† Electronic Supplementary Information (ESI) available:

1 Materials and methods

1.1 Materials

Anhydrous ethanol, hexane, ferric chloride, and manganese chloride were purchased from Sinopharm chemical Reagent Co., Ltd (Shanghai, China). Sodium oleate was obtained from Aladdin Industrial Corporation (Shanghai, China). Halloysite was supplied by Zhongwei mineral materials Corporation, Xingtai, China. All materials were used as received without further purification.

1.2 Preparation of $MnFe_2O_4$/halloysite nanotubular encapsulates

We used "oleate" method [1,2] to prepare ferrite nanoparticles. In a typical procedure, 40 mmol $FeCl_3$, 20 mmol $MnCl_2$, and 160 mmol sodium oleate were added into mixed solvents of 100 mL H_2O, 100 mL ethanol and 200 mL hexane. The mixture was heated and refluxed at 70 ^oC for 4 h. When the reaction completed, the mixed $Mn^{2+}Fe^{3+}$–oleate complex was extracted and was washed three times with distilled water in a separatory funnel. The obtained oleate complex was subsequently heated at 70 ^oC to remove the residual ethanol and hexane, and then 110 ^oC to remove water.

10 g halloysite was added into the oleate complex and mixed for different times (0 h, 1 h, 4 h, 8 h, 24 h). The mixture was then calcinated at 450 ^oC for 2 h. After cooled to room temperature, $MnFe_2O_4$/halloysite nanotubular encapsulates (HF-1) were obtained. In addition, we added 2 g deionized water into the oleate complex during mixing with halloysite to fabricate magnetic halloysite nanocomposites with $MnFe_2O_4$ nanoparticles on the outside of halloysite nanotubes. The obtained sample was named HF-2.

1.3 Characterizations

X-ray diffraction (XRD) data were collected by a D/max 2550 X-ray diffractometer (Riguaku, Japan) with Cu Kα radiation (λ=0.15406 nm) from 3^o to 75^o. The operation voltage and current were maintained at 40 kV and 34 mA, respectively. Morphologies of samples were examined by a JEM-2010 transmission electron microscope (TEM) (JEOL, Japan), at an accelerating voltage of 200 kV. The magnetic measurements were performed by an MPMS-XL-5 SQUID magnetometer (Quantum

Design, USA) at room temperature with applied magnetic field between -20000 Oe and 20000 Oe. The relative complex permittivity and permeability values of samples were determined by a HP8720ES vector network analyzer (Agilent, USA) using T/R coaxial line method at electromagnetic wave frequency of 8-18 GHz and thickness of 2 mm. The samples and paraffin were mixed in a toroidal shape with inner diameter of 3 mm and outer diameter of 7 mm. The volume fractions for all samples are 30%. The relative complex permittivity ($\varepsilon=\varepsilon'-j\varepsilon''$) and permeability ($\mu=\mu'-j\mu''$) were calculated from the measured T/R coefficients. The measurement errors are less than 10% when $\varepsilon' < 15$. The reflection loss values of absorbers were evaluated from the measured complex permittivity and permeability values by using the following equations:

$$Z_{in} = Z_0\sqrt{\frac{\mu}{\varepsilon}}\tanh[j\frac{2\pi fd}{c}\sqrt{\mu\varepsilon}] \quad (1)$$

$$RL(dB) = 20\log\left|\frac{Z_{in}-Z_0}{Z_{in}+Z_0}\right| \quad (2)$$

where Z_{in} and Z_0 are the impedance of absorber and air, respectively. ε and μ are the complex permittivity and permeability of absorber, respectively. f is the frequency of electromagnetic wave. d is the thickness of the absorber. c is the velocity of light.

2 Supporting figures and tables

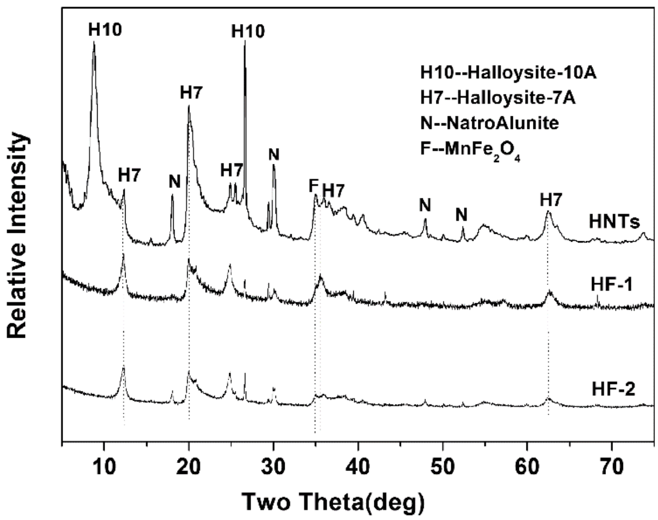

Fig. S1 XRD patterns of natural halloysite (HNTs), $MnFe_2O_4$/halloysite nanotubular encapsulates (HF-1), and $MnFe_2O_4$/halloysite nanocomposites (HF-2).

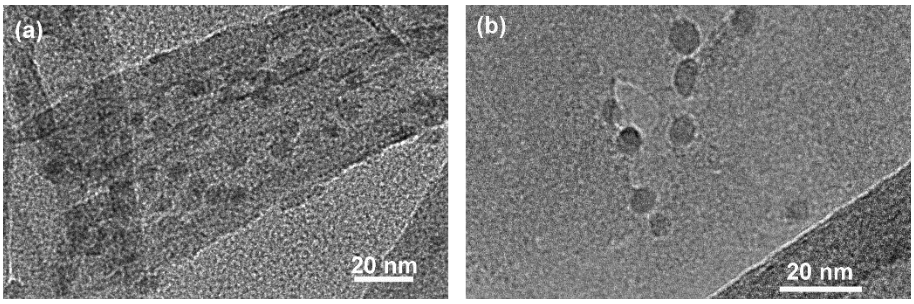

Fig. S2 TEM images of $MnFe_2O_4$/halloysite nanocomposites (HF-2) with ferrite nanoparticles located the outer surface of halloysite nanotubes.

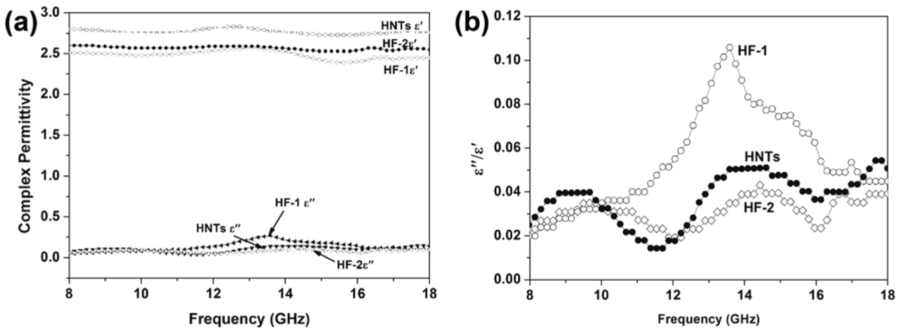

Fig. S3 Frequency dependence of (a) relative complex permittivity, and (b) dielectric loss tangents of natural halloysite (HNTs), $MnFe_2O_4$/halloysite nanotubular encapsulates (HF-1), and $MnFe_2O_4$/halloysite nanocomposites (HF-2).

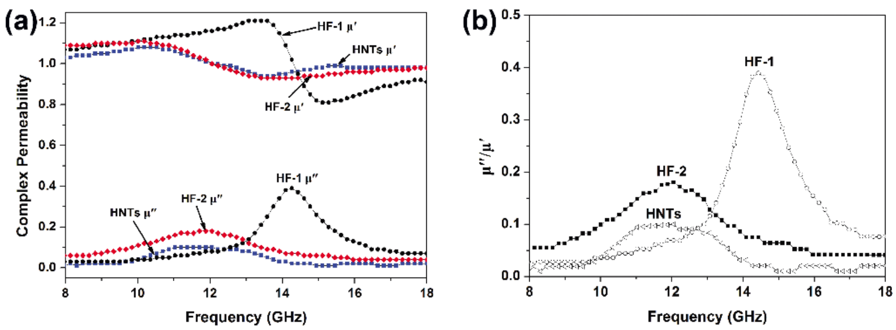

Fig. S4 Frequency dependence of (a) relative complex permeability, and (b) magnetic loss tangents of natural halloysite (HNTs), $MnFe_2O_4$/halloysite nanotubular encapsulates (HF-1), and $MnFe_2O_4$/halloysite nanocomposites (HF-2).

Table S1 BET surface area and pore volumes of natural halloysite (HNTs), $MnFe_2O_4$/halloysite nanotubular encapsulates with different contacting times (HF1-x) and $MnFe_2O_4$/halloysite nanocomposites (HF-2).

Samples	BET surface area/m^2 g^{-1}	Total pore volume/ cm^3 g^{-1}	Mesopore Volume /$cm^3 g^{-1}$	Micropore Volume /cm^3 g^{-1}	Average pore size /nm
HNTs	47.8	0.314	0.221	0.020	2.63
HF1-0	48.8	0.360	0.265	0.020	2.95
HF1-1	45.2	0.298	0.205	0.019	2.64
HF1-4	47.3	0.321	0.223	0.020	2.72
HF1-8	45.2	0.305	0.220	0.019	2.69
HF-2	51.6	0.373	0.268	0.021	2.89

3 Supporting references

1 N. Bao, L. Shen, Y. Wang, P. Padhan, A. Gupta, *J. Am. Chem. Soc*, 2007, **129**, 12374-12375.

2 J. Park, K. An, Y. Hwang, J. Park, H. Noh, J. Kim, J. Park, N. Hwang, T. Hyeon, *Nat. Mater*, 2004, **3**,891-895.