Supporting Information for

NiFe₂O₄@ Nitrogen-Doped Carbon Hollow Spheres with highly efficient and recyclable adsorption of tetracycline hydrochloride

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1. Characterizations

A D/Max-RB X-ray diffractometer (Rigaku, Japan) was used to obtain X-ray diffraction (XRD) patterns. TEM images were obtained on a transmission electron microscope (JEM-2100F, JEOL, Japan) with an accelerating voltage of 200 kV. FESEM images were recorded on a field emission scanning electron microscope (JSM-7500, JEOL, Japan) with an accelerating voltage of 15 kV. Chemical states of elements were measured on an X-ray photoelectron spectroscopy (XPS) equipped with an ultra-high vacuum VG ESCALAB 210 electron spectrometer. Fourier transform infrared spectra (FTIR) were measured on an IR Affinity-1 FTIR spectrometer (Shimadzu, Japan). N_2 adsorption-desorption isotherms were measured on a Micromeritics ASAP 3020 equipment (USA). All as-synthesized samples were degassed at 180 °C for 5 h prior to adsorption measurements. Specific surface area was determined by the multipoint Brunauer–Emmett–Teller (BET) method. The magnetization curve was conducted on Quantum Design MPMS-7 SQUID magnetometer at 300 K under varying magnetic field.

2. Adsorption experiment

Table 1. The corresponding physicochemical properties of prepared samples.

Sample	Average crystal size [nm] (standard deviation)	S_{BET} [m^{2}g^{-1}]	Pore volume [cm^{3}g^{-1}]
$NiFe_2O_4$	6.46	21.85	0.04
$NiFe_2O_4$/NCHS(before)	5.34	335.9	0.68
$NiFe_2O_4$/NCHS(after)	3.38	268.8	0.22

Table 2. Pseudo-first-order and pseudo-second-order kinetic parameters of the as-prepared samples.

Samples	$q_{e,exp}$ (mg.g^{-1})	Pseudo-first-order model			Pseudo-second-order model		
		$q_{e,cal}$ (mg.g^{-1})	k_1 (×10^{-2}min^{-1})	R^2	$q_{e,cal}$ (mg.g^{-1})	K_2 (×10^{-2}g.mg^{-1}·min^{-1})	R^2
NiFe	29.11	29.33	13.68	0.890	29.96	1.60	0.989
NiFe- NCHS	41.28	27.45	3.39	0.891	41.84	3.44	0.999

Table 3. Parameters of Langmuir isotherm and Freundlich isotherm for TC adsorption on NiFe$_2$O$_4$/NCHS.

Samples	Langmuir model			Freundlich model		
	q_{max} (mg.g^{-1})	K_L (L mg^{-1})	R^2	K_F (mg/g)(L/mg)$^{1/n}$	n	R^2
NiFe- NCHS	271.739	0.048	0.996	32.478	2.258	0.960

Table 4. The maximum adsorption capacity for TC by different adsorbents for comparison

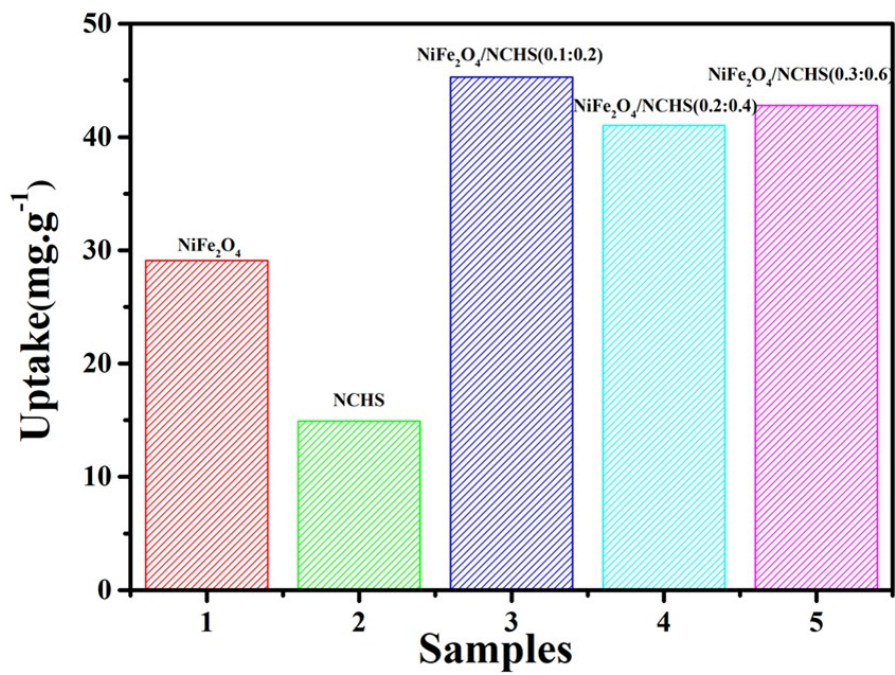| Samples	q_{max} (mg/g)	References
Mag@ZnO-Co$_3$O$_4$	128	1
graphene oxide	212	2
GO-MPs	39.1	3
Ni nanoparticles/silica (Ni NPs/SiO$_2$)	381.3	4
MWCNT	269.54	5
activated sludge	91	6
NHCS-NiFe	271.739	This work

Figure S1. Adsorption of TC by magnetic composites with different concentration of
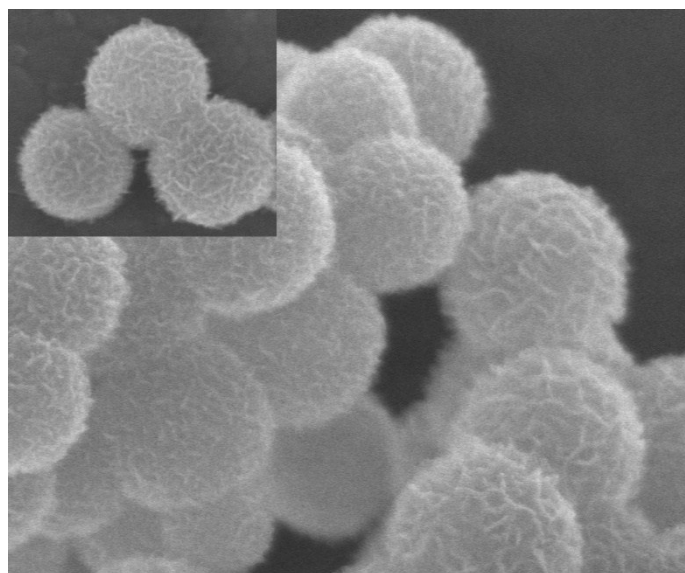
raw materials.

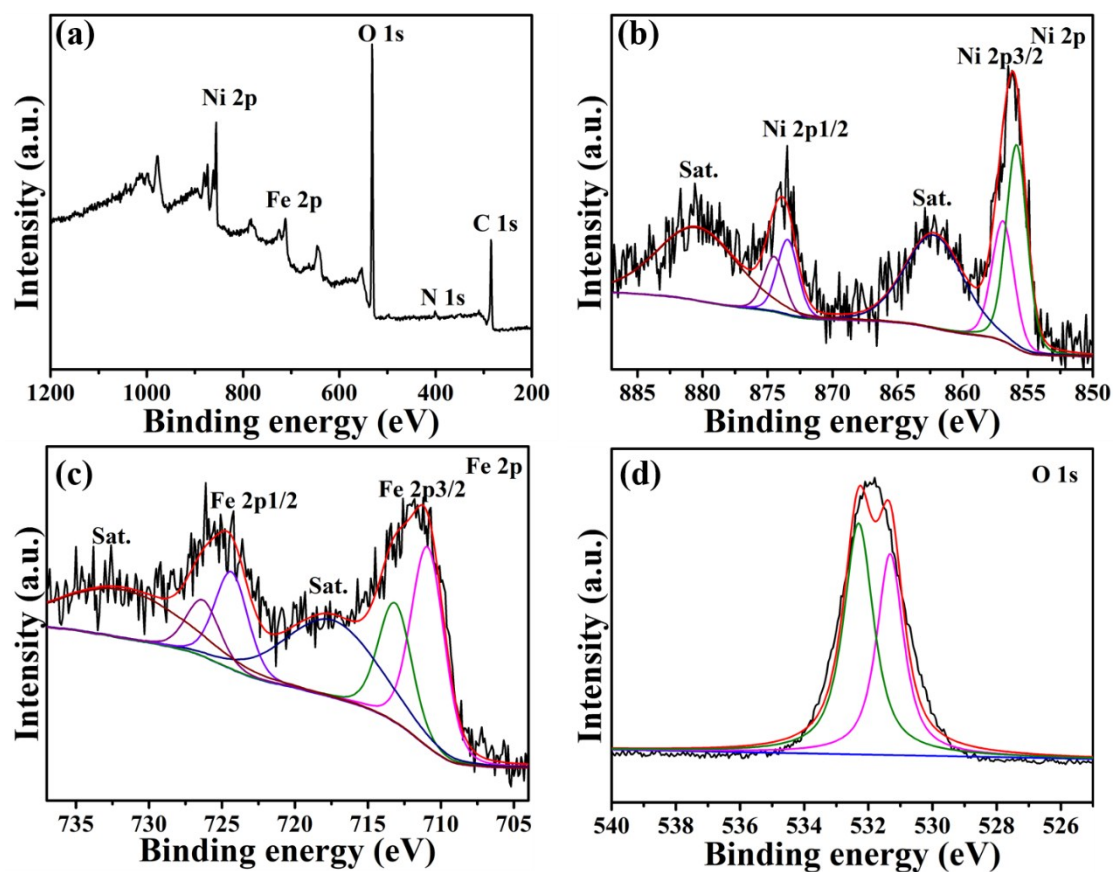**Figure S2.** The SEM images of NiFe$_2$O$_4$/C samples after absorption.

Figure S3. X-ray photoelectron spectroscopy (XPS) spectra of (a) the survey spectrum, (b) Ni 2p, (c) Fe 2p, (d) O 1s for NiFe$_2$O$_4$/NCHS composite after adsorption.
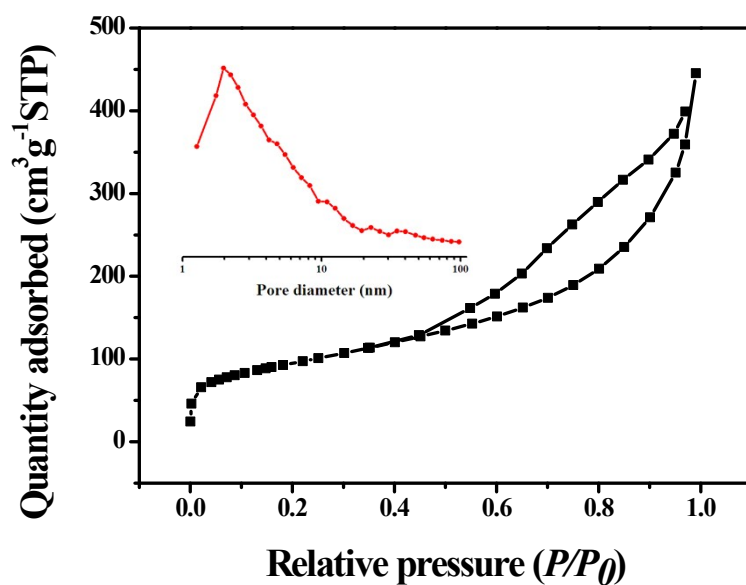

Figure S4. N$_2$ adsorption/desorption isotherm and the corresponding pore size distribution (inset) of the adsorbed NiFe$_2$O$_4$/NCHS composite.
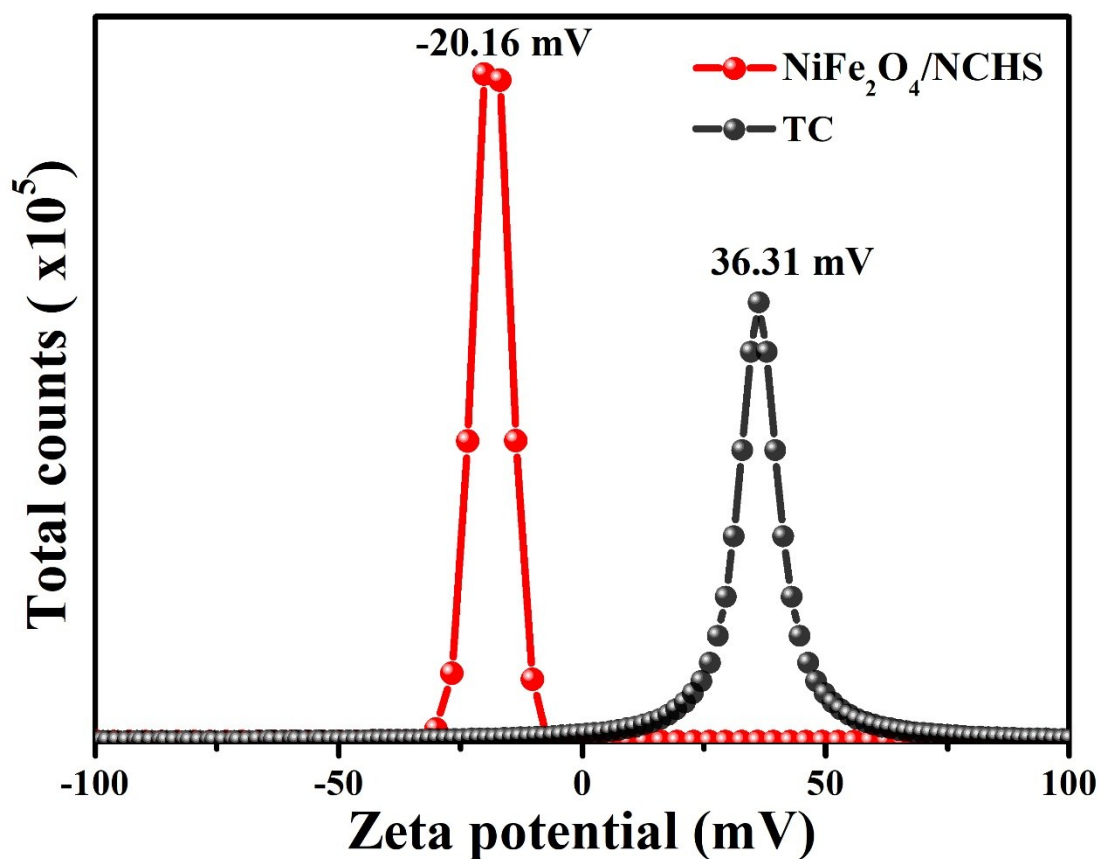
Figure S5. ζ potentials of samples (pH 7)
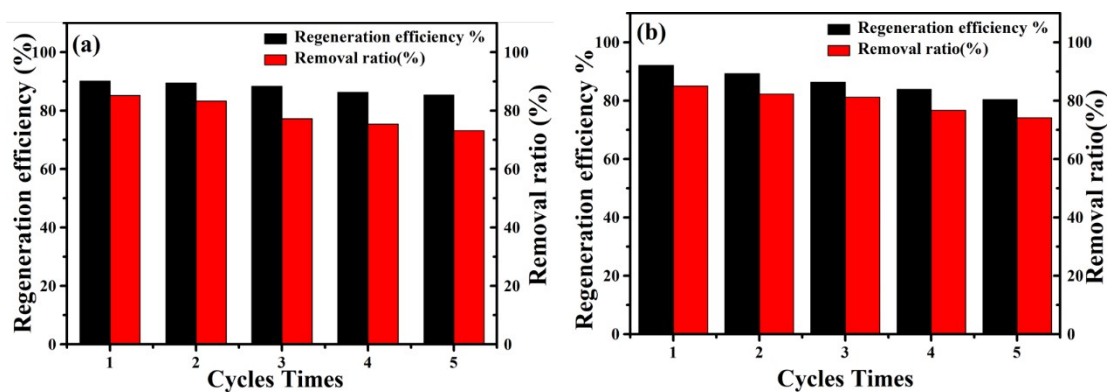

Figure S6. (a) Regeneration efficiency of NaOH (0.001 M) for NiFe2O4/NCHS with tetracycline (b)Regeneration efficiency of NH3·H2O (0.001 M) for NiFe2O4/NCHS with tetracycline (initial tetracycline concentration: 20 mg·L −1, pH: 5, adsorbent dosage: 0.05 g,temperature: 298 K).

■ **REFERENCES**

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