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Supporting Information

Amorphous Molybdenum Selenide Intercalated Magnetite as Recyclable Trap for Effective Sequestration of Elemental Mercury

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Sorbent characterizations. The crystallinity of the sorbents was analyzed using X-ray diffraction (XRD, D8 Bruker AXS) with 2θ from 10° to 80° in Cu α (λ = 0.15406 nm) radiation. A vibrating sample magnetometer (VSM, SQUID) was employed to characterize the magnetization of the sorbents. A scanning electron microscopy (SEM, FEI-50, USA) was adopted to characterize the morphologies of the sorbents. The amorphism of the $MoSe_x$ was further evidenced by a transmission electron microscopy (TEM, JEOL 2100F) at 200 kV. The Brunauer-Emmett-Teller (BET) surface area of the sorbents was determined by the N_2 adsorption and desorption method with a BET analyzer (ASAP 2020, Micromeritics, USA). The Raman spectroscopy (LabRAM HR800, HORIBA JOBIN YVON S.A.S) was recorded to further analyze the bond vibrations in the sorbents. With a reference of the C 1s binding energy value of 284.8 eV, X-ray photoelectron spectroscopy (XPS) spectra (Thermo ESCALAB 250Xi) were recorded for fresh and spent $MoSe_x$(inter)Fe_3O_4 [the spent $MoSe_x$(inter)Fe_3O_4 was obtained by pretreating fresh $MoSe_x$(inter)Fe_3O_4 under 500 μg m^{-3} Hg^0 for 12 h].

Kinetic models. pseudo-first-order and pseudo-first-order kinetic models were chosen to study the Hg^0 uptake rate over $MoSe_x$(inter)Fe_3O_4. In the pseudo-first-order model, the Hg adsorption rate was proportional to the difference between the equilibrium capacity and the adsorbed amount at any time, as described as follows:

$$\frac{\mathrm{d}Q_t}{dt} = k_1(Q_e - Q_t) \tag{S1}$$

Equation (3) could be modified to the following equation based on the initial conditions of t=0, Q_t=0:

$$Q_t = Q_e(1 - e^{-k_1 t}) \tag{S2}$$

where the pseudo-first-order kinetic constant (k_1, min^{-1}) can be determined by fitting the adsorption breakthrough curve.

The pseudo-second-order model represents that the surface diffusivity is inversely proportional to the square of concentration variation on sorbent surface, which could be described by equation (S3):

$$\frac{dQ_t}{dt} = k_2(Q_e - Q_t)^2 \tag{S3}$$

Equation (S3) could be modified to the following equation based on the initial conditions of t=0, Q_t=0:

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{1}{Q_e} t \tag{S4}$$

where the pseudo-second-order kinetic constant (k_2, mg g^{-1} min^{-1}) can be determined by fitting the adsorption breakthrough curve.

Thermodynamics. The activation energy for Hg^0 adsorption over $MoSe_x$(inter)Fe_3O_4 was determined by Arrhenius equation with the assistance of k_2:

$$\ln k_2 = \frac{E_a}{R}\left(\frac{1}{T}\right) + C \tag{S5}$$

in which R = 8.314 J mol^{-1} K^{-1}, T (K) is the reaction temperature, and the activation energy (E_a, kJ mol^{-1}) could be hence obtained by simulating a linear relationship between different ln k_2 and 1/T.

Mercury temperature programmed desorption/decomposition (Hg-TPD). To properly determine the Hg^0 species over Hg-laden $MoSe_x$(inter)Fe_3O_4, a Hg-TPD test was conducted. In a typical procedure, 10 mg of $MoSe_x$(inter)Fe_3O_4 mixed with 500 mg of SiO_2 was pretreated in the presence of 500 μg m^{-3} of Hg^0 feed for a continuous 30 min at 50 ^{o}C. Then, the Hg^0 feed was cut off and the Hg-laden sorbent was purged by pure N_2 until the outlet Hg^0 concentration equaled zero. The Hg-TPD test was hence started from 50 ^{o}C to 450 ^{o}C with a heating rate of 10 ^{o}C min^{-1} to monitor the possible decomposition of both HgSe and HgO. The outlet Hg^0 concentration was unceasingly recorded by the mercury analyzer.

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Table S1. Experimental conditions.

Experiments	**Materials**	**Carrier gas (1 L min^{-1})**	**Reaction temperature (^oC)**	**Mass loaded (mg)**	**C_{in} (μg m^{-3})**
Set I	Bare Fe_3O_4; Bare $MoSe_x$; $MoSe_x/Fe_3O_4$; $MoSe_x$(inter)Fe_3O_4	Pure N_2	50	10	500
Set II	$MoSe_x$(inter)Fe_3O_4	Pure N_2	25-125	10	500
Set III	$MoSe_x$(inter)Fe_3O_4	Pure N_2; N_2 + 4%-20% O_2; N_2+0.1%-1% SO_2 (+4% O_2); N_2+8%-20% H_2O (+4% O_2)	50	10	500
Set IV	$MoSe_x$(inter)Fe_3O_4	Pure N_2	50	10	50-1500
Set V	$MoSe_x$(inter)Fe_3O_4	Pure N_2	50	2-50	500
Set VI	$MoSe_x$(inter)Fe_3O_4	Pure N_2	50	10	500
Set VII	Recycled $MoSe_x$(inter)Fe_3O_4	Pure N_2	50	As recycled	500

Table S2. Adsorption performance comparison of various magnetic sorbents.

Sorbents	C_{in} (μg m^{-3})	Sorbent mass (mg)	Hg^0 adsorption capacities (μg g^{-1})	Hg^0 adsorption rate (μg $g^{-1}\cdot min^{-1}$)	Reference
$MoSe_x$(inter)Fe_3O_4 (100%)	500	10	134250	28.3	This work
Fe_3O_4@CuS (100%)	90	5	80730	13.2	11
FeS_x (4%)	120	200	220	0.3	36
H_2S modified Fe-Ti spinel (5%)	100-120	200	690	1.9	37
H_2S modified ilmenite (6%)	100-120	200	170	0.5	38
Mn/γ-Fe_3O_4 (19%)	1000	30	3640	-	40
$(Fe_{3-x}Mn_x)_{1-\delta}O_4$ (100%)	670-1130	25	5100	-	39
Magnetic zeolite supported Ag^0 (20%)	310	~50	44	-	41
Co-MF (100%)	85	500	30	-	42
Magnetic biochar (100%)	85	50	950	-	43

The ratios in the bracelets represent to which extent the sorbents were penetrated.