Supporting Information

Synthesis of magnetic short-channel mesoporous silica SBA-15 modified with polypyrrole/polyaniline copolymer for the removal of mercury ions from aqueous solution

Jingtao Shen[a,†], Shuyuan Zhang[a,†], Zheng Zeng[a], Jialun Huang[a], Yi Shen[a], Yongfu Guo[a,b,*]

[a] *School of Environmental Science and Engineering, Suzhou University of Science and Technology, Suzhou, 215009, China*

[b] *Jiangsu Collaborative Innovation Center of Technology and Material of Water Treatment, Suzhou University of Science and Technology, Suzhou, 215009, China*

[*]Corresponding author: Tel: +86 512 68092987 (Y. Guo).

E-mail address: yongfuguo@163.com (Y. Guo)

[†] These authors contributed equally to this work.

Figure S1

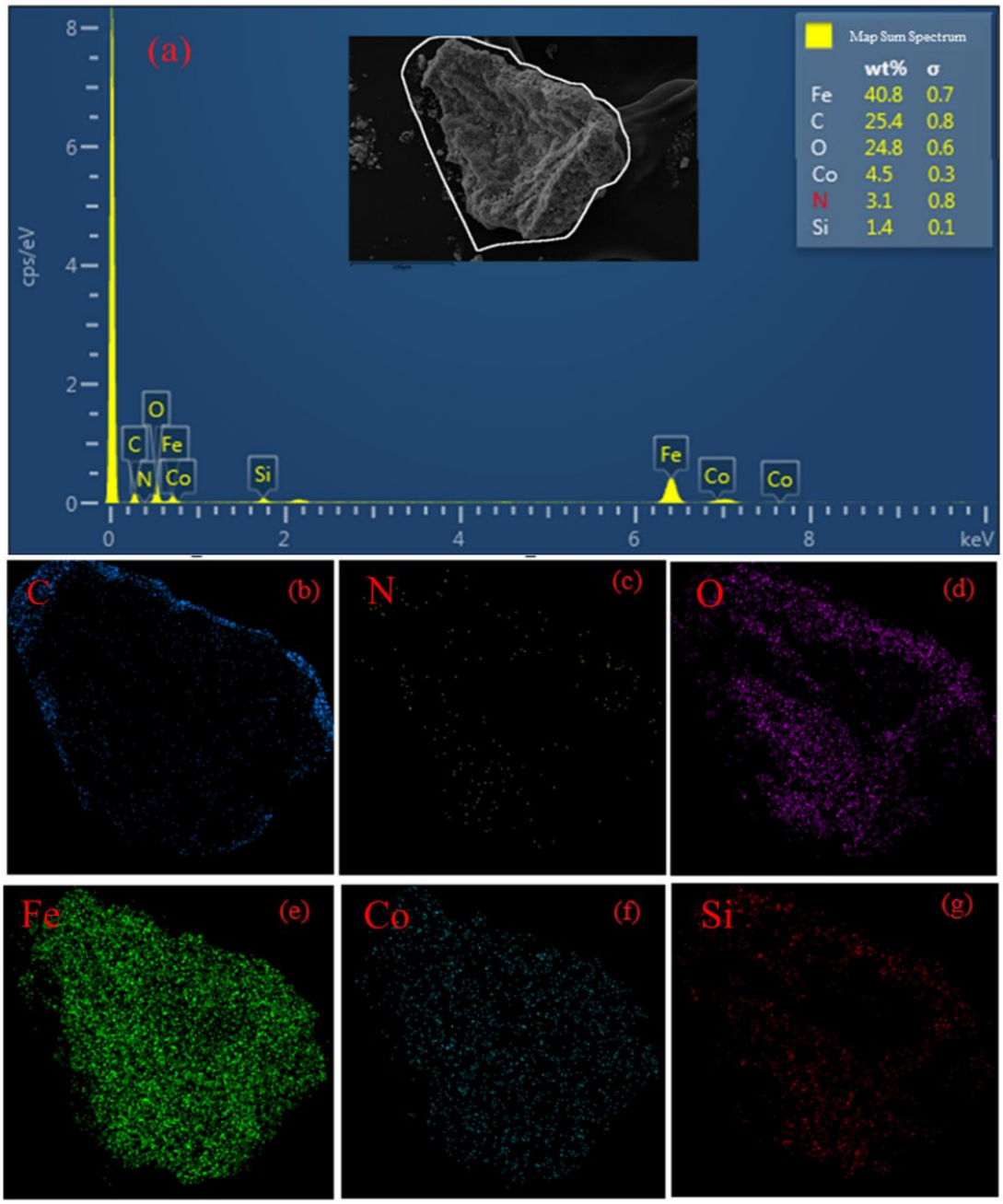

Figure S1. EDS spectrum (inserted: Map Sum Spectrum) of PPy-PANI/M-SBA-15 (a), and EDX elemental mapping of PPy-PANI/M-SBA-15 (b-g).

Table S1.

Kinetic fitting for adsorption of Hg(Ⅱ) onto PPy-PANI/M-SBA-15.

Pseudo-first-order						
$q_{e,exp}$	$q_{e,fit}$	k_1	R^2	x^2	SSE	RMSE
253.8	231.6	0.1159	0.8580	0.8491	11608	26.9
Pseudo-second-order						
$q_{e,exp}$	$q_{e,fit}$	k_1	R^2	x^2	SSE	RMSE
253.8	241.5	0.0008	0.9309	0.9266	5650.3	18.8

Intra-particle diffusion								
K_{d-1}	C_1	R_1^2	K_{d-2}	C_2	R_2^2	K_{d-3}	C_3	R_3^2
33.68	37.36	0.906	4.95	167.29	0.967	0.20	255.74	1

Elovich					
α	β	R^2	x^2	SSE	RMSE
276.6	0.03236	0.9924	0.9913	625.1	6.445

Table S2.

Adsorption isotherm parameters of PPy-PANI/ M-SBA-15.

Langmuir							
T (K)	Q_m (mg/g)	R_L	K_L (L/mg)	R^2	x^2	SSE	RMSE
298	363.9	0.256	0.0967	0.9697	0.9568	672.4	14.97
308	355.8	0.153	0.1839	0.9838	0.9784	338.7	10.63
318	366.1	0.025	1.3095	0.9898	0.9864	239.1	8.93
Freundlich							
T (K)	K_F ($L^n/mg^{n-1}/g$)	$1/n_F$		R^2	x^2	SSE	RMSE
298	67.5	0.4082		0.9173	0.8897	1171.6	23.92
308	101.4	0.3256		0.9794	0.9725	430.9	11.99
318	214.6	0.1646		0.9334	0.9113	1560	22.8
Temkin							
T (K)	b_T	k_T		R^2	x^2	SSE	RMSE
298	84.6	0.820		0.9574	0.9431	885.1	17.18
308	73.45	2.096		0.9922	0.9896	162.4	7.36
318	48.35	75.163		0.9695	0.9593	715.6	15.44
Dubinin-Radushkevich							
T (K)	q_{max} (mg/g)	E (KJ/mol)		R^2	x^2	SSE	RMSE
298	38.78	121.09		0.9193	0.8925	52.9	3.252
308	35.75	134.07		0.9280	0.9040	46.7	3.055
318	37.55	166.71		0.9603	0.9471	33.6	2.592

Table S3.

Comparison of adsorption capacity for mercury (II) ions.

Adsorbents	BET (m^2/g)	pH	Fitting model	Q_m (mg/g)	Ref.
$CoFe_2O_4$@SiO_2-NH_2	17.08	7	Langmuir	149.3	1
$CoFe_2O_4$–rGO	69.9	4.6	Langmuir	157.9	2
SBA-15-SH	50.9	8	Freundlich	195.6	3
Coal-based activated carbon	900	4	Langmuir	48.0	4
Polypyrrole/SBA-15	97.6	8	Langmuir	200	5
PDA@Fe_3O_4	/	5.36	Langmuir	307	6
Polypyrrole multilayer cellulose	/	6	Langmuir	31.68	7
Magnetic GO	58.6	6	Langmuir	71.3	8
Magnetic CNTs/Fe_3O_4	97.16	6.5	Langmuir	65.52	9
PPy-PANI/M-SBA-15	12.3	7	Langmuir	253.8	This work

Regeneration performance

In the experimental part of studying the adsorption effect of pH on the material, it can be seen that the change of pH has a significant impact on the adsorption of PPy-PANI/M-SBA-15. Therefore, an acidic solution can be selected to clean the material to achieve material regeneration.

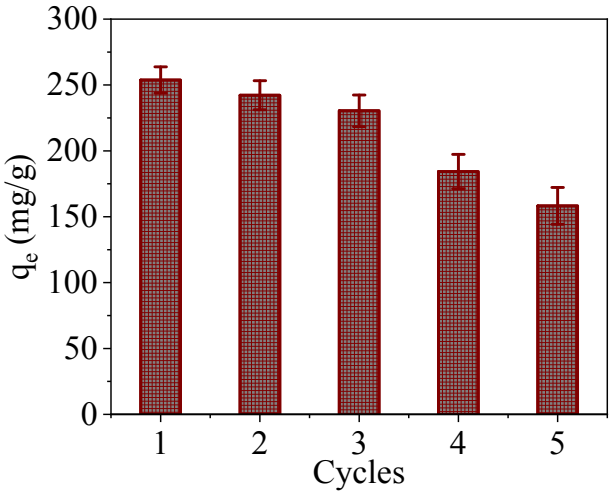

Figure S2. Adsorption performance after repeated use of PPy-PANI/M-SBA-15. (pH = 7, dosage of PPy-PANI/M-SBA-15 = 0.05 g/L, T = 298 K， C_0 = 30 mg/L, t = 6 h)

In this experiment, 0.1 M dilute HCl solution was used as the regeneration solution. The PPy-PANI/M-SBA-15 after adsorption equilibrium was placed in the regeneration solution and oscillated at room temperature for 2 h. After that, a magnet was used to separate the adsorbent from the regeneration solution, then the material was washed with pure water and dried. To show the reusability of the adsorbent, the same method was used for five cycles. The results in Figure S2 show that the adsorption and regeneration performance of PPy-PANI/M-SBA-15 remain good in the first three processes, only a decrease of 9.2%. However, there was a significant decrease after the fourth regeneration, which may be caused by the blockage of some pores of the material or the loss of some active adsorption sites on the surface of the material during multiple adsorption/desorption processes. In short, PPy-PANI/M-SBA-15 has good regeneration performance.

Table S4.

Thermodynamic parameters of adsorption of mercury by PPy-PANI/M-SBA-15.

C_0	ΔH^0	ΔS^0	ΔG^0		
			298 K	308 K	318 K
20	45.17	218.16	-20.01	-21.61	-24.40
30	20.64	134.81	-19.64	-20.64	-22.36
40	16.37	117.53	-18.72	-19.68	-21.08

REFERENCES

(1) Wang, X.; Zhang, Z.; Zhao, Y.; Xia, K.; Guo, Y.; Qu, Z.; Bai, R., A mild and facile synthesis of amino functionalized $CoFe_2O_4$@SiO_2 for Hg(II) removal. *Nanomaterials* **2018,** *8* (9), 673.

(2) Zhang, Y.; Yan, L.; Xu, W.; Guo, X.; Cui, L.; Gao, L.; Wei, Q.; Du, B., Adsorption of Pb(II) and Hg(II) from aqueous solution using magnetic $CoFe_2O_4$-reduced graphene oxide. *J. Mol. Liq.* **2014,** *191*, 177-182.

(3) Shen, Y.; Jiang, N.; Liu, S.; Zheng, C.; Wang, X.; Huang, T.; Guo, Y.; Bai, R., Thiol functionalization of short channel SBA-15 through a safe, mild and facile method and application for the removal of mercury (II). *J. Environ. Chem. Eng* **2018,** *6* (4), 5420-5433.

(4) Guo, Y.; Wang, Z.; Zhou, X.; Bai, R., Removal of mercury (II) from aqueous solution with three commercial raw activated carbons. *Res. Chem. Intermed.* **2016,** *43* (4), 2273-2297.

(5) Shafiabadi, M.; Dashti, A.; Tayebi, H.-A., Removal of Hg (II) from aqueous solution using polypyrrole/SBA-15 nanocomposite: Experimental and modeling. *Synth. Met.* **2016,** *212*, 154-160.

(6) Davodi, B.; Ghorbani, M.; Jahangiri, M., Adsorption of mercury from aqueous solution on synthetic polydopamine nanocomposite based on magnetic nanoparticles using box–behnken design. *J. Taiwan Inst. Chem. Eng.* **2017,** *80*, 673.

(7) Hanif, Z.; Lee, S.; Hussain Qasim, G.; Ardiningsih, I.; Kim, J.-A.; Seon, J.; Han, S.; Hong, S.; Yoon, M.-H., Polypyrrole multilayer-laminated cellulose for large-scale repeatable mercury ion removal. *J. Mater. Chem. A.* **2016,** *4* (32), 12425-12433.

(8) Guo, Y.; Deng, J.; Zhu, J.; Zhou, X.; Bai, R., Removal of mercury(ii) and methylene blue from a wastewater environment with magnetic graphene oxide: adsorption kinetics, isotherms and mechanism. *RSC Adv.* **2016,** *6* (86), 82523-82536.

(9) Zhang, C.; Sui, J.; Li, J.; Tang, Y.; Cai, W., Efficient removal of heavy metal ions by thiol-functionalized superparamagnetic carbon nanotubes. *Chem. Eng. J.* **2012,** *210*, 45-52.