Electronic Supplementary Information for

## Highly Stable Mesoporous Silica Nanospheres Embedded with FeCo/Graphitic Shell Nanocrystals as Magnetically Recyclable Multifunctional Adsorbents for Wastewater Treatment

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Fig. S1. TEM images of FeCo/GC NCs obtained from (a) 65 nm, (b) 130 nm, and (c) 270 nm FeCo/GC NCs@MSNs after HF treatment.



Fig. S2. XRD pattern of MSNs.



Fig. S3. EDX data of (a) 65 nm, (b) 130 nm, and (c) 270 nm FeCo/GC NCs@MSNs.



Fig. S4. (a) Temperature-dependent magnetization curves under an applied field of 100 Oe and (b) Field-dependent magnetization curves at 300 K for 130 nm FeCo/GC NCs@MSNs. Inset in (b) shows the loop on an enlarged x-axis scale.



Fig. S5. Photographs of 130 nm (a, b) FeCo/GC NCs@MSNs-SH and (c) FeCo NCs@MSNs-SH in 35% HCl solutions (a, c) and a 1 mM NaOH (pH 11) solution (b). TEM images of FeCo/GC NCs@MSNs-SH stored over a monitoring period of a week in the (d) HCl and (e) NaOH solutions, respectively. FeCo/GC NCs@MSNs-SH exhibited stability against HCl or NaOH etching over a

monitoring period of a week. However, FeCo NCs@MSNs-SH having FeCo (being unencapsulated with a carbon shell) NCs turned the color to green in the HCl solution right after the addition due to the Fe and Co etching.



Fig. S6. FT-IR data of 130 nm MSNs.



Fig. S7. TEM images of 130 nm (a) MSNs-SH and (b) FeCo/GC@MSNs-SH.



Fig. S8. Effect of pH on the adsorption of (a) MB, (b) MO, and (c)  $Hg^{2+}$  onto the FeCo/GC NCs@MSNs-SH.



Fig. S9. TEM images of 130 nm FeCo/GC NCs@MSNs-SH after the six consecutive adsorption cycles for MO.

Sample	BET surface area (m <sup>2</sup> /g)	Pore volume (cm <sup>3</sup> /g)	pore size (nm)
MSNs	661	0.73	2.45
FeCo/GC NCs@MSNs	442	0.65	2.19

Table S1. Physicochemical properties of selected samples.

Table S2. Langmuir isotherm parameters for MB adsorption on various adsorbents.

Comple	Langmuir model			
Sample	$q_{max}$	b	$R^2$	
MSNs	20.33	1.54	0.99	
FeCo/GC NCs@MSNs	28.99	2.97	0.99	
FeCo/GC NCs@MSNs-SH	37.17	3.49	0.99	

Table S3. Langmuir isotherm parameters for MO adsorption on various adsorbents.

Somalo -	Langmuir model			
Sample –	<b>q</b> max	b	$R^2$	
MSNs	3.33	0.38	0.97	
FeCo/GC NCs@MSNs	13.39	0.48	0.98	
FeCo/GC NCs@MSNs-SH	15.75	0.57	0.99	

Magnetic adsorbents for MB	<b>q</b> e	pH	References
RGO–MnFe <sub>2</sub> O <sub>4</sub> hybrid	34.7	_	S. Bai et al. (2012) <sup>1</sup>
Fe3O4@C	52.5	7.0	S. P. Wu et al. $(2016)^2$
MMWCNT	11.9	7.0	J. L. Gong et al. (2009) <sup>3</sup>
M-MWCNTs	45.8	7.0	L. Ai et al. (2011) <sup>4</sup>
MGO	275.9	9.0	Y. F. Guo et al. $(2016)^5$
CS/Mt-OREC	9.7	7.0	L. Zeng et al. (2015) <sup>6</sup>
x-Fe <sub>2</sub> O <sub>3</sub> /C composites	193.4	-	J. Xiao et al. (2013) <sup>7</sup>
FeCo/GC NCs@MSNs-SH	36.8	7.0	This Study
Magnetic adsorbents for MO	<b>q</b> e	pH	References
m-CS/c-Fe <sub>2</sub> O <sub>3</sub> /MWCNTs	61.4	-	H. Y. Zhu et al. (2010) <sup>8</sup>
CS/Mt-OREC	5.0	7.0	L. Zeng et al. (2015) <sup>6</sup>
CANF	102.0	4.0	B. Tanhaei et al. (2015) <sup>9</sup>
r-Fe <sub>2</sub> O <sub>3</sub> /chitosan	28.5	2.9	R. Jiang et al. (2012) <sup>10</sup>
AC/NiFe <sub>2</sub> O <sub>4</sub>	93.5	3.0	T. Jiang et al. (2015) <sup>11</sup>
FeCo/GC NCs@MSNs-SH	14.6	7.0	This study
Magnetic adsorbents for Hg(II)	<b>q</b> e	pH	References
MAF-SCMNPs	240.0	6.0	S. Bao et al. (2017) <sup>12</sup>
$Fe_3O_4@Cu_3(btc)_2$	158.2	6.0	F. Ke et al. (2017) <sup>13</sup>
Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -SH	148.8	6.5	S. Zhang et al. (2013) <sup>14</sup>
PR-MNPs	133	4.0	J. Song et al. (2011) <sup>15</sup>
CG-MCS	220.1	7.0	Y. Wang et al. (2013) <sup>16</sup>
AEPE-PS-MPs	28.7	7.0	K. Jainae et al. (2015) <sup>17</sup>
TETA-PGMA	468	6.0	Y. Wang et al. (2016) <sup>18</sup>
Thiol-functionalized MGO	30.9	_	J. Bao et al. (2013) <sup>19</sup>
HMSMCs	62.8	6.5	X. Zhang et al. (2015) <sup>20</sup>
MGO	59.9	6.0	Y. F. Guo et al. $(2016)^5$
$rGO-Fe(0)-Fe_3O_4$	22.0	7.0	P. Bhunia et al. (2012) <sup>21</sup>
FeCo/GC NCs@MSNs-SH	221.4	4.0	This study

Table S4. Comparison of adsorption capacities of FeCo/GC NCs@MSNs-SH with different adsorbents.

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