Supporting Information

Effectively Immobilize Lead through Melanotekite Structure Using Low-Temperature Glass-Ceramic Sintering

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(9 pages including 1 text, 3 tables and 5 figures)

List of Supporting Information

Text S1. Synthesis of Fe_3O_4@SiO_2 nano composites

For the synthesis of Fe_3O_4@SiO_2 nano-composites, 2.0 g ferrous chloride hydrate ($FeCl_2 \cdot 4H_2O$, Alfa Aesar China, Ltd) and 5.4 g ferric chloride hydrate ($FeCl_3 \cdot 6H_2O$, Sigma-Aldrich) were dissolved in 100 ml deionized water with magnetic stirring under N_2 atmosphere in a three-necked flask. About 50 ml ammonia ($NH_3 \cdot H_2O$, 30-33 wt. %, E. Merck, Ltd) was added into the $FeCl_2$-$FeCl_3$ solution drop-by-drop to keep the pH at 10.0. The reactive system was maintained at 50 ℃ by water bath for 2 h. Then 125 ml solution containing Fe_3O_4 nanoparticles was ultrasonically dispersed with 300 ml ethanol (95%, BDH Merck Chemical Co.) in a 500 ml beaker flask. After 5 min ultrasonic dispersion, 0.5 ml tetraethyl orthosilicate (TEOS, Alfa Aesar China, Ltd) was added to Fe_3O_4-NH_3-H_2O-Ethanol solution. The reaction solution was kept in ultrasonic condition with ice bath for about 2 h. Raw products were collected by magnet and washed by deionized water for several times. The final product of Fe_3O_4@SiO_2 was obtained after raw material was dried in oven at 60 ℃ for 12 h. The materials obtained are referred as Fe_3O_4@SiO_2 nanoparticles.

Table S1. Quality of the Rietveld refinement analyses in quantifying the phase compositions of the sintered products from the PbO/SiO_2/hematite (PSH) and PbO/SiO_2/magnetite (PSM) system sintered under different temperatures for 5 h. The values indicate the pattern factors and the goodness-of-fit (R_{exp}, R_{wp}, R_p, and *GOF*)

Temperature	Quality of Refinement Analysis (PSH system)			
(℃)	R_{exp} [(a)] (%)	R_{wp} [(a)] (%)	R_p [(a)] (%)	GOF[(b)]
500	2.09	4.99	3.70	2.39
600	2.25	4.62	3.55	2.05
650	2.14	3.76	2.91	1.76
700	2.11	4.11	2.93	1.95
750	2.15	4.06	3.04	1.88
800	2.13	4.16	3.16	1.95
850	2.11	3.89	3.01	1.85
900	2.10	3.90	3.00	1.85

Temperature	Quality of Refinement Analysis (PSM system)			
(℃)	R_{exp} [(a)] (%)	R_{wp} [(a)] (%)	R_p [(a)] (%)	GOF[(b)]
500	2.09	4.81	3.61	2.31
600	2.10	4.64	3.51	2.21
650	2.14	3.83	2.94	1.79
700	2.12	3.81	2.92	1.80
750	2.13	3.88	3.01	1.82
800	2.12	4.19	3.16	1.97
850	2.12	3.93	3.01	1.85
900	2.11	3.97	3.04	1.88

Note:

(a) $R_P = \frac{\sum |Y_i(obs) - Y_i(calc)|}{\sum Y_i(obs)}$, $R_{WP} = \left\{ \frac{\sum \omega_i [Y_i(obs) - Y_i(calc)]^2}{\sum \omega_i [Y_i(obs)]^2} \right\}^{1/2}$, $R_{exp} = \left\{ \frac{\sum M - P}{\sum \omega_i [Y_i(obs)]^2} \right\}^{1/2}$

$$GOF = chi^2 = \frac{R_{WP}}{R_{exp}} = \left\{ \frac{\sum \omega_i [Y_i(obs) - Y_i(calc)]^2}{M - P} \right\}^{1/2}$$

where $Y_{i(obs)}$ and $Y_{i(calc)}$ are the observed and calculated data, respectively, at data point m; M is the number of data points; P is the number of parameters; i is the corresponding data point; ω_i is the weighting given to data point m. The counting statistics is given by $\omega_i = 1/\delta(Y_{i(obs)})^2$, where

$\delta(Y_{i(obs)})$ is the error in $Y_{i(obs)}$.

(b) The goodness of fit (GOF) indicated the fitting quality and it is generally considered that the GOF value between 1.0 and 2.9 is satisfactory.

Table S2. Chemical compositions obtained by EDX for the observed areas in Figure 5

Area	Pb		Fe		Si		O	
	(wt. %)	(Atom %)	(wt. %)	(Atom %)	(wt. %)	(Atom %)	(wt. %)	(Atom %)
1	58.07	15.35	18.98	18.61	8.49	16.55	14.46	49.50
2	59.57	16.29	18.37	18.63	8.56	17.26	13.51	47.82
3	70.80	20.09	4.58	4.82	9.74	20.40	14.88	54.69
4	—	—	73.18	43.87	—	—	26.82	56.13

Table S3. Parameters for the adsorption process by Fe_3O_4@SiO_2 adsorbent treating aqueous solutions of different Pb concentrations

Pb concentration	Initial pH	Adsorbed Pb (mg)	$m_{(adsorbent)}$ (mg)	Removal efficiency (%)
1000 ppm	6.56	18.1	100	67.5
100 ppm	6.60	3.22	100	99.7
10 ppm	6.63	0.36	100	99.5

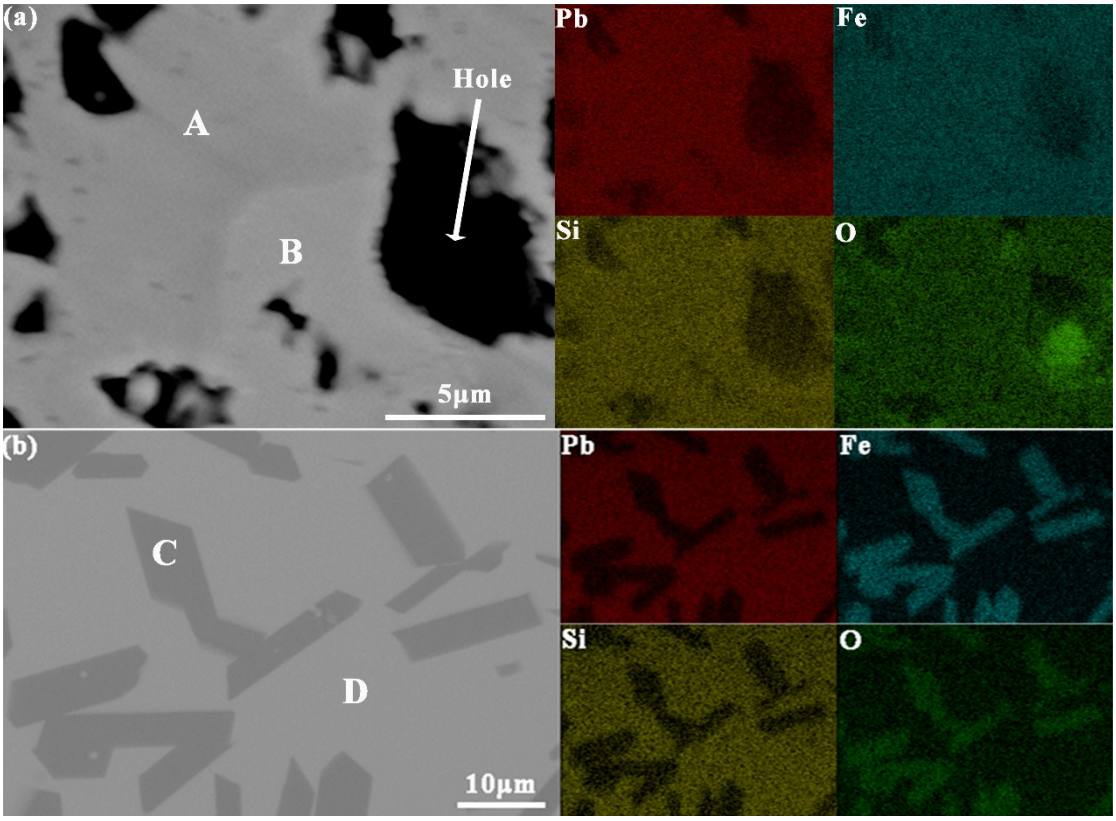

Figure S1. Elemental mapping of PbO/SiO_2/hematite samples sintered at (a) 850℃ and (b) 950℃. Area A and B corresponded to melanotekite and amorphous phase; area C and D corresponded to hematite and amorphous phase.

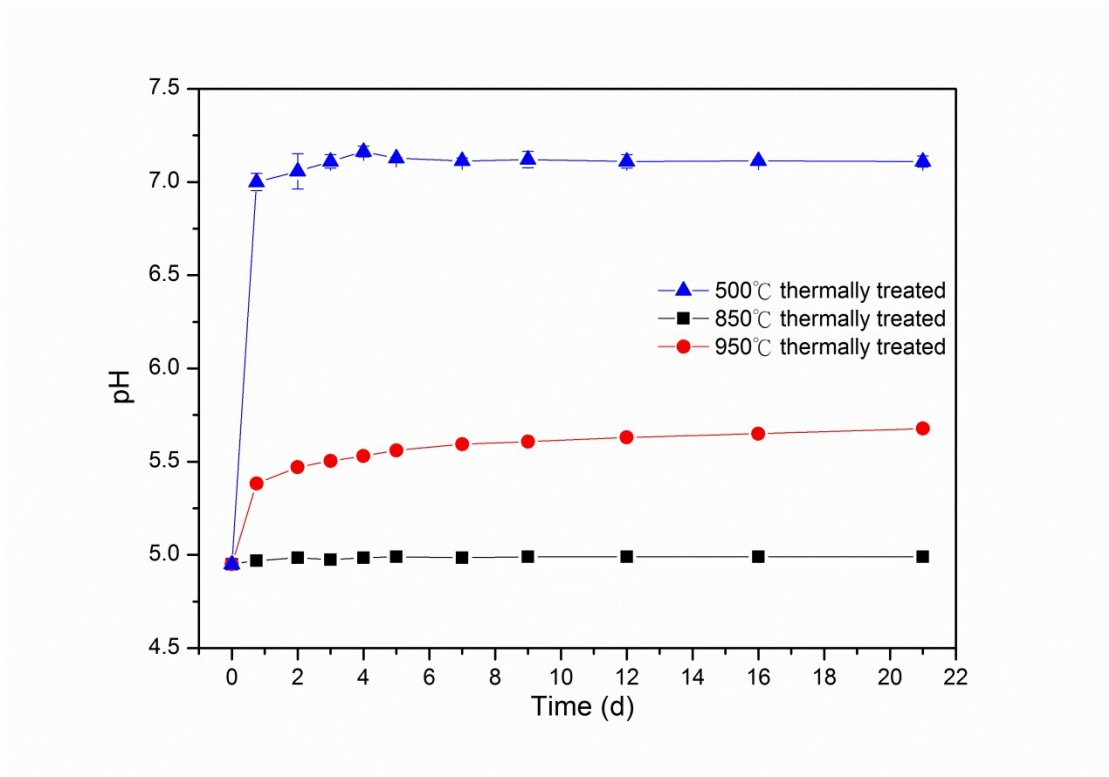

Figure S2. The pH variations in the prolonged leaching tests for the thermally treated PbO/SiO_2/hematite samples sintered at different temperatures.

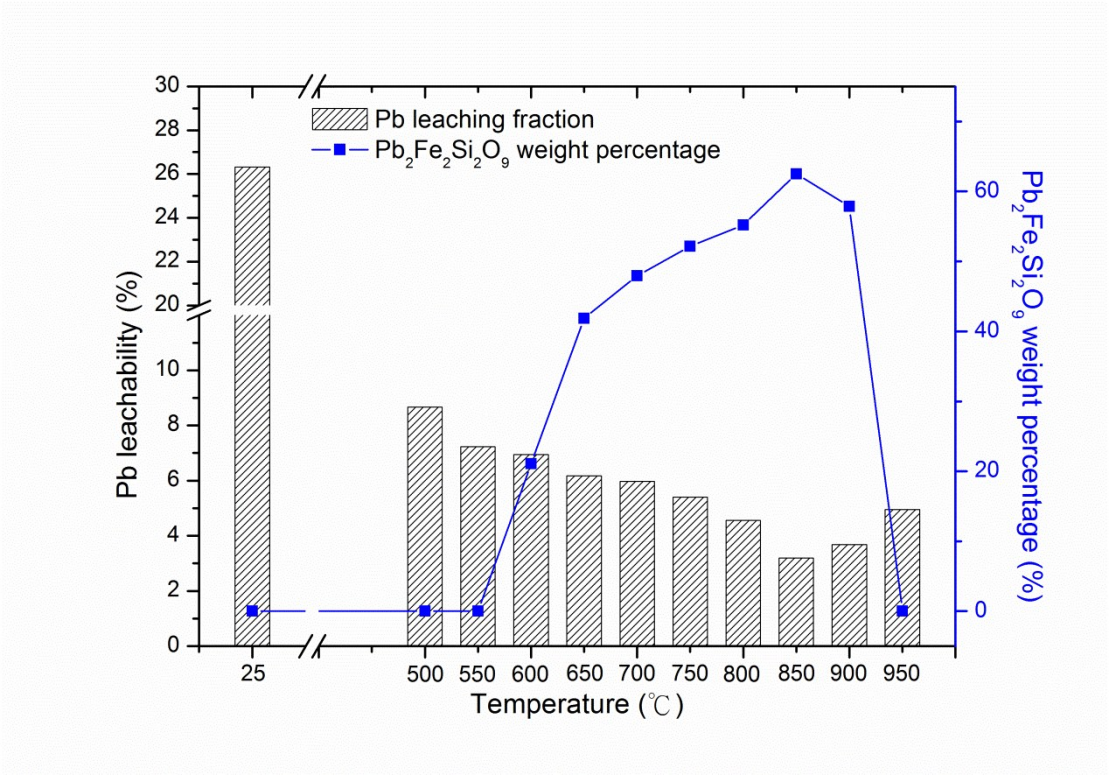

Figure S3. Lead leachability from the thermally-treated PbO/SiO_2/hematite samples after sintering at 500-950 ℃ for 5 h with their corresponding $Pb_2Fe_2Si_2O_9$ contents. The leaching solution was TCLP extraction fluid no. 1 (acetic acid solution) with the pH of 4.9 and the contact time of 18 h.

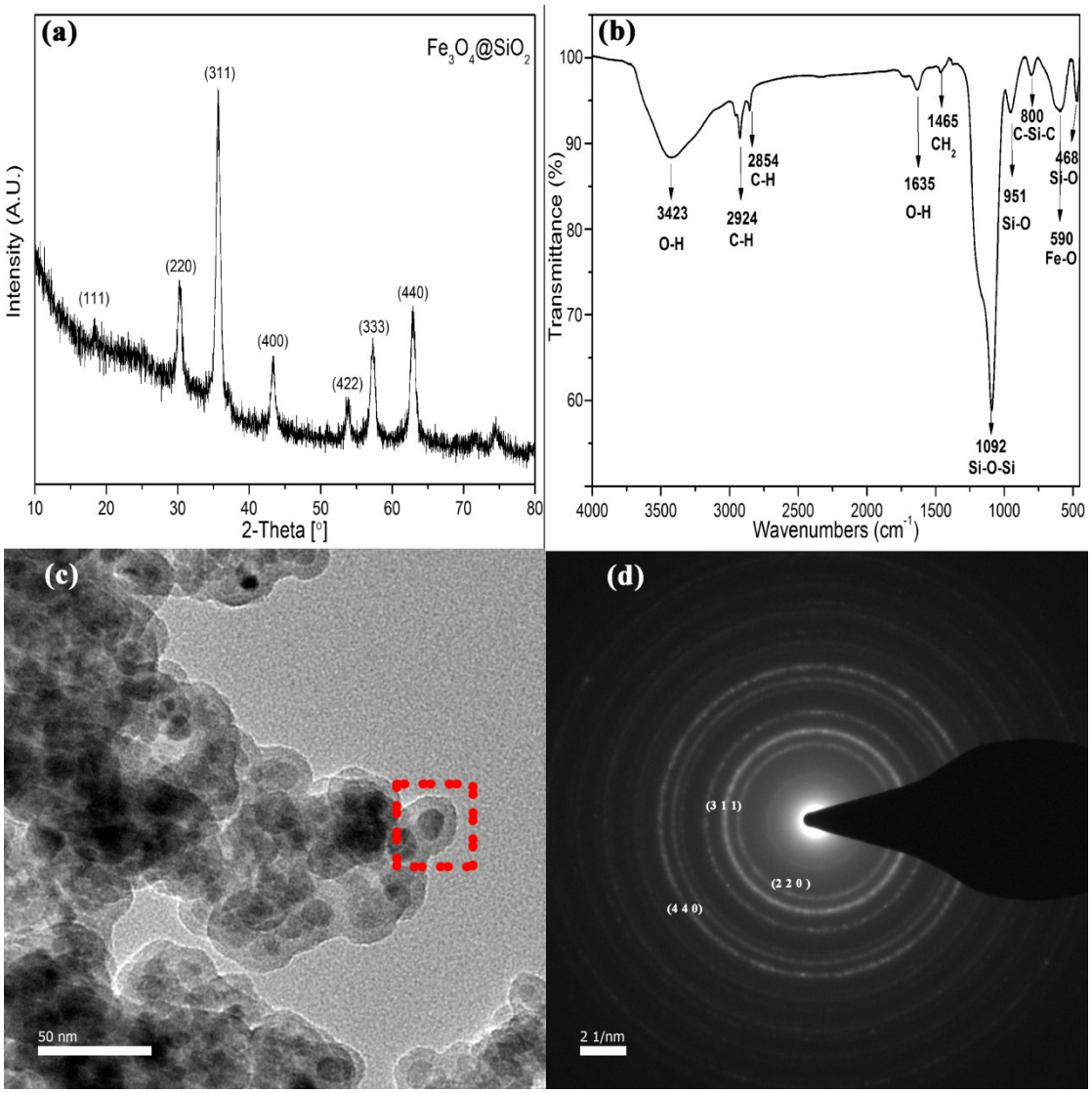

Figure S4. Characterization of synthetic $Fe_3O_4@SiO_2$. (a) XRD pattern of the prepared $Fe_3O_4@SiO_2$; (b) FT-IR spectrum of $Fe_3O_4@SiO_2$; (c) TEM image of the core shell $Fe_3O_4@SiO_2$ structure and (d) electronic diffraction pattern of the as-synthesized $Fe_3O_4@SiO_2$.

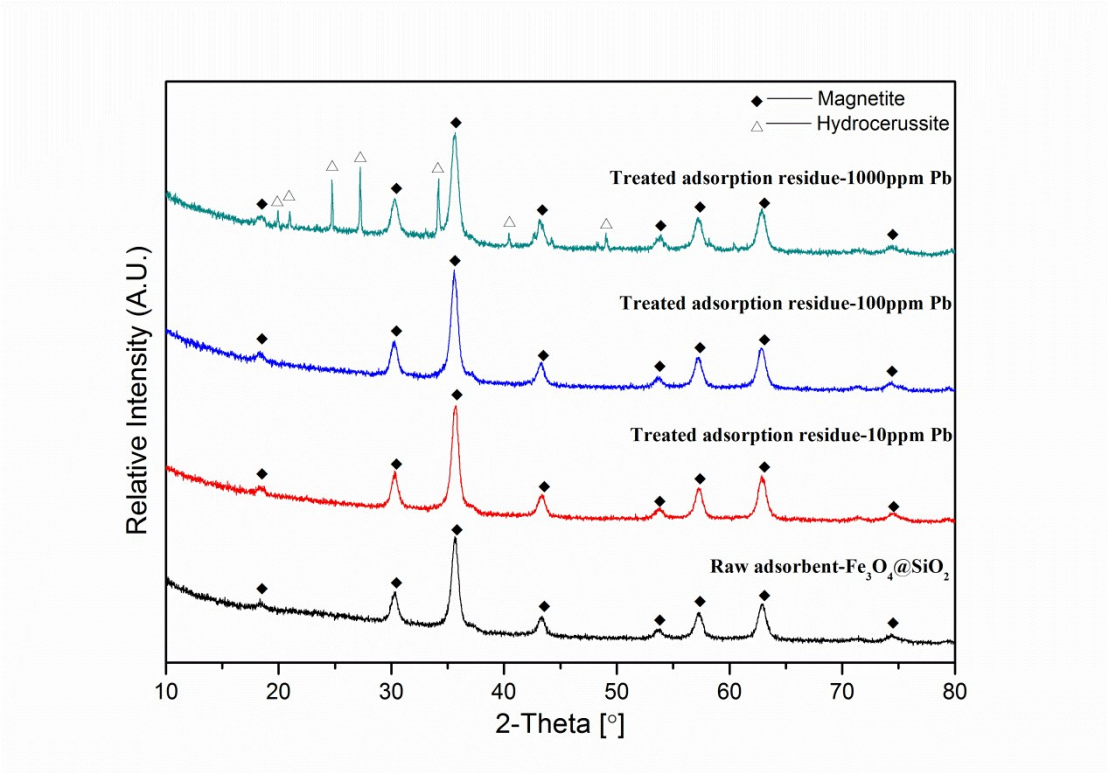

Figure S5. XRD patterns of the spent Fe_3O_4@SiO_2 adsorbents treating different Pb concentration solutions.