



Influence of organic acids on kinetic release of chromium in soil contaminated with leather factory waste in the presence of some adsorbents



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HIGHLIGHTS

- We studied the effect of adsorbents and organic acids on release of Cr in soil contaminated with leather factory waste.
- In all treatments, organic acids released more Cr than inorganic salt.
- In the presence of organic acids, Cr absorption by adsorbents increased.
- The organic acids were effective in reducing the mobile fractions of Cr in all treatments, whereas.
- Nanoparticles have a higher adsorption capacity for Cr rather than clay minerals.

ARTICLE INFO

Article history:

Received 30 October 2015

Received in revised form

10 April 2016

Accepted 18 April 2016

Available online 30 April 2016

Handling Editor: X. Cao

Keywords:

Chromium

Leather factory waste

Organic acid

Clay mineral

Nanoparticles

ABSTRACT

In this study, batch experiments were conducted to investigate the effects of nanoparticles (NPs) (MgO, ZnO, TiO₂) and clay minerals (bentonite, zeolite) on the release of chromium (Cr) from leather factory waste (LFW) and LFW treated soil using organic acids. Chromium release from all treatments was studied in the presence of citric acid, oxalic acid and CaCl₂ solutions. The results showed that, in all treatments, organic acids released more Cr than inorganic salt (CaCl₂). The release of Cr by citric acid was higher than that by oxalic acid. In LFW treated soil and LFW, the release of Cr from the all treatments with NPs was less than that from the clay mineral treatments. On the other hand, in the presence of organic acids, Cr release by NPs and clay minerals decreased. Two kinetic models including pseudo-first- and pseudo-second-order model were tested to describe the time dependent Cr release data. Among the kinetic models used, the pseudo-second-order model generally gave the best fits to experimental data. Before and after release experiments, Cr in LFW, treated LFW, control soil and LFW treated soils were fractionated. In all treatments, the greatest amounts of Cr were found in the residual fraction (RES). The organic acids were effective in reducing the exchangeable (EXC), bound to organic matter (OM) and bound to carbonate (CAR) fractions of Cr in all treatments, whereas, after release of Cr from treated soils, Cr remained mainly in the RES fraction. The application of NPs and clay minerals in soil led to a significant transformation of Cr from mobile fractions to the RES fraction. Therefore, organic ligands played a dominant role in mobility and bioavailability of Cr and the removal of Cr by adsorbents.

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1. Introduction

Soil contamination by chromium (Cr) is a significant environmental concern, hence recognizing its behavior in soils is of increasing scientific concern. The main sources of Cr in soils and water are anthropogenic activities such as leather tanning, mining,

electroplating, wood treatment and chemical industries (Fahim et al., 2006). Therefore, wastewater and sludge from these industries have accelerated soil contamination by Cr. Leather factory is one of the most pollution sources of Cr in the soil. Many researchers have reported Cr contamination of soils from the vicinity of leather industrial sites (Singh et al., 2001; Bini et al., 2008). Pollution problem increases when heavy metals are mobilized into the soil solution and transported to the ground water.

Low molecular weight organic acids (LMWOAs) have a major

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impact on the availability and toxicity of heavy metals (Xu and Ji, 2003; Liao and Xie, 2004). Organic acids such as acetic acid, tartaric acid, citric acid and oxalic acid are widely spread in rhizosphere soil that are originated from the decomposition of plants and animal residues, microbial secretions, canopy drip and root exudates (Huang et al., 2003; Hu et al., 2007). Some studies have reported that the organic acids strongly altered the bioavailability of heavy metals in contaminated soils (Wasay et al., 2001; Lim et al., 2004). Lumsdon and Farmer (2005) stated that the complexation of metals by organic compounds can accelerate heavy metal dissolution. Wang et al. (2009) also indicated that LMWOAs played an important role for mobilization of heavy metals and increased the total amount of dissolved cations in soil. Organic acids may influence Cr solubility and uptake through chelation, precipitation, and oxidation–reduction reactions (Han et al., 2004; Banks et al., 2006; Afshan et al., 2015).

The bioavailability and leaching of metals can be minimized through chemical and biological amendments. Recently there has been increasing interest in the immobilization of metals using nanoparticles (NPs). Wang et al. (2014) demonstrated that nano-scale zero-valent iron can be used as an effective agent for remediation of Cr contaminated soils. Zhang et al. (2012) also reported that the mixture of bentonite and zero-valent iron enhanced Cr (VI) removal.

Despite the extensive investigations on influence of organic acids and adsorbents on the solubility of Cr in soil, there is still little information in the literature with respect to interactions among Cr, organic acids, and sorbent in soils contaminated with leather factory wastes (LFW). On the other hand, there is no information about the effect of organic acids and NPs on the kinetic release of Cr from leather tanning waste. The objectives of this study were to illustrate the kinetics release of Cr in LFW and soil contaminated with LFW in the presence of organic acids, clay minerals and NPs and their impacts on Cr fractionation. The results of this study can be used to predict the fate and transport of Cr in the soil and LFW to find a useful technique for remediation of contaminated soils.

2. Materials and methods

2.1. Soil, leather factory waste and adsorbents

The soil sample used in this study was collected from an agricultural area and the waste sample was collected from leather factory (LFW) in Hamadan, Iran. The samples were air dried and ground to pass through a 2-mm sieve and stored for the laboratory experiments. Characteristics of the soil, LFW, were previously reported by Taghipour and Jalali (2015) and characteristics of NPs (MgO, ZnO and TiO₂) and clay minerals (bentonite and zeolite) were reported by Mahdavi et al. (2012, 2013) and Moharami and Jalali (2013), respectively.

2.2. Release of Cr

Two organic acids (citric and oxalic acid) and CaCl₂ were selected to study Cr release. We selected citric and oxalic acids, existing commonly in soils, to investigate release of Cr from contaminated soils. These organic acids are commonly present ranging from 0 to 10.0 mM in the rhizosphere soils (Jones, 1998) and they have also different pK_a, ligand forms, and numbers of functional groups (Serjeant and Dempsey, 1979). CaCl₂ was often used as soil background electrolytes in desorption study (Robbins et al., 1999; Qin et al., 2004). In this study it was chosen as contrastive extractor. The experiments were carried out using 13 treatments: LFW, control soil, 10% of LFW plus soil (L-10), 5% of two clay minerals, bentonite and zeolite, (L-10-bentonite, L-10-zeolite,

LFW-bentonite and LFW-zeolite) and 1% of three NPs, MgO, TiO₂ and ZnO, (L-10-MgO, L-10-TiO₂, L-10-ZnO, LFW-MgO, LFW-TiO₂ and LFW-ZnO). All treatments (2 g) were extracted with 20 mL of a 10 mM citric acid, oxalic acid and CaCl₂ solutions. The suspensions were shaken for 30 min at 200 rpm, equilibrated for 0.5, 1, 4, 15, 30, 45, 57, 76, and 90 h at 25 °C, centrifuged at 5000 rpm for 10 min. Concentration of Cr was analyzed in each extract by atomic absorption spectrophotometer. All experiments were performed in two replicates.

Two equations, namely pseudo-first-order and pseudo-second-order (Chen and Li, 2010) kinetic were used to investigate the release of Cr with time. The pseudo-first-order (Eq. (1)) and pseudo-second-order (Eq. (2)) equations are expressed in the following forms:

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (1)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (2)$$

where q_t (mg kg⁻¹) is the amount of Cr released at time t (h), q_e (mg kg⁻¹) is cumulative Cr released at time t , k_1 (h⁻¹) and k_2 (mg kg⁻¹ h⁻¹) are the rate constants of pseudo-first-order and pseudo-second-order models, respectively. The non-linear Chi-square (χ^2) also was used for evaluation of the best fit among the kinetic equations (Boparai et al., 2011) and could be calculated according to the following equation:

$$\chi^2 = \sum \frac{(q_{e, \text{exp.}} - q_{e, \text{cal.}})^2}{q_{e, \text{cal.}}} \quad (3)$$

where $q_{e, \text{exp}}$ and $q_{e, \text{cal}}$ are the measured and fitted Cr release to the experimental data, respectively. If data from the model were similar to the experimental data, χ^2 would be a small value.

2.3. Fractionation of Cr

Chromium fractionation was carried out before and after release of Cr from control and treated soil. Different fractions of Cr were determined by Sposito et al. (1982) method. Similar experiments were also conducted on the LFW and treated LFW.

3. Results and discussion

3.1. Release of Cr from LFW and LFW treated soil in the presence of organic acids

Chromium release from LFW by successive extraction with 0.01 M citric acid, oxalic acid and CaCl₂ were shown in Fig. 1 and Table 1. The release was initially rapid followed by a slower reaction. The rapid release continued for 4 h or 15 h, followed with slower release. It is also noted from Table 1, that there were significant differences ($P < 0.05$) among release of Cr by different extractors. Citric acid had the greatest release of Cr, followed by oxalic acid; and the smallest release was resulted by CaCl₂. For instance, after 90 h, CaCl₂, oxalic acid and citric acid extracted 695.7, 807.2 and 1025.8 mg kg⁻¹ Cr from LFW, respectively.

Chromium release from LFW treated soil by extraction with CaCl₂ and citric and oxalic acids is shown in Fig. 2 and Table 2. Similar with the trend observed in LFW, the Cr from LFW treated soil released in two steps: an initial rapid step (15 h), followed by a slow step. This two phases release has also been reported in other studies for other heavy metals in soils and sediments

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