Supporting information

Synthesis and characterization of magnetic mesoporous nanoparticles Fe_3O_4@mSiO_2-DODGA for adsorption of 16 rare earth elements

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Material preparation

Synthesis of Fe_3O_4@mSiO_2@NH_2

Fe_3O_4 magnetic nanoparticles were synthesized by solvothermal reaction according to the literature[1]. Typically, $FeCl_3$ (6 m mol, 1.62 g) and carbamide (30 m mol, 1.80 g) were dissolved in ethylene glycol (58mL) and water (2mL). The obtained mixture was stirred vigorously at room temperature for 30 min and added subsequently into a Teflon-lined stainless autoclave. The autoclave was sealed and placed in a muffle furnace. The increasing temperature programmed was set from room temperature to 200 ^oC in 1 h and remained 200 ^oC for 16 h, followed by cooling to room temperature. The black precipitate was collected by an external magnetic field and purified by washing sequentially with ethanol and ultrapure water for three times, respectively.

The Fe_3O_4@SiO_2 particles were synthesized according to the reported method[2,3]. Briefly, Fe_3O_4 magnetic nanoparticles (0.3 g) was added into a mixed solution of ethanol (160 mL) and ultrapure water (40 mL). The dispersion mixture was sonicated for 30 min to make the nanoparticles disperse evenly. After that, the concentrated ammonia aqueous solution (5 mL, 25 wt%) and TEOS (3.1 m mol, 0.7 mL) was added to make sol-gel reaction occurred on the surface of the particles. The mixture was stirring for 8 h at 40 ^oC. Finally, the Fe_3O_4@SiO_2 particles were separated and washed with ethanol and ultrapure water respectively, dried under vacuum.

The mesoporous SiO2 layer coated on SiO2 shell was prepared with the following method. Typically, Fe_3O_4@SiO_2 particles (0.4 g) was added into a mixed solution of ethanol (30 mL) and ultrapure water (20 mL). The dispersion mixture was sonicated for 10 min. the concentrated ammonia aqueous solution (1 mL, 28 wt%), CTAB(0.9 g) and TEOS (9 mmol, 2.0 mL) was added subsequently. The mixture was stirring for 10 h at room temperature. The Fe_3O_4@mSiO_2 particles were separated and washed with ethanol and ultrapure water. To remove CTAB[4], the obtained Fe_3O_4@mSiO_2 particles were added into a mixed solution of ethanol (150 mL) and $NH_4 \cdot NO_3$ (0.9 g). The mixture was refluxed for 3 h.

Amine functionalization nanoparticles Fe_3O_4@mSiO_2@NH_2 were prepared by reacting with APTES[5]. Briefly, anhydrous toluene (50 mL) and Fe_3O_4@mSiO_2 particles (0.4 g) were added in a three-neck flask. The mixture was sonicated for 30min to make the particles disperse. APTES (4 mL) was added into the flask. The reaction was going on under nitrogen gas protection at 120 ^oC for 24 h. The resultant Fe_3O_4@mSiO_2@NH_2 nanoparticles were washed with anhydrous toluene and acetone for three times, respectively, and dried under vacuum.

Synthesis of DODGA-Cl

The synthesis of DODGA-Cl was referred the literature[6,7]. Diglycolic acid (37.3 mM, 5.0 g) was dissolved in acid anhydride (132.2 mM, 12.5mL) under a nitrogen atmosphere. The mixture was stirred and heated to reflux. After refluxing for 10 min, 3 drops of

phosphoric acid were added into the reaction mixture to catalysis the reaction. The solution was stirred under reflux conditions for 2 hour. The resulting solution was concentrated through rotary evaporation under reduced pressure. The final solution was poured into the toluene to recrystallize the products. After filtrating the solid generated in toluene, a white needle-like product (3.6 g, 83% yield) was obtained to be used directly in the next synthesis step.

In the second step[8], didoctylamine (16.0 m mol, 3.86 g) was placed in a 250 ml round bottom flask and dissolved in DCM (150 ml), glycolic anhydride (22.4 m mol, 2.62 g) was added, and the mixture was stirred at room temperature for 2 h. The end of the reaction was controlled by TLC (DCM/MeOH 8:2). The mixture was concentrated; and the residue was taken up in ethyl acetate, washed successively with 0.1N HCl (×2) and sat. NaCl, and dried over Na_2SO_4. Evaporation of the solvent gave the title compound as an oil which crystallized on standing (5.00 g, 87.5%).

Converting the DODGA-OH to its acyl chloride derivative was referred the normal organic reaction operation. Typically, DODGA-OH (10 m mol, 3.60 g) was dissolved in the solvent of CH_2Cl_2 (30 mL). Anhydrous oxalyl chloride(15 m mol, 1.3mL) was added into the mixture. After stirring for 10 min, 2 drops of DMF was added to catalyze the reaction. The reaction was monitored by thin layer chromatography. At the end of the reaction, the solvent and excessive oxalyl chloride was removed by rotary evaporating. The residue was DODGA-Cl (3.30 g, 87.2%) that could be used directly to modify magnetic particles.

Modifying the Fe_3O_4 particles with the functional unit of DODGA

The functionalized Fe_3O_4 particles were prepared by the reaction between the anime groups on the surface of the Fe_3O_4@$mSiO_2$@NH_2 and the DODGA-Cl. Briefly, the Fe_3O_4@$mSiO_2$@NH_2(0.3 g) and TEA(15 m mol, 2.1 mL) was added into the solvent of anhydrous CH_2Cl_2 (30 mL). The mixture was sonicated for 15min. the solution of DODGA-Cl (10 m mol, 3.75 g) in anhydrous CH_2Cl_2 (10 mL) was dropped slowly into the mixture. The reaction was going on at room temperature for 8 h. The resultant particles were washed with CH_2Cl_2 and methanol three times respectively, and dried under vacuum.

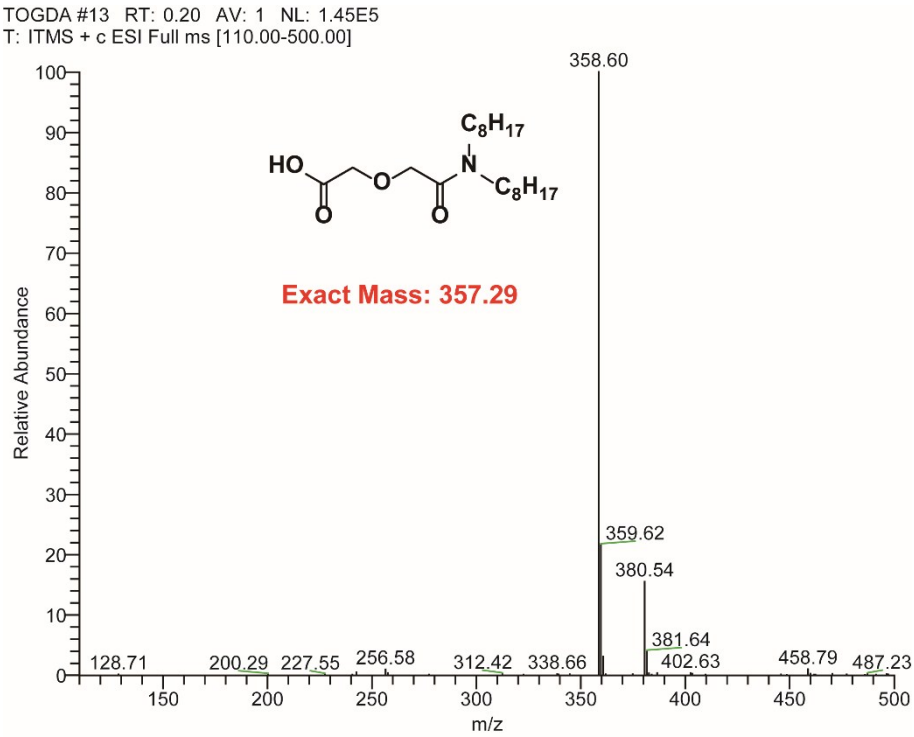

Figure S1. The mass spectra of DODGA

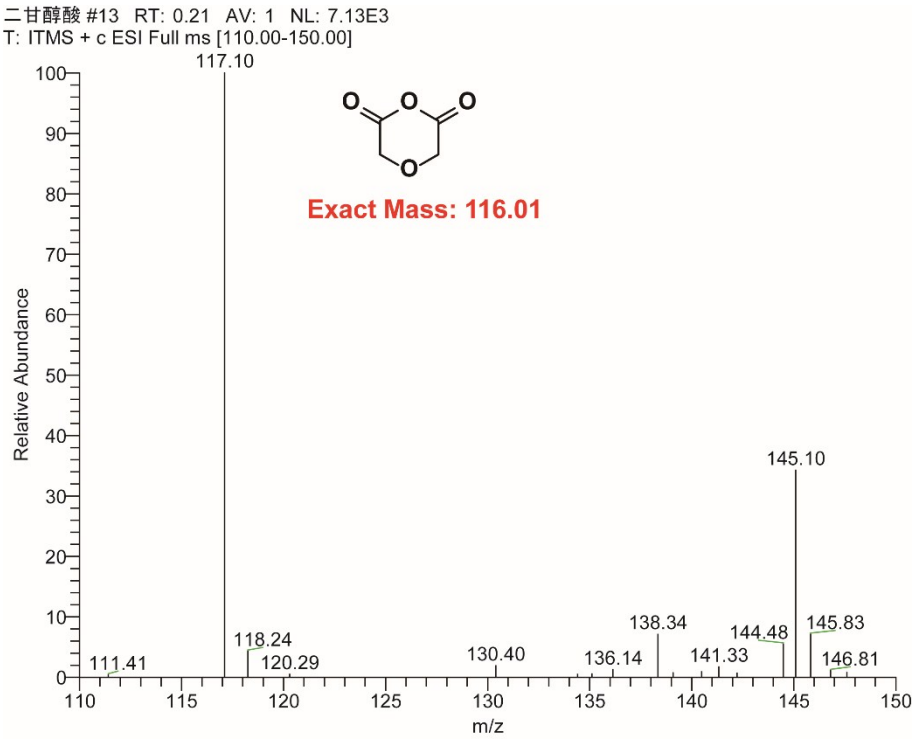

Figure S2. The mass spectra of Glycolic anhydride

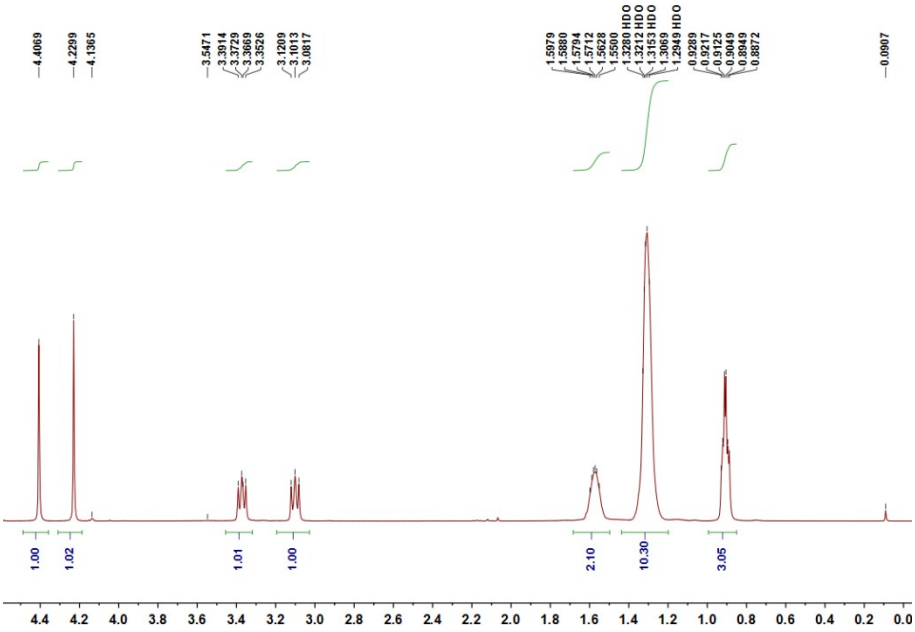

Figure S3. The ^{1}H NMR spectra of DODGA

^{1}H NMR (400 MHz, Chloroform-*d*) δ 4.41 (s, 1H), 4.23 (s, 1H), 3.37 (dd, J = 9.0, 6.6 Hz, 1H), 3.19-3.03 (m, 1H), 1.68-1.50 (m, 2H), 1.31 (s, 10H), 0.91 (td, J = 6.8, 3.0 Hz, 3H).

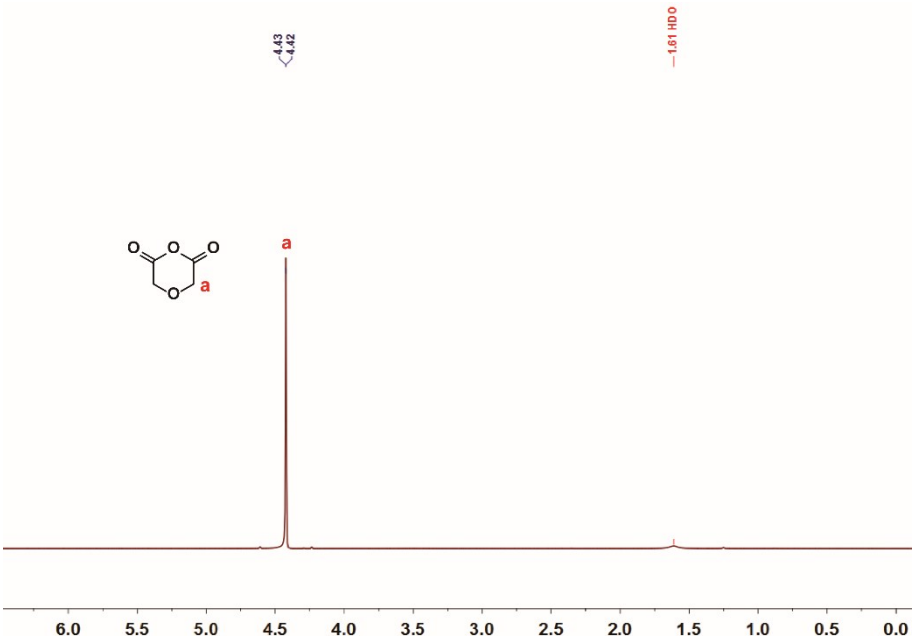

Figure S4. The ^{1}H NMR spectra of Glycolic anhydride

^{1}H NMR (400 MHz, Chloroform-*d*) δ 4.42 (d, *J* = 1.4 Hz, 4H).

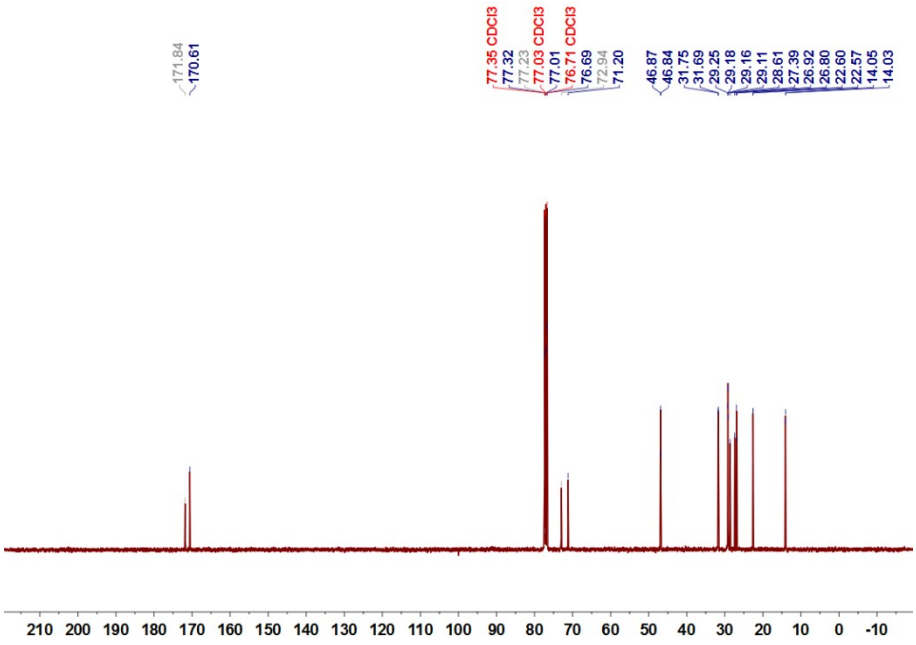

Figure S5. The ^{13}C NMR spectra of DODGA

^{13}C NMR (101 MHz, $CDCl_3$) δ 171.84, 170.61, 72.94, 71.20, 46.87, 46.84, 31.75, 31.69, 29.25, 29.18, 29.16, 29.11, 28.61, 27.39, 26.92, 26.80, 22.60, 22.57, 14.05, 14.03. MS (ESI): calcd. for $C_{20}H_{39}NO_4$: 357.29; found:358.60.

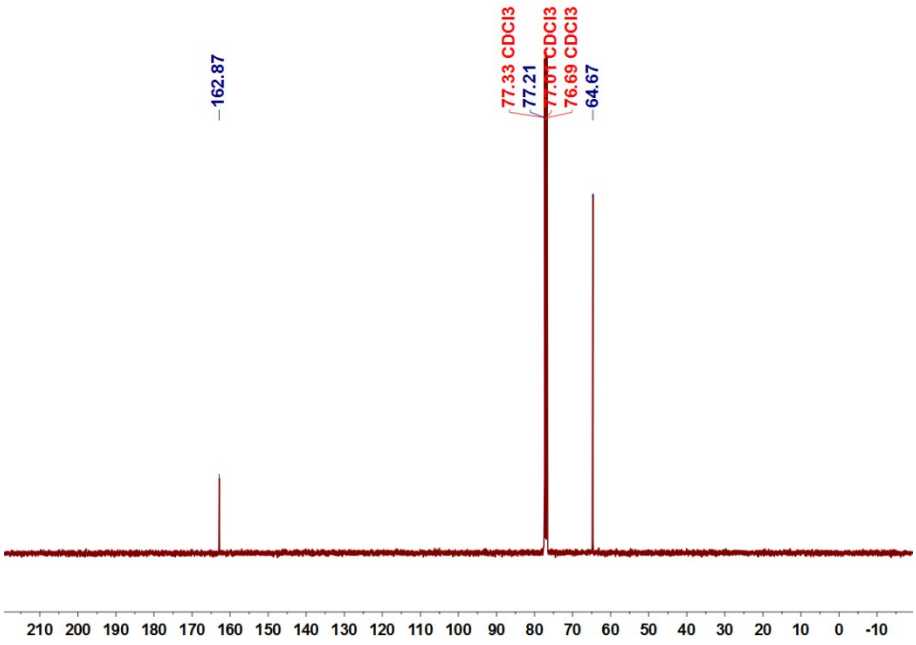

Figure S6. The ^{13}C NMR spectra of Glycolic anhydride

^{13}C NMR (101 MHz, Chloroform-*d*) δ 162.87, 64.67. MS (ESI): calcd. for $C_4H_4O_4$: 116.01; found:117.10.

Adsorption kinetics

Pseudo-first order kinetic model[9]

$$q_t = q_e\left(1 - e^{-k_1 t}\right)$$

Where q_e(mg·g^{-1}) and q_t(mg·g^{-1}) refer to the amounts of REEs adsorbed at equilibrium and designed time, respectively. k_1 (min^{-1}) Pseudo-first order reaction rate constant.

Table S1. The result of data processing according to pseudo-first order kinetic model

Elements	Equation	Parameter		
		R^2	q_e mg·g^{-1}	k_1 min$_{-1}$
Nd	q_t=57.36(1-e$^{-0.089t}$)	0.9000	57.362	0.0891
Sm	q_t=26.65(1-e$^{-0.066t}$)	0.9755	26.6529	0.0665
Eu	q_t=35.40(1-e$^{-0.188t}$)	0.7948	35.3960	0.1882
Ho	q_t=16.43(1-e$^{-0.093t}$)	0.9561	16.428	0.0927
Yb	q_t=33.16(1-e$^{-0.105t}$)	0.9761	33.1592	0.1052
Lu	q_t=39.29(1-e$^{-0.285t}$)	0.5491	39.2897	0.2851
Y	q_t=15.60(1-e$^{-0.103t}$)	0.9592	15.6051	0.1032
Sc	q_t=13.85(1-e$^{-0.088t}$)	0.9106	13.8479	0.0881

Pseudo-second-order model[10]

$$\frac{t}{q_t} = \frac{1}{2k_2 q_e^2} + \frac{1}{q_e} t$$

Where k_2 was pseudo-second-order reaction rate constant (g·mg^{-1}·min^{-1}), the initial adsorption rate h(mg·g^{-1}·min^{-1}) was calculated from the following equation: $h=k_2 q_e^2$

Table S2. The result of data processing according to pseudo-second-order model

Elements	Equation	Parameter		
		R^2	q_e mg·g^{-1}	k_2 g·mg^{-1}·min^{-1}
Nd	t/q_t=0.1484+0.0162t	0.9997	61.7584	282.7313
Sm	t/q_t=0.3136+0.0357t	0.9998	28.0112	123.0296
Eu	t/q_t=0.0828+0.027t	1.0000	37.0370	56.7901
Ho	t/q_t=0.4107+0.0578t	0.9997	17.3010	61.4666
Yb	t/q_t=0.1515+0.0289t	0.9999	34.6021	90.6958
Lu	t/q_t=0.095+0.0237t	0.9998	42.1941	84.5662
Y	t/q_t=0.3802+0.0608t	0.9999	16.4474	51.4251
Sc	t/q_t=0.5058+0.0687t	0.9997	14.5560	53.5840

Intra-particle diffusion model[11]

$$q_t = k_i t^{1/2} + C$$

Where k_i was intra-particle diffusion rate constant ($mg \cdot g^{-1} \cdot min^{-1/2}$), C represented the boundary layer diffusion effect. The larger the value of C, the greater the diffusion effect of the boundary layer.

Table S3. The result of data processing according to intra-particle diffusion model

Elements	Equation	Parameter		
		R^2	k_i $mg \cdot g^{-1} \cdot min^{-1/2}$	C
Nd	q_t=35.996 + 1.4109 $t^{1/2}$	0.6592	1.4109	35.996
Sm	q_t=14.789 + 0.7398 $t^{1/2}$	0.5851	0.7398	14.789
Eu	q_t=28.686 + 0.4675 $t^{1/2}$	0.5898	0.4675	28.686
Ho	q_t=10.433 + 0.3864 $t^{1/2}$	0.4838	0.3864	10.433
Yb	q_t=22.532 + 0.6897 $t^{1/2}$	0.5001	0.6897	22.532
Lu	q_t=33.574 + 0.4466 $t^{1/2}$	0.8311	0.4466	33.574
Y	q_t=10.419 + 0.3384 $t^{1/2}$	0.4985	0.3384	10.419
Sc	q_t=8.7572 + 0.3277 $t^{1/2}$	0.4386	0.3277	8.7572

Elovich model[12]

$$q_t = \frac{\ln(\alpha\beta)}{\beta} + \frac{\ln t}{\beta}$$

Where α ($mg \cdot g^{-1} \cdot min^{-1}$) was the initial adsorption rate constant, β($mg \cdot g^{-1} \cdot min^{-1}$) was a parameter related to the surface coverage of the adsorbent and the activation energy of chemical adsorption.

Table S4. The result of data processing according to Elovich model

Elements	Equation	Parameter		
		R^2	α $mg \cdot g^{-1} \cdot min^{-1}$	β $mg \cdot g^{-1} \cdot min^{-1}$
Nd	q_t=26.945lnt+14.218	0.9612	45.6711	0.03711
Sm	q_t=14.724lnt+2.5879	0.9266	17.5533	0.06792
Eu	q_t=9.2842lnt+21.003	0.9298	89.1700	0.10771
Ho	q_t=8.2165lnt+3.3692	0.8744	12.38142	0.12171
Yb	q_t =14.55lnt+10.074	0.8899	29.0774	0.06873
Lu	q_t =7.696lnt+27.774	0.9867	284.1745	0.12994
Y	q_t=7.1041lnt+4.3537	0.8782	13.1118	0.14076
Sc	q_t=7.0795lnt+4.3537	0.8181	13.0943	0.14125

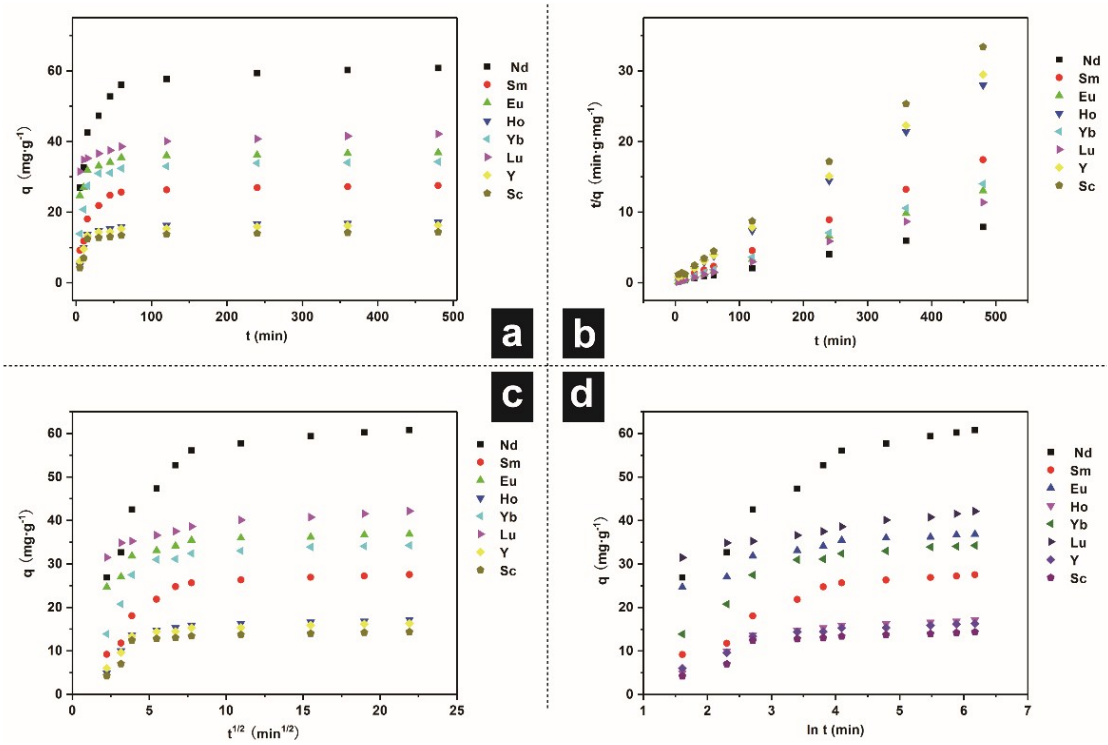

Figure S7. Different adsorption kinetic model of Fe_3O_4@ $mSiO_2$-TODGA for REEs (a. pseudo-first-order, b. pseudo-second-order, c. intra-particle diffusion, d. Elovich model)

Adsorption isotherms

Langmuir model[13]

$$\frac{C_e}{q_e} = \frac{C_e}{Q_{max}} + \frac{1}{K_L Q_{max}}$$

Where Qe($mg \cdot g^{-1}$) and Ce($mg \cdot L^{-1}$) was the amount of nanoparticles adsorbed and the concentration of REEs in the solution under the equilibrium, respectively; Q_{max}($mg \cdot g^{-1}$) was the saturated monolayer adsorption, and K_L($L \cdot mg^{-1}$) was the Langmuir constant, which is related to the binding energy of the adsorption system.

Table S5. The result of data processing according to Langmuir model

Elements	Equation	Parameter			
		R^2	Q_{max} ($mg \cdot g^{-1}$)	K_L ($L \cdot mg^{-1}$)	R_L
Nd	C_e/q_e=0.0145 C_e +0.0109	0.9327	68.9655	1.3303	0.4291
Sm	C_e/q_e =0.0350 C_e +0.0102	0.9987	28.5714	3.4314	0.2256
Eu	C_e/q_e =0.0249 C_e +0.0211	0.9981	40.1606	1.1801	0.4586
Ho	C_e/q_e =0.0579 C_e +0.0439	0.9892	17.2712	1.3189	0.4312
Yb	C_e/q_e =0.0258 C_e +0.0236	0.9911	38.7597	1.0932	0.4777
Lu	C_e/q_e =0.022 C_e +0.0222	0.9900	45.4545	0.9910	0.5022
Y	C_e/q_e =0.0568 C_e +0.0318	0.9919	17.6056	1.7862	0.3589
Sc	C_e/q_e =0.0674 C_e +0.0105	0.9983	14.8368	6.4190	0.1347

Freundlich model[14]

$$\ln\left(Q_e\right) = \ln\left(K_f\right) + \frac{1}{n}\ln\left(C_e\right)$$

where K_f and n was the Freundlich isotherm constant, respectively. K_f was related to the adsortpion amount, and n was related to the strength of the adsorption. The value of 1/n between 0.1-0.5 indicated that the adsorption process was favorable.

Table S6. The result of data processing according to Freundlich model

Elements	Equation	Parameter		
		R^2	K_f	n
Nd	$\ln(q_e)=0.4574\ln(C_e)+3.4593$	0.9972	31.7947	2.1863
Sm	$\ln(q_e)=0.2941\ln(C_e)+2.7919$	0.9366	16.3120	3.4002
Eu	$\ln(q_e)=0.1542\ln(C_e)+3.2662$	0.9838	26.2115	6.4851
Ho	$\ln(q_e)=0.1808\ln(C_e)+2.3297$	0.9950	10.2794	5.5310
Yb	$\ln(q_e)=0.7722\ln(C_e)+2.9209$	0.9828	18.5580	1.2950
Lu	$\ln(q_e)=0.4854\ln(C_e)+2.8683$	0.9444	17.6071	2.0602
Y	$\ln(q_e)=0.6547\ln(C_e)+2.1791$	0.9025	8.8383	1.5274
Sc	$\ln(q_e)=0.3962\ln(C_e)+2.2898$	0.9429	9.8730	2.5240

D-R model[15]

$$\ln Q_e = \ln Q_m - \beta\varepsilon^2$$

$$\varepsilon = RT\ln\left(1 + {}^1/_{C_e}\right)$$

$$E = (2\beta)^{-0.5}$$

Where β was a constant ($mol^2 \cdot J^{-2}$) and related to adsorption energy; Q_m was the thoeretical saturated adsorption capacity ($mg \cdot g^{-1}$); ε was Polanyi potential; Rwas gas constant ($8.314 J \cdot mol^{-1} \cdot K^{-1}$), T was the absolute tempertautre (K); E was the average adsorption energy (kJ·mol-1) and represented the energy required to transfer 1 mol of metal ions from the solution to the surface of the solid phase. The value of E could reflect whether the adsorption mechanism was a physical reaction or a chemical reaction. When E>8 $kJ \cdot mol^{-1}$, the adsorption process follows chemical adsorption, while E<8 $kJ \cdot mol^{-1}$, the adsorption process was a physical reaction.

Table S7. The result of data processing according to D-R model

Elements	Equation	Parameter			
		R^2	β ($mol^2 J^{-2}$)	E ($kJ \cdot mol^{-1}$)	Q_{max} ($mg \cdot g^{-1}$)
Nd	$\ln(Q_e)=3.6081-8.001\times10^{-9}\varepsilon^2$	0.9543	8.001×10^{-9}	15.81	36.8959
Sm	$\ln(Q_e)=3.2104-1.137\times10^{-8}\varepsilon^2$	0.9574	1.137×10^{-8}	13.26	24.7890
Eu	$\ln(Q_e)=3.4517-2.801\times10^{-8}\varepsilon^2$	0.953	2.801×10^{-8}	8.45	31.5540
Ho	$\ln(Q_e)=2.6778-1.200\times10^{-}$	0.9603	1.200×10^{-8}	12.91	14.5530

	$^{8}\varepsilon^{2}$				
Yb	ln（Q_e）=3.4329-3.197×10-	0.9578	3.197×10^{-8}	7.91	30.9663
	$^{8}\varepsilon^{2}$				
Lu	ln（Q_e）=3.4410-2.000×10-	0.9569	2.000×10^{-8}	10.00	31.2182
	$^{8}\varepsilon^{2}$				
Y	ln（Q_e）=2.8273-4.001×10-	0.9555	4.001×10^{-8}	7.07	16.8998
	$^{8}\varepsilon^{2}$				
Sc	ln（Q_e）=2.6359-1.600×10-	0.9666	1.600×10^{-8}	11.18	13.9559
	$^{8}\varepsilon^{2}$				

Tempkin model [16]

$$Q_e = B_T \ln K_T + B_T \ln C_e$$

Where the value of the B_T was calculated from the following equation: $B_T=(RT)/b_T$, R was the gas constant (8.314J·mol^{-1}·K^{-1}), b_T was the Tempkin constant (J·mol^{-1}) and related to adsorption heat. K_T was the equilibrium binding constant (L·g^{-1}) and related to the maximum binding energy.

Table S8. The result of data processing according to Tempkin model

Elements	Equation	Parameter		
		R^2	B_T	K_T
Nd	Q_e=9.0897lnC_e+37.799	0.8545	9.0897	63.9718
Sm	Q_e=3.8971lnC_e+19.61	0.9702	3.8971	153.2310
Eu	Q_e=7.7306lnC_e+21.578	0.9665	7.7396	16.2485
Ho	Q_e=2.2484lnC_e+10.392	0.9805	2.2484	101.6925
Yb	Q_e=8.0659lnC_e+20.264	0.9395	8.0659	12.3333
Lu	Q_e=7.9296lnC_e+23.234	0.9650	7.9196	18.7977
Y	Q_e=3.4884lnC_e+9.9655	0.8066	3.4884	17.4049
Sc	Q_e=2.3076lnC_e+10.189	0.8240	2.3076	82.7157

Table S9. 16 kinds of REEs linear standard curve

Elements		R^2	Liner range
La	Y=242.396X+190.248	0.99969	5~1000 μg/L
Ce	Y=41.843X+23.1529	0.99986	5~1000 μg/L
Pr	Y=33.372X+32.615	0.99997	5~1000 μg/L
Nd	Y=29.463X+214.024	0.99781	5~1000 μg/L
Sm	Y=53.289X-17.493	0.99986	5~1000 μg/L
Eu	Y=120.374X+23.768	0.99999	5~1000 μg/L
Gd	Y=73.411X+38.238	0.99996	5~1000 μg/L
Tb	Y=49.724X+14.738	0.99995	5~1000 μg/L
Dy	Y=83.063X+0.00	0.99784	10~1000 μg/L
Ho	Y=35.289X+17.842	0.99996	5~1000 μg/L
Er	Y=54.089X+2.625	0.99999	5~1000 μg/L
Tm	Y=139.274X+34.453	1.00000	5~1000 μg/L
Yb	Y=611.496X+120.269	1.00000	5~1000 μg/L

Lu	Y=213.229X+1.236	0.99999	5~1000 μg/L
Y	Y=152.899X+74.869	0.99998	5~1000 μg/L
Sc	Y=487.212X+5.708	0.99999	5~1000 μg/L

Table S10. Comparison of the Fe_3O_4@mSiO_2-DODGA with the other conventional materials

Entry	Materials	Recycling times The final adsorption ratio	REEs	Ref.
1	Fe@CS-DGA	5/>90%	Pb(II)	28
2	Fe_3O_4@TODGA	3/80%, 92%	Am(III), Pu(IV)	29
3	Fe_3O_4@ HA-MNPs	6/95%	Eu(III)	23
4	SBA-15-BSEA- Fe_3O_4 -NPs	5/95%	Ce(III)	18

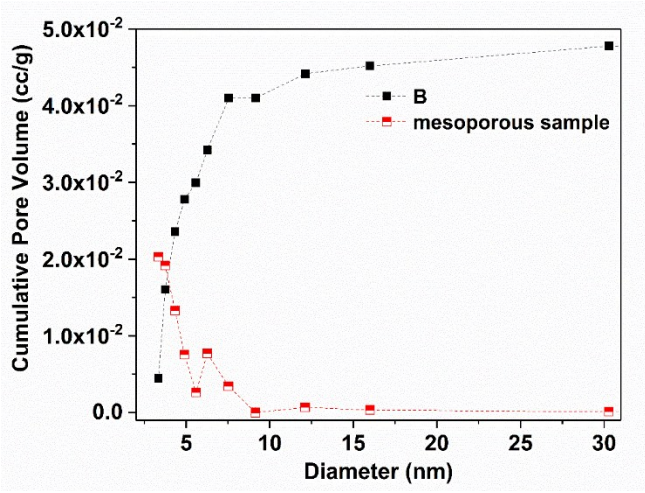

Figure S8. The pore diameter of Fe_3O_4@mSiO_2-DODGA

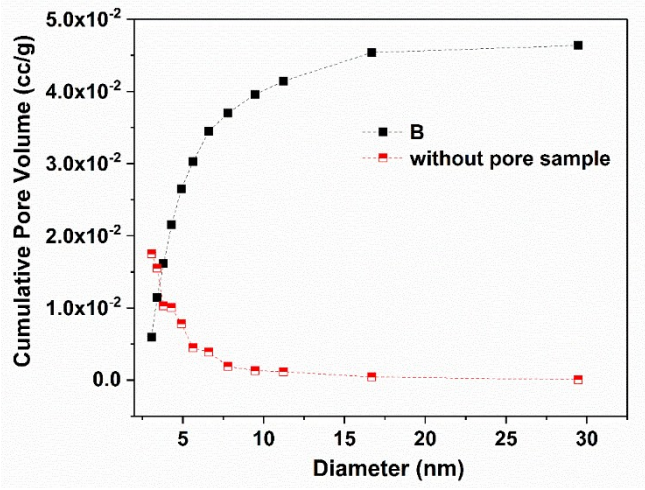

Figure S9. The pore structure of Fe_3O_4@SiO_2

Reference

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