



Research article

Effects of modified zeolite on the removal and stabilization of heavy metals in contaminated lake sediment using BCR sequential extraction

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ARTICLE INFO

Article history:

Received 26 February 2016

Received in revised form

18 April 2016

Accepted 23 April 2016

Available online 29 April 2016

Keywords:

Sediment

Zeolite

Heavy metal

BCR sequential extraction

Speciation

ABSTRACT

Sediment can be applied on land as a soil conditioner. However, toxic substances such as heavy metals within the sediment often lead to soil contamination if no proper management is conducted prior to land application. In order to reduce the bioavailable portion of heavy metals such as Pb, Cu, Zn and Cd, zeolite as a kind of stabilizer was investigated on the effect of metal stabilization in sediment. Zeolite was firstly modified and screened to get the best condition for removal of heavy metals. Results showed that the granulated zeolite with NaCl conditioning had the highest CEC and metal sorption. Using BCR sequential extraction, the selected modified zeolite effectively stabilized Pb, Cu, Zn and Cd in sediment to different extents. It was most suitable for Cd stabilization by reducing its acid exchangeable fraction while increasing the contents of the reducible and residual fractions. Modified zeolite also immobilized Cu, Zn and Pb in sediment by enhancing one stable fraction while decreasing the acid exchangeable fraction.

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1. Introduction

Lake sediment is a crucial composition in lake ecosystem for its role as the storage bank for organic and inorganic nutrients, and for toxics and hazards simultaneously. Heavy metals, as a kind of persistent toxic substances, enter into lakes through various pathways and accumulate in sediment by adsorption, complexation or sedimentation with suspensions or sediments in waters (Ji et al., 2004). Heavy metals in sediment are usually in a certain dynamic equilibrium with the above water body, and could enter the water when the surrounding environment changes, thus resulting in heavy metal contamination in waters (Müller and Pluquet, 1998).

For its characteristic of being nutrient-rich, a conventional way of utilizing the lake sediment is to apply it on land to increase soil fertility, use it in wetlands for biological habitation, or use it to remedy seriously disrupted soils for development of surface plants. However, most land application is by simply mixing the sediment with soil or applying it onto the soil surface (Bedell et al., 2006; Chen et al., 2002; Perin et al., 1985). Due to the fact that heavy metals accumulate in sediments in various forms, part of them are likely to be mobilized to become environmentally toxic in the

course of utilization. Within the exchangeable or carbonates-bond fractions of heavy metals released by sediment, only less than 1% is considered safe to the environment, and over 50% of the total amount possesses high risk and possibly enters the food chain (Perin et al., 1985).

Traditionally, using stabilizer such as fly ash (Rijkenberg and Depree, 2010; Tomasevic et al., 2013), ground granulated blast-furnace slag (Wang et al., 2015), iron/manganese oxides (Wang et al., 2015), phosphates (Rijkenberg and Depree, 2010), compost (Rijkenberg and Depree, 2010), limestone (Wildt et al., 2004) or Ca-rich clay minerals (Yin and Zhu, 2016) can minimize the mobile metals in the environment, hence controlling the spread of pollution. Some stabilizers are found to be potentially toxic to environment. For example, red mud is usually abundant with chromium and Al³⁺, and sludge from waste water treatment plant has arsenic of high quantity (Müller and Pluquet, 1998). Therefore, efficiency, source, cost and potential toxicity should be taken together to suggest a suitable stabilizer for a specific heavy metal remediation project. In recent years, zeolite has received much attention in scientific community due to its special physico-chemical property, accessible source and low cost, and is being widely used in industry, agriculture and pollution control. Zeolite has a crystal structure that is made by silicon (aluminium) oxygen tetrahedron arranged into three dimensional lattice (Breck, 1974). Various large or small holes and channels in the structure give rise

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to the great openness of zeolite, making the material selectively absorb molecules of appropriate size. In addition, substitution of some of the silicon (Si) with aluminium (Al) (or other metals) leads to a negative charge on the framework, with cations (principally sodium (Na^+), potassium (K^+), and calcium (Ca^{2+}), less frequently lithium (Li^+), magnesium (Mg^{2+}), strontium (Sr^{2+}) and barium (Ba^{2+})) distributed within the pore structure (Pabalan and Bertetti, 2001) and have weak connection with the framework (Breck, 1974). These structure properties make zeolite possess great ion exchange ability, subsequently resulting in its strong and selective adsorption of some particular metal ions. Examined by X-ray Powder Diffraction, it was assumed that the fraction of clinoptilolite was the main reason for ion exchange and adsorption in all kinds of zeolite components (Mondale et al., 1995) and deemed to be the most effective ion exchange and selection material (Humidpour et al., 2010). The metal removal efficiency of clinoptilolite for different metals follows the order: $\text{Pb}^{2+} > \text{Cd}^{2+} > \text{Cs}^+ > \text{Cu}^{2+} > \text{Co}^{2+} > \text{Cr}^{3+} > \text{Zn}^{2+} > \text{Ni}^{2+} > \text{Hg}^{2+}$ (Tomasevic et al., 2013). It was reported that zeolite could stabilize heavy metals in contaminated soils and reduce metal transfer to upper part of plants (Humidpour et al., 2010; Sun et al., 2014). Recent work utilizing zeolite for metal removal and remediation mainly focused on sewage (Ashmawy et al., 2012; Zorpas et al., 2000), soil (Li et al., 2009; Mahabadi et al., 2007; Shi et al., 2009), and water (Egashira et al., 2012; Wingenfelder et al., 2005). Research of utilizing zeolite to stabilize heavy metals in sediment is yet limited. Using the sediment generally accepted as 'waste' for land application while keeping the accompanied risks to minimal is an optimal and effective solution for sediment reuse, conforming to the demands of resource utilization.

Most of natural zeolites occur in forms of zeolitic-rich tuffs with zeolitic composition accompanied with other impurities (Delkash et al., 2015). Pretreatment of zeolite to cleanse the impurities or alter the properties would affect the adsorption process of zeolite (Song et al., 2015). Zeolite with modifications using mineral acid or base treatment has been proposed to improve zeolite surface areas and porosity (Christidis et al., 2003). Therefore, the objectives of the study were (a) to investigate the impact of modification on the performance of metal removal by zeolite in aqueous environment and (b) to assess the stabilization effect of zeolite on heavy metals in sediment. Samples of sediment from Dongting Lake known for heavy metal contamination (Li et al., 2013) were collected for experiments. For a better metal stabilization effect, zeolite was modified and screened for particle size and modification condition. Following that, BCR sequential extraction was conducted to study the influence of modified zeolite on the speciation of Pb, Cu, Zn and Cd in the sediment.

2. Materials and methods

2.1. Sediment description

Sediment samples were collected from Dongting Lake, Hunan. A map of the sampling site was provided as Fig. 1. Dongting Lake is one of the biggest natural water resources in South China, and due to developed economy in the adjacent area, it suffers from high loads of untreated wastewater (Li et al., 2013). The upper 0–10 cm depths of sediments were randomly collected and then homogenized, after which subsamples were kept at 4 °C prior to laboratory analysis.

Pseudo-total trace metal contents were assessed on sample triplicate after nitric acid digestion employing US EPA standard method (EPA3050B, 1996). Metal content was determined by Atomic Absorption Spectroscopy (AAS) (AAnalyst700, Perkin Elmer). Potential ecological risk index (PER) developed based on

sedimentary theory was introduced to assess the ecological risk degree of heavy metals in the present sediment. Risk index (RI) can be calculated by the following formulas proposed by Hakanson (Hakanson, 1980):

$$C_f^i = C_D^i / C_B^i \quad (1)$$

$$E_r^i = T_r^i \times C_f^i \quad (2)$$

$$RI = \sum_{i=1}^m E_r^i \quad (3)$$

Where RI is the sum of the potential risk of individual heavy metal, E_r^i is the potential risk of individual heavy metal, T_r^i is the toxic-response factor for a given metal, C_f^i is the contamination factor, C_D^i is the present concentration of heavy metals in sediments, and C_B^i is the pre-industrial record of heavy metal concentration in sediments. Based on the Hakanson's approach, the toxic-response factors for Pb, Cu, Zn and Cd are 5, 5, 1, and 40, respectively. In this study area, the pre-industrial concentration records for Pb, Cu, Zn and Cd were replaced by their corresponding background values, namely 23.3, 20.2, 83.3 and 0.33 mg/kg (Li et al., 1986).

2.2. Zeolite modification and screening

The natural zeolite used in the experiments came from Gongyi, Zhengzhou (Henan Province, China). Three grain size fractions were used as received from the supplier: bulk particle of 2–2.5 mm (named as fraction I), granulated particle of 0.5–1 mm (named as fraction II), and powdered zeolite of <0.5 mm (named as fraction III). Samples were washed with distilled water to remove soluble salts possibly present prior to conditioning and capacity measurements. The pH of the zeolite in three forms was tested using pH meter (soil/water = 1:5).

Natural zeolite different in three forms were activated using HCl, NaOH and NaCl solutions to prepare for the modified zeolite. The experimental protocol was modified from a study done by Lin et al. (1998). Five grams of zeolite of each form were immersed in 100 ml solutions in conical flasks containing 2 mol/L HCl, NaOH or NaCl. The conical flasks were then shaken in a water bath at 75 °C for 3 h, followed by centrifugation at 3000 rpm for 15 min. The liquid supernatant was decanted and all treated zeolites were washed with de-ionised water for several times and then dried in muffle at 105 °C for 2 h. Following that, the modification was repeated once and the zeolites were settled for 24 h before centrifugation, washing and drying. Identification of mineral species in the natural and modified zeolite was carried out by X-ray diffraction (XRD) of the random-oriented powder samples using Japan Rigaku 2555 X-ray diffractometer (Cu 40 kV, 250 mA). Selected natural and modified zeolites samples were also submit for BET analysis to examine the surface area (SA) and pore volume (PV).

Cation exchange capacity (CEC) of the natural and modified zeolites was determined by extracting NH_4^+ with 1 mol/L NaCl (pH 7) from NH_4^+ -saturated sample prepared by equilibrating with 1 mol/L ammonium acetate (pH 7) at room temperature. The exact concentrations of ammonium ions in the solutions were determined using US EPA standard method (EPA5220, 1999).

The metal removal efficiency of the natural and modified clinoptilolite was evaluated by polluting the sorbent with 0.1 M KNO_3 solutions containing Pb(II), Cu(II), Zn(II) and Cd(II) at the concentration of 400 mg/L each with a solid/solution ratio of 1:100 in conical flasks. Then the conical flasks were put in a steady rate

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