Supporting online materials for

Lignosulfonate-modified graphene hydrogel with ultrahigh adsorption capacity for Pb(II) removal

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Experimental details for GO synthesis. GO was prepared by the oxidation of natural graphite powder (325 mesh) according to a modified Hummers' method. Briefly, graphite (3.0 g) was added to concentrated sulfuric acid (70 mL) under stirring at room temperature, then sodium nitrate (1.5 g) was added, and the mixture was cooled to 0 °C. Under vigorous agitation, potassium permanganate (9.0 g) was added slowly to keep the temperature of the suspension lower than 20°C. Successively, the reaction system was transferred to a 35–40 °C water bath for about 0.5 h, forming a thick paste. Successively, 140 mL of deionized (DI) water was added, and the solution was stirred for another 15 min. An additional 500 mL of DI water was added and followed by a slow addition of 20 mL of H_2O_2 (30%), turning the color of the solution from brown to yellow. The mixture was filtered and washed with 1:10 HCl aqueous solution (250 mL) to remove metal ions followed by repeated washing with water and centrifugation to remove the acid.

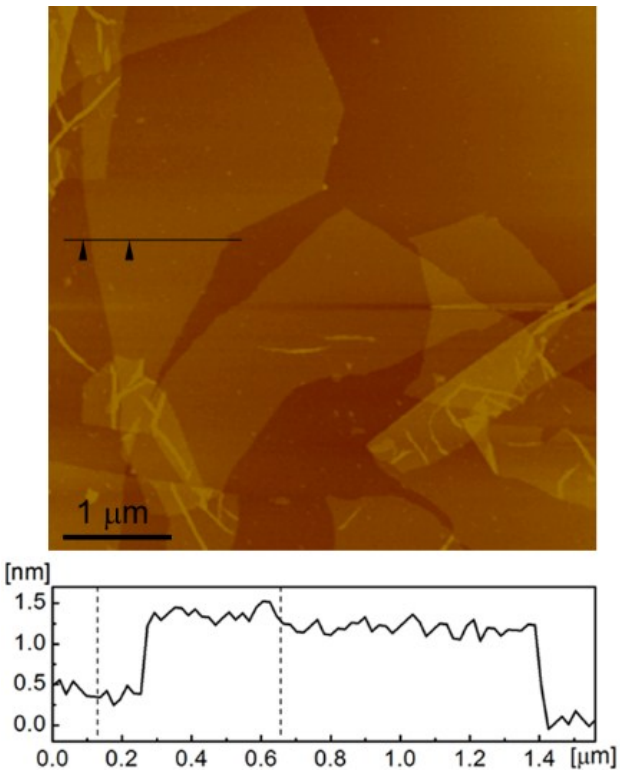

Figure S1. The atomic force microscope (AFM) image and height profile of GO sheets on a freshly cleaved mica surface. The heights of GO sheets range from 0.8–1.0 nm, indicating an individual layer. The lateral sizes of most GO sheets are about several micrometers.

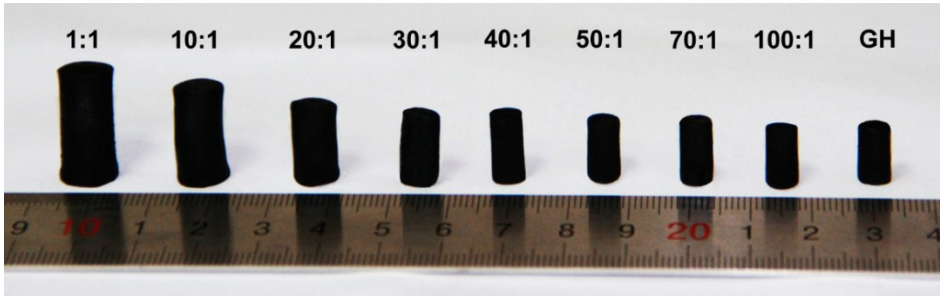

Figure S2. A photograph of free-standing LS-GH with mass ratio of GO:LS being 1:1 , 10:1, 20:1, 30:1, 40:1, 50:1, 70:1, 100:1 and pure GH.

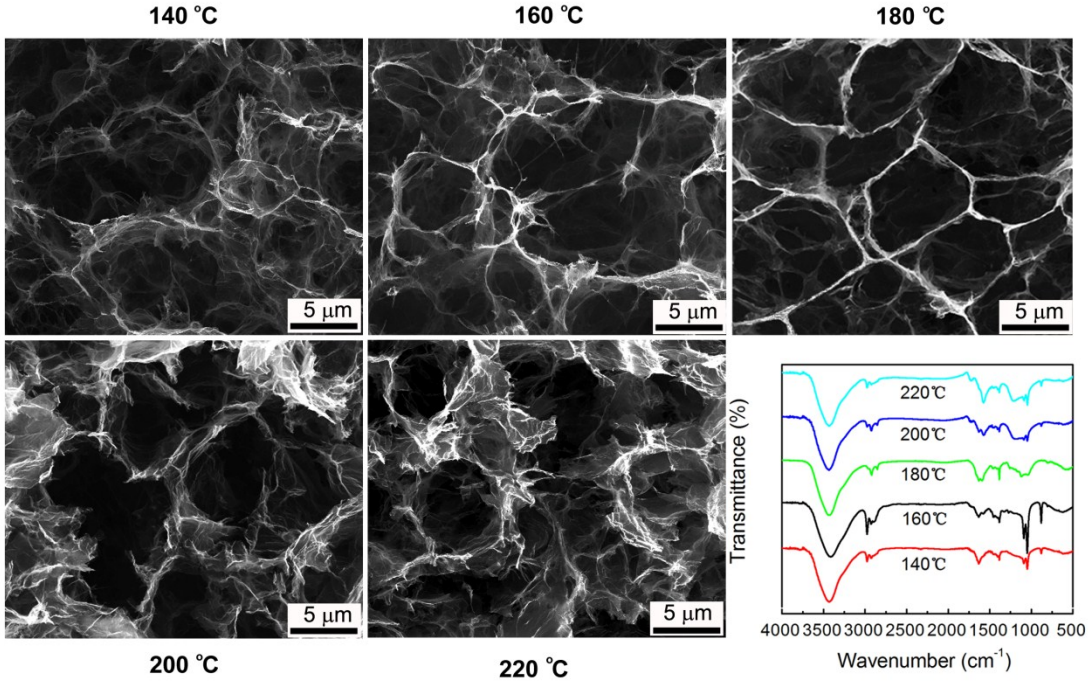

Figure S3. SEM images and corresponding FTIR spectra of LS-GH freeze-dried samples (GO:LS = 20:1, GO = 1 mg mL^{-1}) treated at 140, 160, 180, 200 and 220 °C.

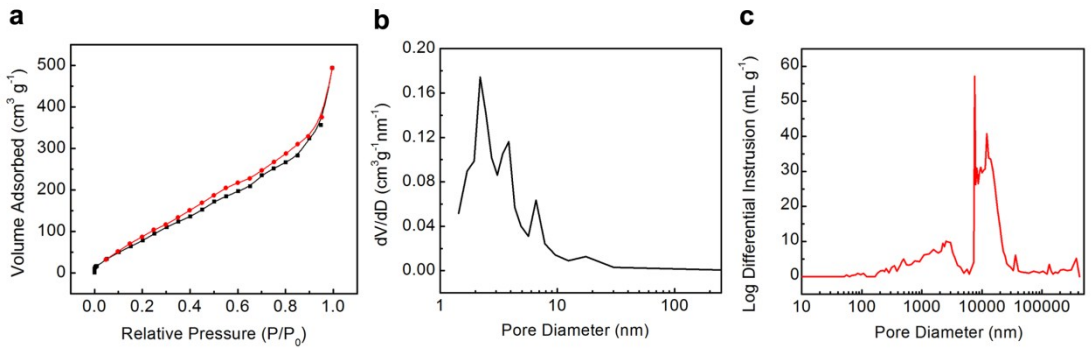

Figure S4. (a) Nitrogen adsorption and desorption isotherms, pore size distributions of LS-GH (GO:LS = 20:1, GO = 1 mg mL^{-1}) calculated by (b) Barrett-Joyner-Halenda (BJH) method in Nitrogen adsorption and (c) mercury intrusion porosimetry (MIP) method.

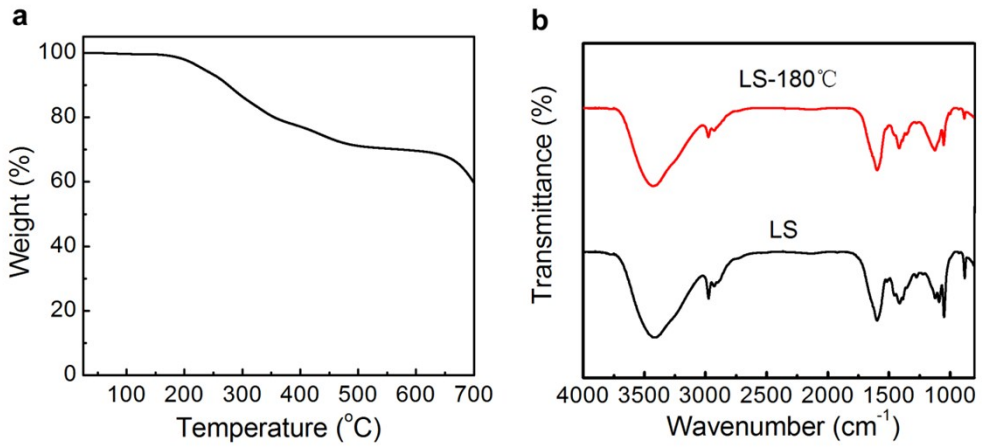

Figure S5. (a) TGA curve of pure LS. (b) FTIR spectra of pure LS before (LS) and after (LS-180 °C) hydrothermal treatment.

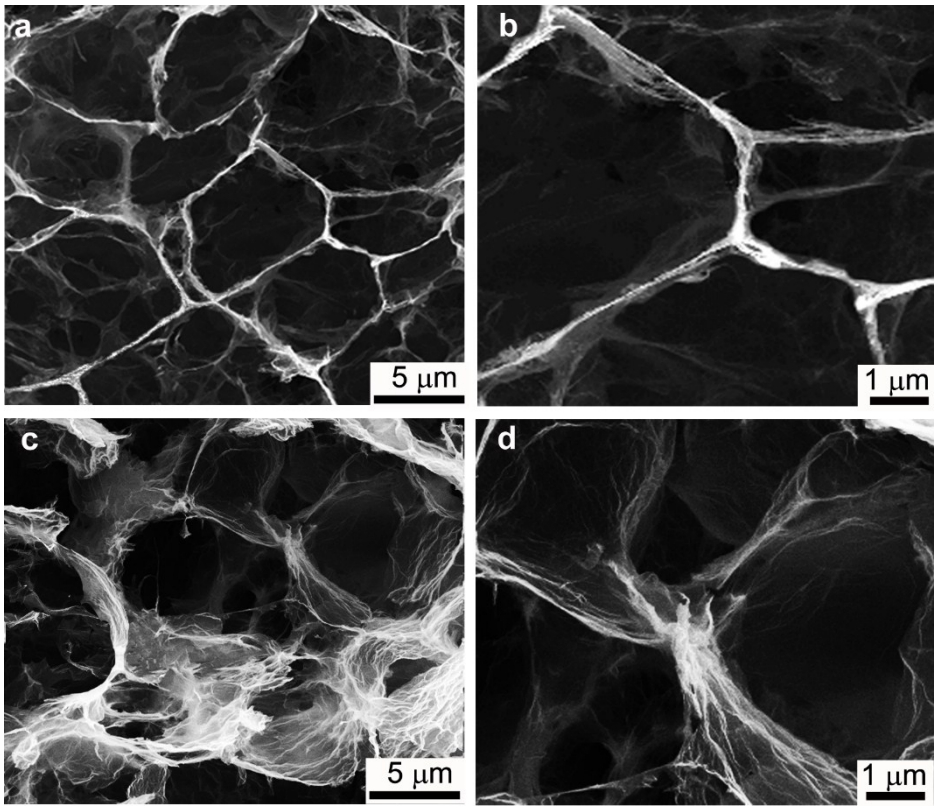

Figure S6. SEM images with different magnifications of (a, b) LS-GH (GO:LS = 20:1, GO = 1 mg mL^{-1}) and (c, d) GH freeze-dried samples.

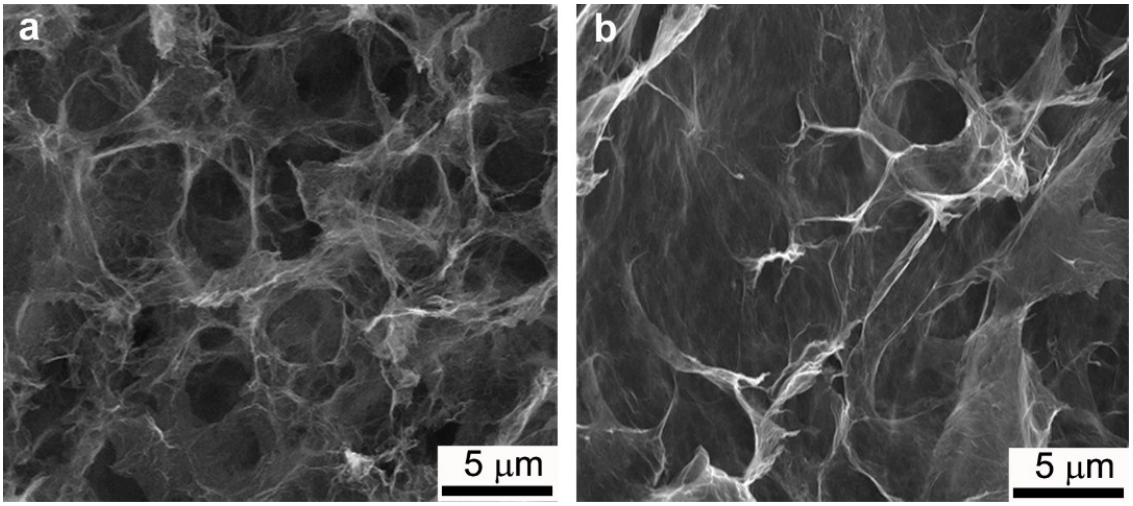

Figure S7. SEM images of LS-GH freeze-dried samples with the mass ratios of GO:LS being (a) 10:1 and (b) 1:1.

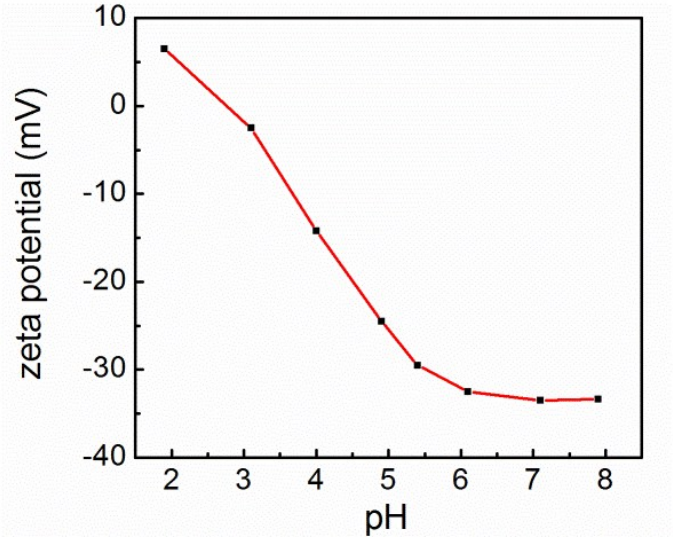

Figure S8. Zeta potentials of LS-GH (GO:LS = 20:1, GO = 1 mg mL^{-1}) under different pH values.

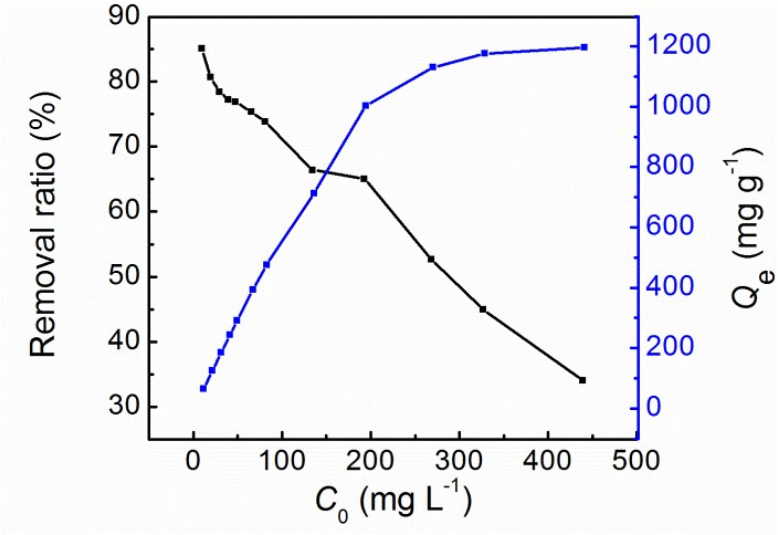

Figure S9. The removal ratio of LS-GH (GO:LS = 20:1, GO = 1 mg mL^{-1}) toward Pb(II) at different initial concentration of Pb(II).

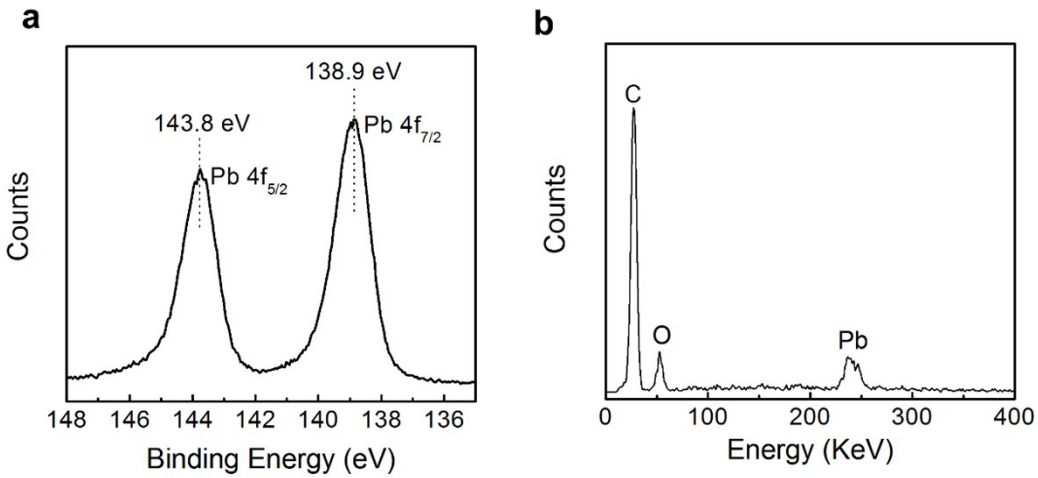

Figure S10. (a) XPS spectra of Pb 4f and (b) EDX spectra after adsorbing Pb (II) on LS-GH (GO:LS = 20:1, GO = 1 mg mL^{-1}).

Table S1. The Pb(II) adsorption capacity of graphene-based and lignin-based adsorbents obtained from the literatures.

Adsorbent	Adsorption capacity ($mg\ g^{-1}$)	Reference
Graphene/α-FeOOH hydrogel	373.8	7
Graphene/δ-MnO_2 aerogel	643.6	8
Oxidized CVD-graphene foam	381.3	9
CVD-grown graphene macroscopic objects	882	10
Graphene aerogel	80	13
Few layered GO sheets	842	14
Graphene/CNT aerogel	104.9	16
Graphene/CNT aerogel /α-FeOOH aerogel	451	17
GO-chitosan hydrogel	90	18
Polydopamine modified graphene hydrogel	336.3	19
GO/CMC porous monolith	76.7	20
Surface functionalized porous lignin powder	188	26
Lignin-based ion-exchange resin	194.5	27
Graft-polymerized lignosulfonate hydrogel	216.3	29
LS-GO-PANI particle	216.4	30
Lignin particle	89.4	31
Lignosulfonate modified graphene hydrogel	1210	Our work

Table S2. Langmuir and Freundlich model fitting parameters for Pb(II) adsorption on LS-GH.

Isotherm model	Parameter	Value
Langmuir model	Q_m (mg g^{-1})	1250
	$K_L \times 10^{-2}$ (L mg^{-1})	5.33
	R^2	0.9931
Freundlich model	K_F (mg g^{-1})(L mg^{-1})$^{1/n}$	115.107
	n	2.2045
	R^2	0.9064

Table S3. Adsorption kinetics fitting results for Pb(II) on LS-GH by pseudo-first-order and pseudo-second-order models.

Isotherm model	Parameter	Value
Pseudo-First-Order Model	Q_e (mg g^{-1})	865
	$k_1 \times 10^{-2}$ (min^{-1})	1.42
	R^2	0.9672
Pseudo-Second-Order Model	Q_e (mg g^{-1})	898
	$k_2 \times 10^{-4}$ [g (mg·min) $^{-1}$]	0.347
	R^2	0.9975