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## Adsorption of lead on organo-mineral complexes isolated from loess in Northwestern China

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### ABSTRACT

Four types of organo-mineral complex (OMC) samples with different sizes were extracted from loess in northwestern China. OMC, which has a size less than 2  $\mu\text{m}$ , was used to investigate the adsorption behavior of lead. The OMC sample had a porous structure with a surface area of 147.49  $\text{m}^2/\text{g}$ , and the adsorption capacity changed with different conditions of OMC content, pH values of the solution, temperature, and ionic strength. The adsorption isotherms suggested that the adsorption process was favorable, and the maximum adsorption capacity of lead on OMC was 1.5877  $\text{mg}/\text{g}$ , which was calculated by using Langmuir equation. The reaction fits better the pseudo-second-order kinetic equation, and the adsorption behavior could be divided into two sections, thereby indicating that the reaction involved physical and chemical adsorption. The adsorption of lead on OMC was spontaneous and exothermic in nature, and the randomness of the system decreased slightly throughout the entire process. The desorption efficiency was acceptable with purified water and with ultrasonic assistance.

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### 1. Introduction

Heavy metal contamination of agricultural soil occurs with rapid urbanization and industrialization, and is attracting increasing public attention in China. Such attention is warranted because the presence of heavy metals in soils might pose a serious threat to human health through the food chain (Chen et al., 2013; Yu et al., 2012). Lead (Pb), as one of the toxic metals that are commonly discovered in agricultural soils, can easily enter the soil system through various approaches, such as atmospheric deposition, sewage irrigation, and application of fertilizers and pesticides (Finster et al., 2004; Huang et al., 2014; Sun et al., 2006). Soil is known to function as a chemical and as a filter that reduces the environmental effect of Pb introduced into the biosphere. The biosphere is the first line of prevention against Pb contamination of groundwater, which occurs partly through the adsorption process. A high level of Pb in soils may affect crop yields significantly, deteriorate soil quality, and inevitably increase the risk of Pb contamination in farm products (Wang et al., 2006). Consequently, an in-depth understanding of the physicochemical behaviors of Pb in soils is necessary.

As the special particle unit of soil, organo-mineral complexes (OMCs) have long been used to study the migration and distribution of organic matter in soils (Feng et al., 2014; Junet and Basile, 2013). Given the small particle size of OMC, the ability of OMC to adsorb foreign substances, including metals, nitrogen and phosphate, might be different from that of other soil constituents because of various characteristics

of specific surface area and pore structures (Acosta et al., 2009; Bi et al., 2013). Relevant research on the matter has been completed (Acosta et al., 2009; Bradl, 2004; Dultz et al., 2005); however, most studies focus on coarse particles, such as soil aggregates, clay minerals, and sediments. Investigating the reaction behavior between OMC samples and heavy metals is important.

The main factors that affect the adsorption process include metal speciation and concentration, contact time, temperature, and pH value. pH value is the most important environmental factor because of its strong effect on chemical speciation and solubility of heavy metals (Harter, 1983). The major objective of this paper was to identify the composition and surface structure of OMC isolated from loess in northwestern China and to investigate the relationship between environmental parameters and the adsorption process. This paper also investigates the reaction kinetics, isotherms, and thermodynamics of the adsorption of lead on OMC. This research is significant in improving the understanding of the adsorption mechanism between OMC and Pb, and in further evaluating the transformation behavior of Pb in the pedosphere or ecosystem in a reasonable manner.

### 2. Materials and methods

#### 2.1. Isolation of OMC

Loess samples were collected from a 0–20 cm deep surface horizon of farmland located in Shaanxi University of Science and Technology, Xi'an, China. The samples were transported to the laboratory in polyethylene bags and divided into four subsamples through the sedimentation

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method. The division of the subsamples is as follows: <2  $\mu\text{m}$  particles, 2–20  $\mu\text{m}$  particles, 20–200  $\mu\text{m}$  particles and >200  $\mu\text{m}$  particles (Bradley, 2004). The <2  $\mu\text{m}$  particles were used in the study, and all samples were air dried and kept in a desiccator over silica gel for subsequent utilization.

## 2.2. Physicochemical analysis of OMC

The composition of OMC was determined by chemical analysis. A Brunauer–Emmett–Teller (BET) surface analyzer (Nova2200e, QUANTACHROME) was used to measure nitrogen adsorption at 77 K. The surface area of OMC was calculated from the isotherms by using the BET equation. The mineral phases of OMC were revealed with an X-ray diffractometer (D/max2200PC, RIGAKU) that ran at 40 kV and 40 mA by using a Cu K $\alpha$  radiation. The surface morphology of OMC was identified by using a scanning electron microscope (TM1000, HITACHI). The Fourier transform infrared (FT-IR) spectra were obtained in the range of 4000–400  $\text{cm}^{-1}$  on a spectrometer (Vector22, BRUKER) with KBr pellet method at room temperature.

## 2.3. Adsorption studies

A stock solution of 1000 mg/L of Pb was prepared from  $\text{Pb}(\text{NO}_3)_2$ . Experimental solutions of the desired concentration were obtained by diluting the stock solution using purified water. The effects of OMC content, original pH value of solution, temperature, and ionic strength on adsorption behaviors were investigated. A constant-temperature air bath shaker (ZD-85, Changzhou, China) was used to control the desired temperature. The pH values of solution were adjusted by adding HCl or NaOH solutions. All pH measurements were performed by using a pH meter (pHS-3C, Shanghai, China).

The OMC sample was placed in a 250 mL conical flask with a stopper; the flask contained 20 mL of the experimental solution of known concentration. The suspensions were shaken at 120 rpm for 4 h. The supernatant liquids were then centrifuged at 4000 rpm for 5 min and then filtered with 0.45  $\mu\text{m}$  filter. All the experiments were performed for three times. The initial and final concentrations of Pb were measured by using a flame atomic adsorption spectrometer (Z-2000, HITACHI). The obtained data were applied to calculate the adsorption capacity ( $q_e$ , mg/g) by using the following relationship:

$$q_e = (C_0 - C_e) / m \times V \quad (1)$$

The removal efficiency (%) of lead on OMC can be calculated as follows:

$$\text{Removal efficiency}(\%) = (C_0 - C_e) / C_0 \times 100 \quad (2)$$

where  $C_0$  and  $C_e$  (mg/L) are the initial and equilibrium concentrations of Pb, respectively,  $V$  (mL) is the volume of the solution, and  $m$  (mg) is the content of OMC.

## 2.4. Desorption studies

Desorption experiments were carried out by using purified water as a desorbing agent with ultrasonic assistance. The effect of the different volumes of purified water (50, 100, 150 and 200 mL) and ultrasound power (25, 50, 75 and 100 W) on desorption efficiency was studied by contacting 0.1 g of Pb-loaded OMC samples. The mixture was stirred until desorption equilibrium was achieved at 25  $^{\circ}\text{C}$  and then filtered. The concentration of desorbed Pb in the filtrate was checked. The desorption efficiency of Pb was calculated by using the following equation:

$$\text{Desorption efficiency}(\%) = C_d V_d / q_e m \times 100 \quad (3)$$

where  $C_d$  (mg/L) is the concentration of Pb in the purified water at desorption equilibrium,  $V_d$  (L) is the volume of purified water,  $m$  (g) is the mass of OMC used for desorption studies, and  $q_e$  (mg/g) is the adsorption capacity of OMC for Pb.

## 3. Results and discussion

### 3.1. Characterization of OMC

#### 3.1.1. BET and XRD

The BET surface area of OMC derived from loess was 147.49  $\text{m}^2/\text{g}$ , which is lower than that of the loess sample described in our previous investigation (Fan et al., 2013), thereby indicating acceptable adsorption sites on OMC samples. The XRD pattern of OMC is shown in Fig. 1. The crystal phase of the OMC was mainly composed of quartz (26.64 $^{\circ}$ ), muscovite (34.94 $^{\circ}$ ), clinocllore (19.80 $^{\circ}$ ), and polyolithionite (27.94 $^{\circ}$ ). Results showed that the clay minerals of the OMC were poorly organized, and many disoriented particles were stacked randomly.

#### 3.1.2. SEM

To determine some physical characteristics of the surface morphology of OMC, a morphological investigation was conducted by SEM. Fig. 2 displays the SEM images of OMC, and micrometric particles of different sizes could be observed. The surface of the OMC, which involved a majority of tiny pores, was clearly uneven.

#### 3.1.3. FT-IR

The nature of OMC surface configuration was examined by spectroscopic analyses, and the FT-IR spectra are presented in Fig. 3. In the spectra, the typical bands at 3471 and 3416  $\text{cm}^{-1}$  were related to the stretching vibration of O–H groups likely indicating the presence of  $\text{H}_2\text{O}$  in the OMC. A band within the 1620–1639  $\text{cm}^{-1}$  region was also visible and might be attributed to CONH or the bending vibration of the hydroxyl and interlayer water molecules. The peak recorded at 1452  $\text{cm}^{-1}$  might be attributed to carbonate, and a band at 877  $\text{cm}^{-1}$  could be attributed to Si–O bending vibrations.

### 3.2. Adsorption process

#### 3.2.1. Influential factors

**3.2.1.1. Effect of OMC content on lead adsorption.** The effect of OMC content on lead adsorption was studied by changing the OMC content from 0.01 to 0.4 g at room temperature; results are presented in Fig. 4. The removal rate increased evidently from 5.05 to 92% within the OMC content from 0.01 up to 0.2 g and remained nearly constant thereafter. This condition occurred because the number of available

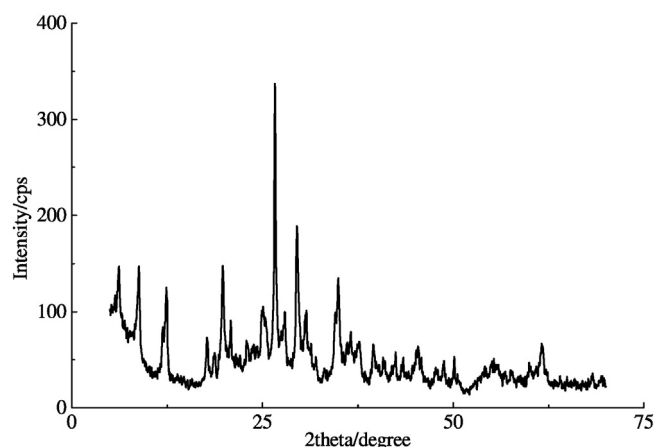


Fig. 1. The X-ray diffraction pattern of OMC samples.

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