Reduced Graphene Oxide Modified Mesoporous FeNi Alloy/Carbon Microspheres for Enhanced Broadband Electromagnetic Wave Absorber

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ESI-1: Preparation of graphene oxide (GO)

Natural graphite (3.0 g), $K_2S_2O_8$ (2.8 g), P_2O_5 (2.5 g) and H_2SO_4 (12 mL) were added into a 50 mL conical flask, and then the mixture was maintained at 80 ^oC and stirred for 4.5 h. After cooling to room temperature, the solution was diluted with 500 mL deionized water and the suspension was filtered and washed with the deionized water until the filtrate was neutral. The precipitate was oven-dried at 60 ^oC for 24 h. Then, the pretreated graphite powders were added to 120 mL of H_2SO_4 (98%) at 0 ^oC, and continue to add slowly 15 g $KMnO_4$ to above solution. The mixture was carefully increased to 35 ^oC and stirred for 2 h. Subsequently, 250 mL of deionized water was added to the mixture and stirred for 2 h. The solution was diluted with 700 mL of deionized water and 20 mL of H_2O_2 was slowly dropped into the solution. Then, the mixture was centrifuged and washed with hydrochloric acid solution, acetone and ultrapure water, respectively, and graphite oxide was obtained. Finally, to prepare graphene oxide, graphite oxide was sonicated in water for 4 h and freezed-dried about 2 days.

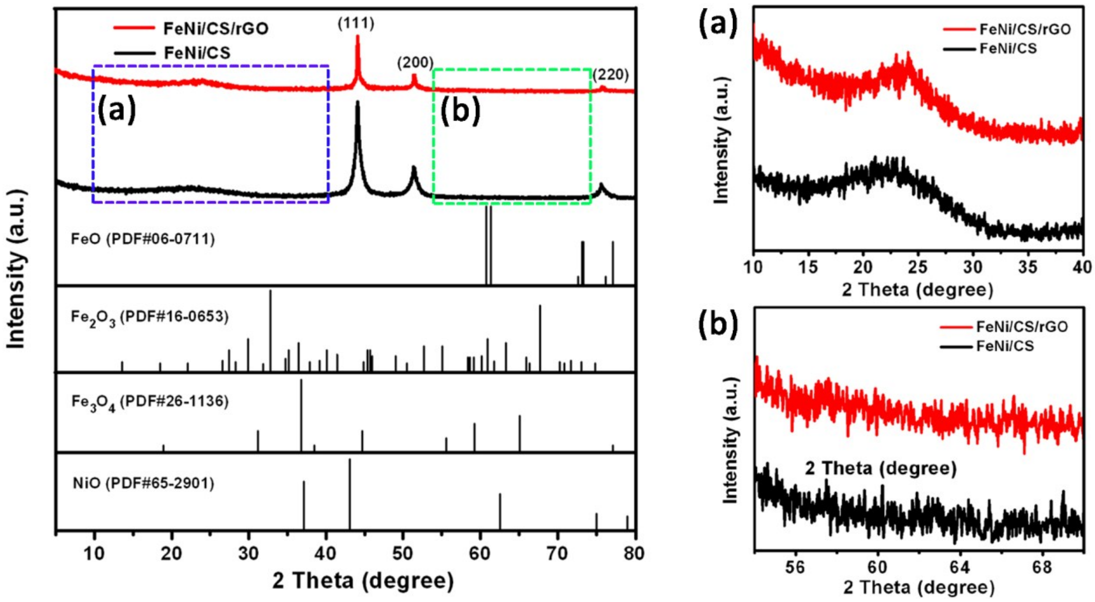

Fig. S1. Comparative XRD patterns of FeNi/CS composites, FeNi/CS/rGO composites, FeO, Fe_2O_3, Fe_3O_4 and NiO. (a) XRD patterns with an expanded region of 2θ = 10°-40°. (b) XRD patterns with an expanded region of 2θ = 54°-70°.

The XRD result is further checked through the comparative diagram of FeNi/CS composites, FeNi/CS/rGO composites, FeO, Fe_2O_3, Fe_3O_4 and NiO (Fig. S1). We display the XRD patterns with an expanded regions of 2θ = 10°-40° and 2θ = 54^{o}-70^{o} in Fig. S1(a) and Fig. S1(b), respectively, which shows no signal of metal oxide peaks. This result means that the surface oxidation of metastable Fe/Ni(zero) when exposed to air has not affected the majority composition of FeNi alloy in the bulk phase. That is to say, FeNi/CS and FeNi/CS/rGO are composite materials containing FeNi alloy and carbon-based materials.

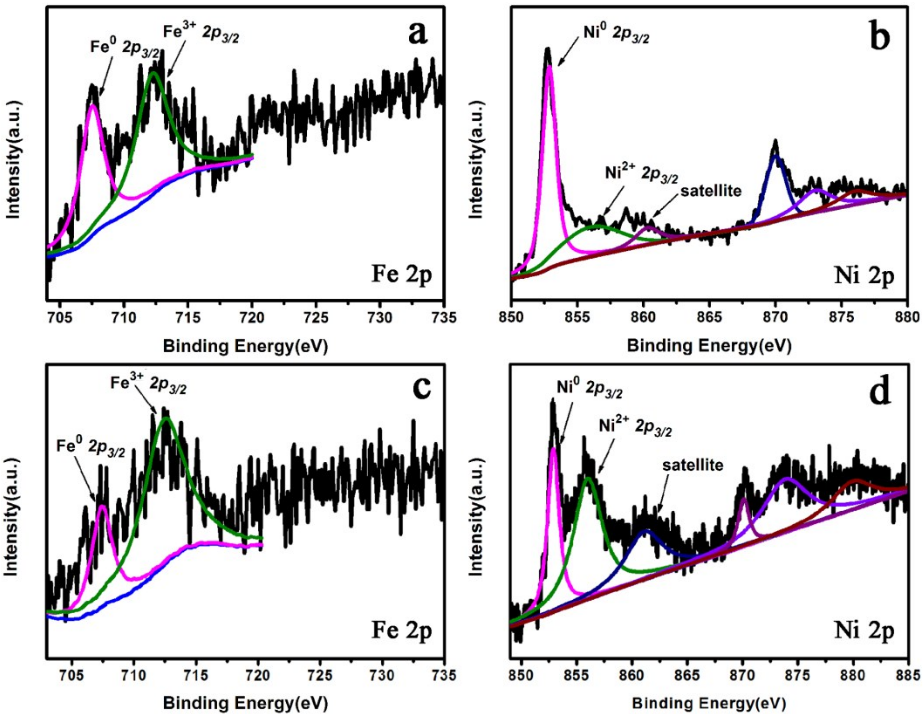

Fig. S2. XPS spectrum of (a) Fe 2p and (b) Ni 2p of FeNi/CS; (c) Fe 2p and (d) Ni 2p of FeNi/CS/rGO composite.

ESI-2:Preparation of pure FeNi alloy nanoparticles

40 mL ethanol and 60 mL deionized water were used for the mixture solvent. The 7.5 mmol $FeSO_4·7H_2O$ and 22.5 mmol $NiCl_2·6H_2O$ were dissolved into the above-mentioned mixed solution. Then, the mixture was maintained at 80 ^{o}C and kept for 30 min. Afterwards, 5 M NaOH solution was drop-wise added to the solution until it reached a pH of ca. 11. Subsequently, 13 mL hydrazine hydrate was added and kept for 30 min. The precipitation were collected by magnetic separation and washed with deionized water/ethanol. Finally, the sample was dried at 80 ^{o}C in the oven overnight.

Table S1. The various weight fractions in the FeNi/CS and FeNi/CS/rGO composites based on the CHNS elemental analysis.

Samples	***FeNi fraction (wt%)***	***C-based fraction (wt%)***	
		$C_{glucose}$[a] ***FeNi:$C_{glucose}$***	***rGO*** ***FeNi:rGO***
FeNi/CS	53.44	46.56 1.15	-----
FeNi/CS/rGO	43.41	37.82 1.15	18.77 2.3

[a] $C_{glucose}$, the fraction of carbonized product of glucose in the composites.

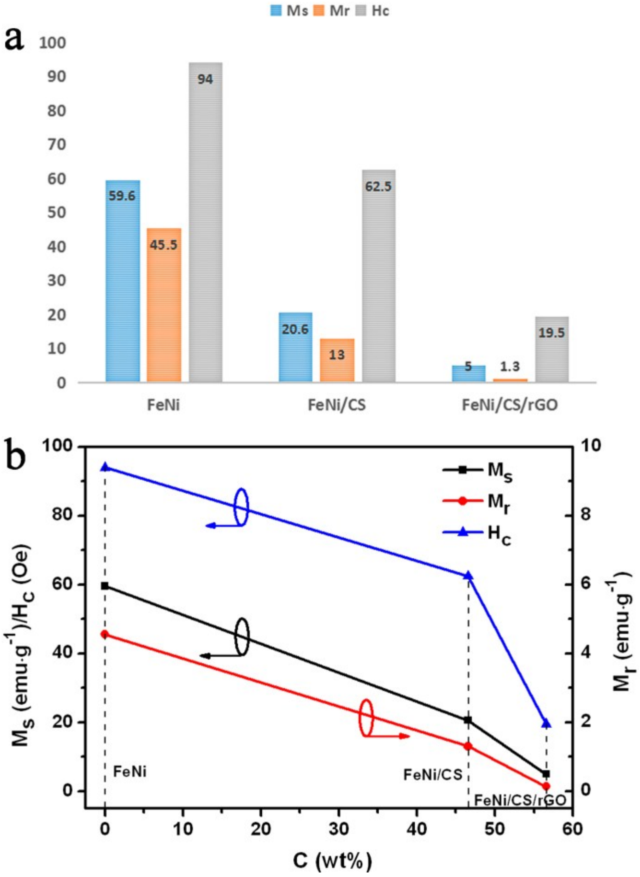

Fig. S3. (a) Histogram of M_s, M_r and H_c values for FeNi, FeNi/CS and FeNi/CS/rGO composite (Note: the M_r values are expanded 10-fold to be easily observed); (b) M_s, M_r and H_c as a function of C content of FeNi, FeNi/CS and FeNi/CS/rGO composite.

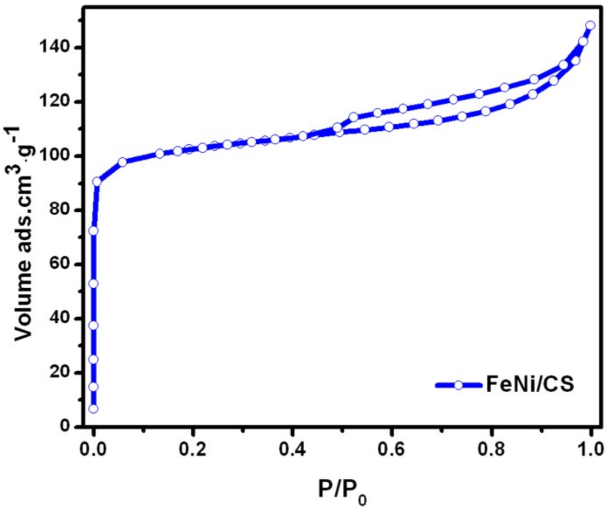

Fig. S4. Nitrogen adsorption-desorption isotherm of FeNi/CS.

To investigate the pored structure of as-obtained magnetically carbon material, nitrogen adsorption-desorption isotherm of FeNi/CS is shown in Fig. S4. Typical IV isotherm with a H2-typical hysteresis loop (P_0>0.4) is found for FeNi/CS composite, suggesting that magnetically carbon material possesses representative mesoporous structure. The calculated Brunauer−Emmett−Teller (BET) surface area and pore volume of FeNi/CS composite are 315.12 $m^2 \cdot g^{-1}$ and 0.148 $cm^3 \cdot g^{-1}$, respectively. These porous may effectively enhance surface areas and capture microwaves in favour of improving EM absorption capacity of material.