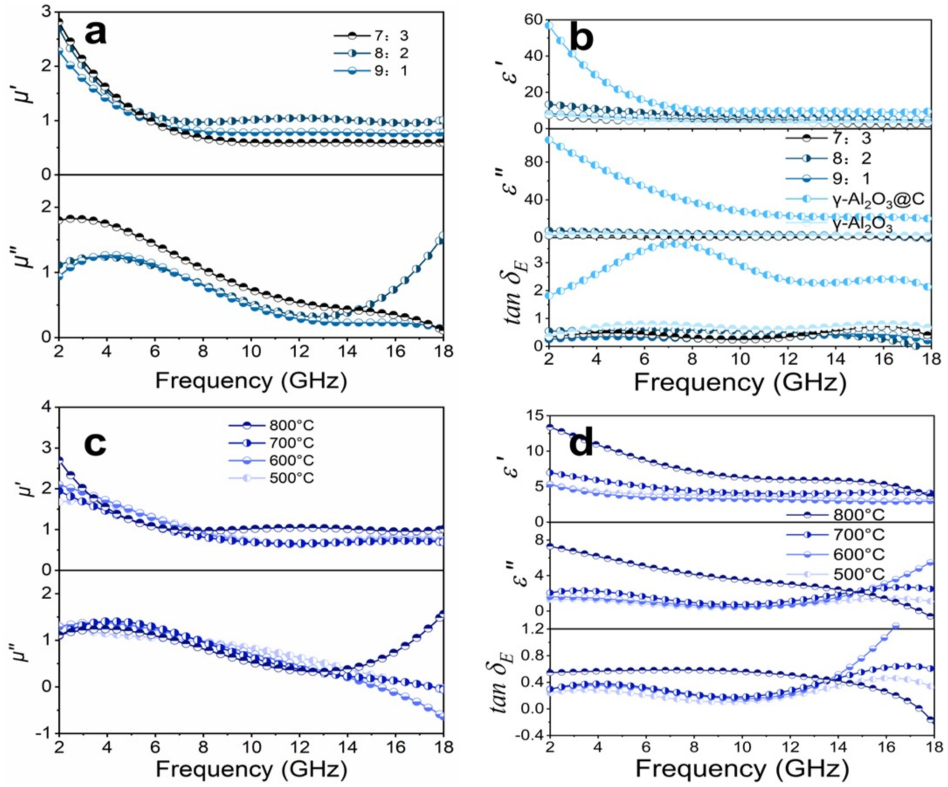

Fig. S4. Frequency characteristics of pure γ-Al_2O_3, γ-Al_2O_3@C, γ-Al_2O_3@$FeAl_2O_4$@Fe@C, and γ-Al_2O_3@Fe_3O_4@C YSMSs obtained at different β and T_a: (a, c) the relative complex permeability, (b, d) the real part (ε') and imaginary part (ε'') of relative complex permittivity and dielectric loss.

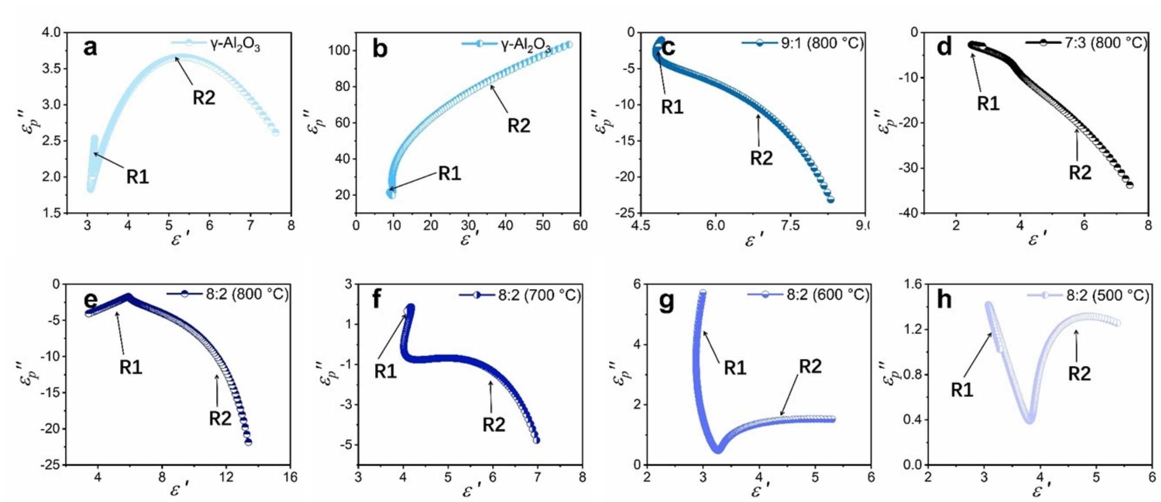

Fig. S5. Cole-Cole curves (a–h) of γ-Al_2O_3, γ-Al_2O_3@C, γ-Al_2O_3@$FeAl_2O_4$@Fe@C, and γ-Al_2O_3@Fe_3O_4@C YSMSs obtained at different β and T_a.