

Supplementary Information

Graphene oxide-iron oxide and reduced graphene oxide-iron oxide hybrid materials for removal of organic and inorganic pollutants

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Characterization of the GO-iron oxide and RGO-iron oxide composites

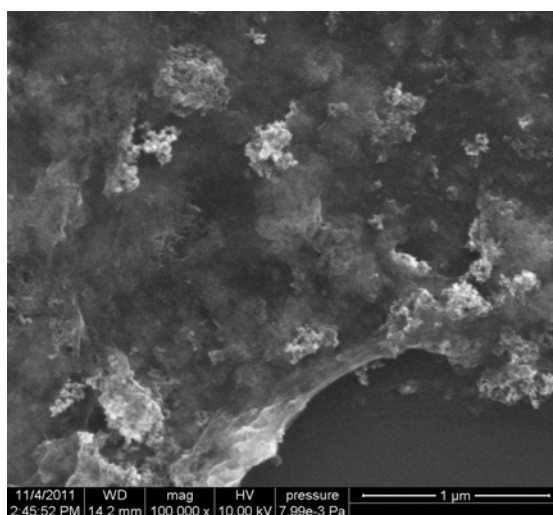


Fig. S1 SEM image of GO-iron oxides.

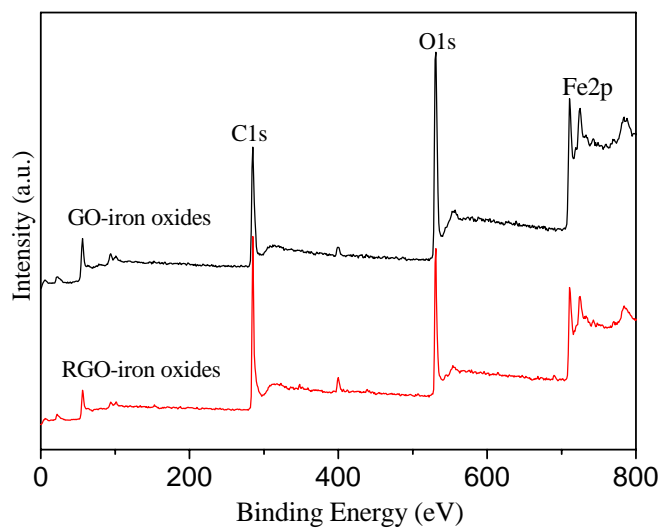


Fig. S2 Wide scan XPS spectra of GO-iron oxides and RGO-iron oxides.

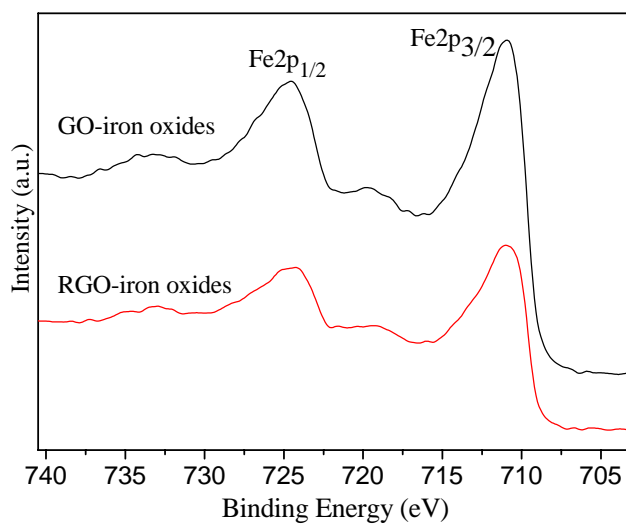


Fig. S3 High resolution XPS spectra of Fe2p.

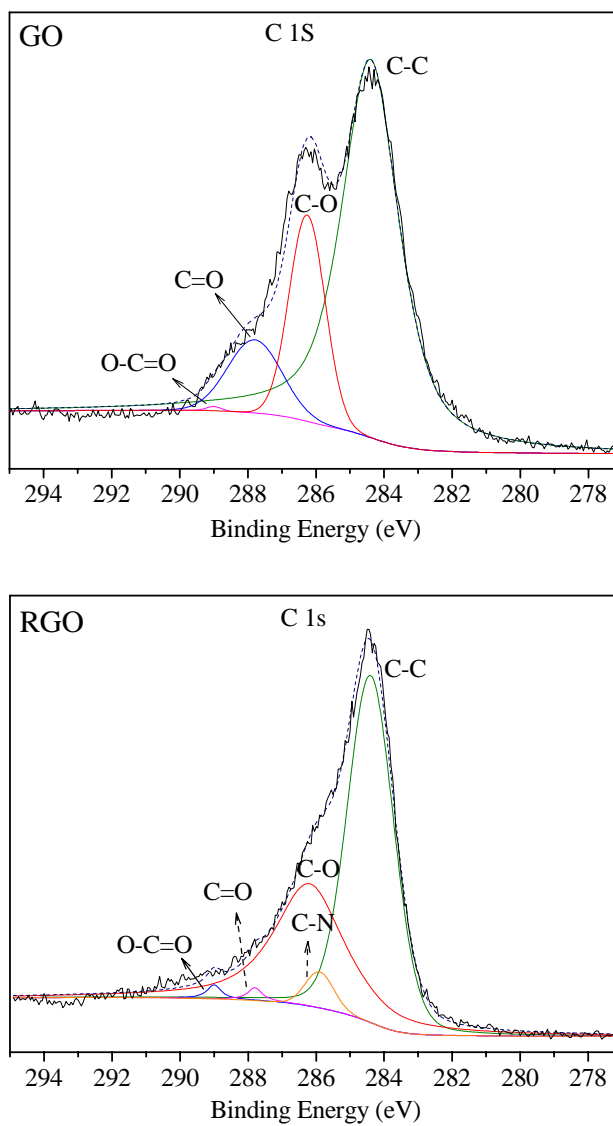


Fig. S4 C1s spectra of raw GO and RGO.

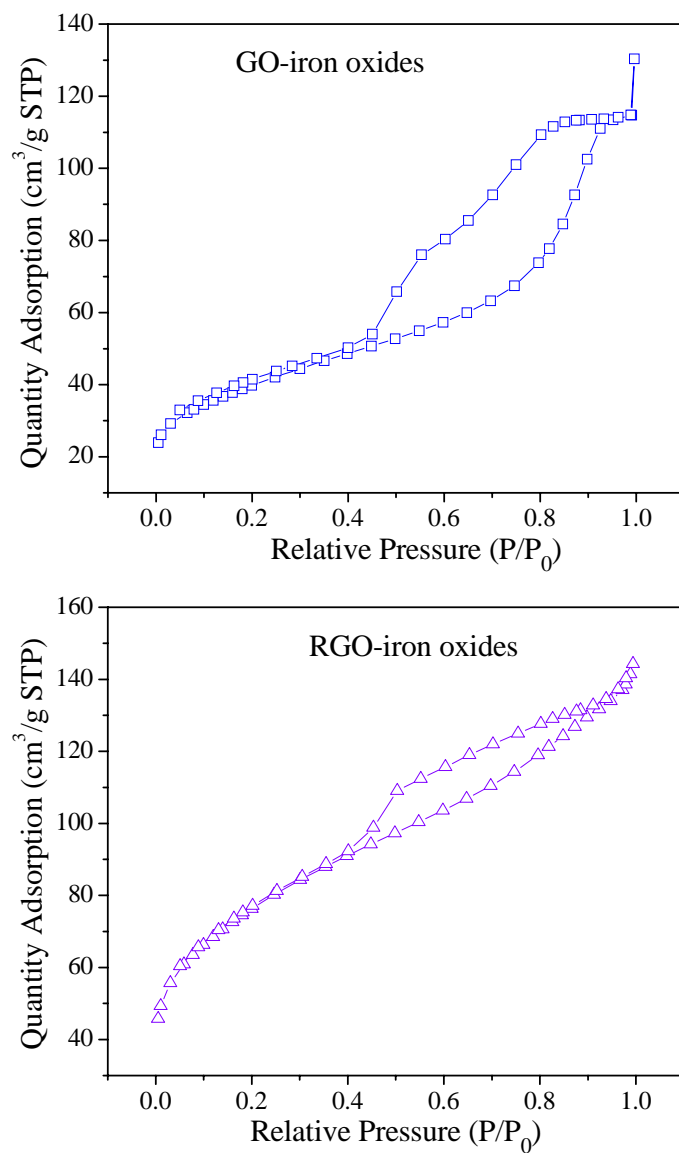


Fig. S5 Nitrogen adsorption and desorption isotherms on GO-iron oxides and RGO-iron oxides.

Surface charge properties

The surface of adsorbent contains a large number of binding sites. Assuming that surface sites (=SOH) are amphoteric, they may become positively charged at low pH due to the protonation reaction on the surfaces (i.e., $SOH + H^+ \rightleftharpoons SOH_2^+$). At high pH values, the surface of adsorbent surfaces becomes negatively charged due to the deprotonation process (i.e., $SOH \rightleftharpoons SO^- + H^+$). Surface sites density can be estimated from acid-base titration.

The acid-base surface chemistry of GO-iron oxides and RGO-iron oxides was investigated by potentiometric titration experiments, which was conducted in a 100 mL Teflon vessel with a polyethylene lid. The vessel was surrounded with a glass jacket to maintain a temperature of 20 ± 1 °C. A Teflon bar was used for stirring. All titrations were conducted using a computer controlled PC-titration system (DL50 Automatic Titrator, Mettler Toledo) with pH electrode (Delta 320). Argon was bubbled successively through NaOH, HClO₄, and distilled water to exclude CO₂(g). Before beginning the titrations, GO-iron oxides and RGO-iron oxides (2.0 g/L) and background electrolyte were added to the vessel and purged with argon for at least two hours. The pH was quickly lowered to approximately 3.5 by addition of 1.4721 M HClO₄. After 1 hour of equilibrium, the suspensions were slowly back-titrated at a variable increment (0.008 up to 0.15 mL, which is automatically adjusted to keep a stable pH change value) with 0.04668 M NaOH solution to pH 10. Each step was allowed to stabilize until the pH drift was less than 0.005 pH unit per minute. Our experience documents that chosen concentrations of acid and base contribute to

decrease titration errors. Titrations were carried out in 0.01 M NaClO₄ background electrolytes. The values of the Gran function (*G*) were calculated as:

$$\text{On the acidic side: } G_a = (V_0 + V_{at} + V_b) \times 10^{-\text{pH}} \times 100 \quad (1)$$

$$\text{On the alkaline side: } G_b = (V_0 + V_{at} + V_b) \times 10^{-(13.8-\text{pH})} \times 100 \quad (2)$$

where, *V*₀ represents the initial volume of the suspension. *V*_{at} and *V*_b are the total volume of acid solution and of OH⁻ added at the different titration points, respectively. The hydroxide ions added to GO-iron oxides or RGO-iron oxides suspension were consumed by the following steps reflected in the Gran plots: neutralization of excess H⁺ in the suspension (before *V*_{eb1}), reactions with the various receivers on GO-iron oxides or RGO-iron oxides surfaces (between *V*_{eb1} and *V*_{eb2}), and adjustment of the system pH (after *V*_{eb2}). Therefore, the specific volumes, *V*_{eb1} and *V*_{eb2}, obtained from the results of linear regression analysis of the Gran plots, corresponded to the equivalent points.

Effect of pH and ionic strength on Pb(II) adsorption onto GO-iron oxides

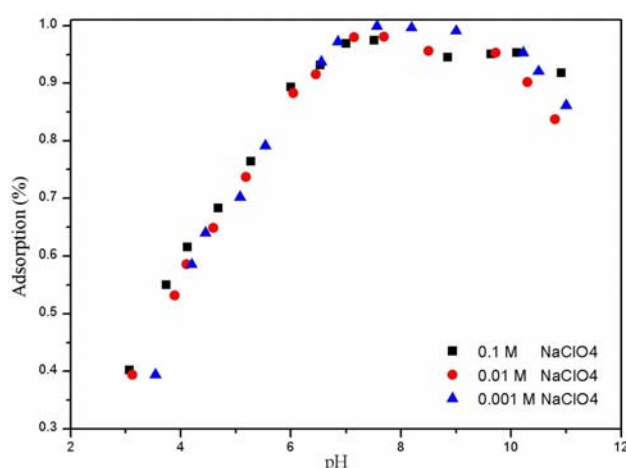


Fig. S6 Effect of pH and ionic strength of Pb(II) adsorption on GO-iron oxides. *T* =

303 K, [C_{Pb(II)}]_{initial} = 40 mg/L, *m/V* = 0.1 g/L.

Table S1 The pH values before and after adsorption of Pb(II) on GO-iron oxides. m/V

= 0.1 g/L, $[C_{\text{Pb(II)}}]_{\text{initial}}=40$ mg/L, $T = 303$ K.

<i>0.1M NaClO₄</i>		<i>0.01M NaClO₄</i>		<i>0.001M NaClO₄</i>	
<i>pH before</i>	pH after	pH before	pH after	pH before	pH after
3.07	3.05	3.12	3.09	3.54	3.48
3.74	3.47	3.89	3.83	4.20	4.16
4.12	4.09	4.11	4.09	4.45	4.39
4.68	4.52	4.59	4.52	5.08	4.99
5.27	5.12	5.19	5.13	5.54	5.48
6.00	5.85	6.05	5.99	6.55	6.49
6.53	6.52	6.45	6.36	6.85	6.70
7.00	6.88	7.15	7.06	7.56	7.43
7.51	6.64	7.69	7.63	8.19	7.81
8.85	8.30	8.50	7.99	9.00	8.49
9.63	8.49	9.72	9.02	10.22	9.68
10.10	9.48	10.30	9.86	10.50	9.96
10.91	10.73	10.80	10.30	11.00	10.59

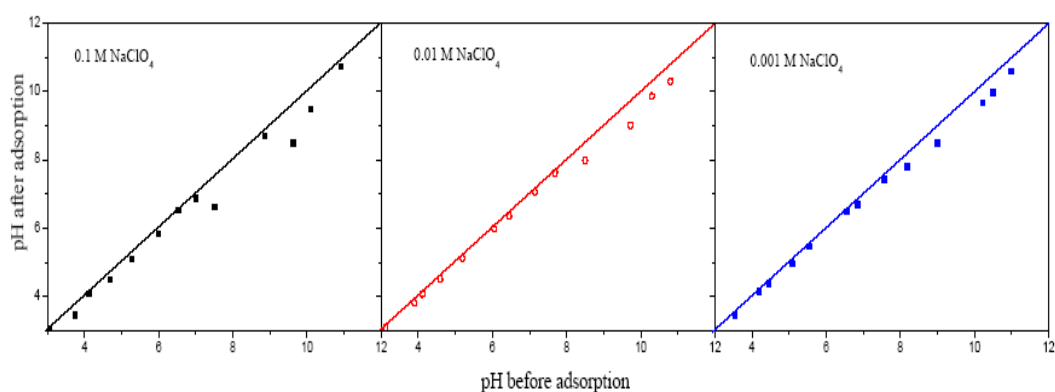


Fig. S7 Variation of pH values before and after adsorption of Pb(II) on GO-iron oxides.

Possible interaction mechanisms of GO-iron oxides-Pb(II), RGO-iron oxides-naphthol,
and RGO-iron oxides-naphthylamine

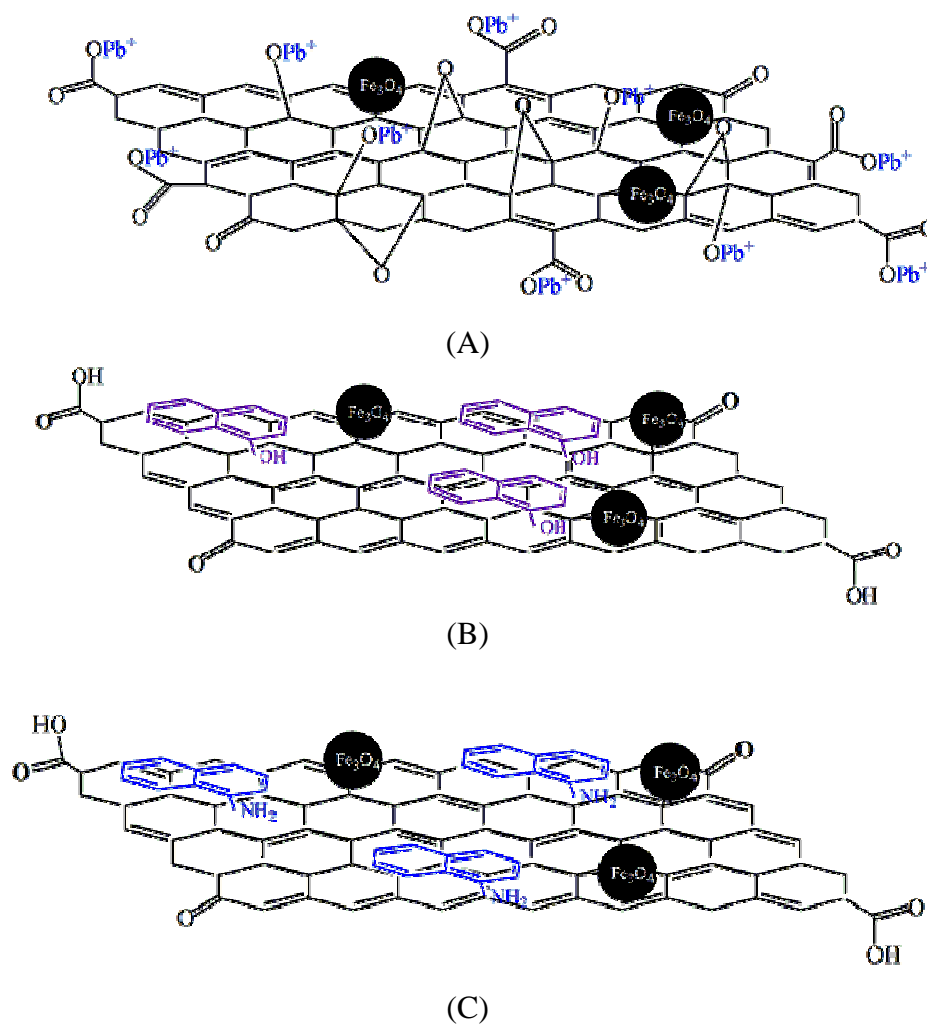


Fig. S8 Schematics showing possible interaction mechanisms, (A) GO-iron oxides-Pb(II), (B) RGO-iron oxides-naphthol, and (C) RGO-iron oxides-naphthylamine.

Linear plots of $\ln K^0$ vs. $1/T$

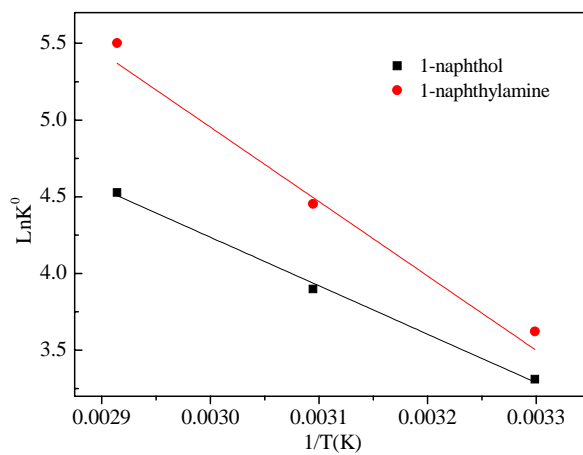


Fig. S9 Linear plots of $\ln K^0$ vs. $1/T$ for the adsorption of 1-naphthol and 1-naphthylamine on RGO-iron oxides, $m/V = 0.1$ g/L, $pH = 6.5 \pm 0.1$, $I = 0.01$ M $NaClO_4$.