

Heavy Metal Release from Some Industrial Wastes: Influence of Organic and Inorganic Acids, Clay Minerals, and Nanoparticles



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(Received December 28, 2017; revised January 28, 2018)

ABSTRACT

Knowledge about heavy metal release from industrial solid wastes (ISWs) is crucial for better management of their environmental risks. This study was conducted to investigate the effect of organic and inorganic acids, clay minerals, and nanoparticles (NPs) on the release of heavy metals from sugar factory waste, ceramic factory waste, leather factory waste, and stone cutting waste. The influence of the extractants on heavy metal release from these ISWs was in the following descending order: citric acid > oxalic acid > nitric acid \geq sulfuric acid > CaCl₂. Addition of clay minerals and NPs as adsorbents decreased heavy metal release, which was significantly lower in NP-treated wastes than in the clay mineral-treated wastes. On the other hand, the presence of organic and inorganic acids increased heavy metal adsorption by NPs and clay minerals. These results suggest that NPs can be applied successfully in waste remediation, and organic and inorganic acids play an important role in the removal of heavy metals from the studied adsorbents.

Key Words: adsorbents, environmental risks, extractants, industrial solid wastes, metal removal, remediation

Citation: Taghipour M, Jalali M. 2018. Heavy metal release from some industrial wastes: Influence of organic and inorganic acids, clay minerals, and nanoparticles. *Pedosphere*. 28(1): 70–83.

INTRODUCTION

Rapid urbanization and subsequent industrialization have increased environmental pollution levels. The number of contaminated sites in industrial areas is increasing in developing countries (LaGrega *et al.*, 2001). Environmental challenges for different industries are associated with liquids, gaseous, and solid wastes (Meawad *et al.*, 2010). Industrial wastes in solid form have been recognized as one of the major polluting materials, and their management is recognized as an important problem of modern society. Unfortunately, industrial solid wastes (ISWs) have been dumped into the land. One of the main problems with ISWs is that they contain high concentrations of heavy metals. Appropriate management should be applied to avoid contamination of soil, water, and plants (Amir *et al.*, 2005).

Due to environmental risks of ISWs, especially the pollution of soil and water by heavy metals and their adverse effects on both plant and human health, it is necessary to take action towards remediation of these contaminations. Nanoparticles (NPs) can be used as adsorbents for metal contaminant remediation owing to their special properties, such as high adsorption ca-

pacities, greater activities, and unsaturated surfaces (Taghipour and Jalali, 2015). Many experiments have been carried out in order to study the effectiveness of NPs in heavy metal removal from soil, water, and industrial wastes (Liu and Zhao, 2007; Engates and Shipley, 2011; Mahdavi *et al.*, 2013). The use of natural clay minerals is another way to remove heavy metals from water and soil. Many studies have been conducted using different clay minerals (Inglezakis *et al.*, 2016; Stylianou *et al.*, 2016). For good management of industrial wastes, it is necessary to obtain exact information and data about the waste characteristics. In the past, characteristics of industrial wastes as well as their impacts on soil pollution, plants, and animals have been studied extensively (Mishra *et al.*, 2009; Kumar and Chopra, 2010). However, few studies have investigated industrial wastes in solid form, and most of these studies essentially focused on quantification and characterization of the wastes (Mbuligwe and Kaseva, 2006; Chang *et al.* 2010). So far, less attention has been paid to the release of heavy metals from industrial wastes. In addition, there is no previous report on the effect of NPs (especially ZnO and MgO) on the release of heavy metals from industrial wastes in solid form.

Therefore, the determination of heavy metals in

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ISWs and the development of a strategy to minimize heavy metals in wastes are important challenges. This study was designed to ascertain the levels of lead (Pb), cadmium (Cd), chromium (Cr), nickel (Ni), zinc (Zn), and copper (Cu) in four types of ISWs, sugar factory waste (SFW), ceramic factory waste (CFW), leather factory waste (LFW), and stone cutting waste (SCW) in the presence of organic and inorganic acids and to study the effect of NPs and clay minerals on heavy metal release from these wastes.

MATERIALS AND METHODS

Industrial solid wastes and adsorbents

The four types of ISW samples were collected from four factories in Hamadan Province, Iran. In general, SFW, CFW, LFW, and SCW were collected from the mixture of all solid wastes produced at the sugar factory, broken fractions of pottery and ceramic, last stage of leather production (tanning), and raw materials used during cutting and production processes in sedimentation basins, respectively. All waste samples were passed through a 2-mm sieve and stored for further analysis. Samples were digested in nitric acid (Spósito *et al.*, 1982), and the total heavy metal concentrations of the digests were measured using an atomic absorption spectrometer (AAS, Varian SpectrAA 220 FS, Varian, Palo Alto, USA) at the wavelengths of 228.8, 324.8, 357.9, 232.0, 217.0, and 213.9 nm, respectively. The detection limits of the AAS method were 0.02, 0.03, 0.06, 0.10, 0.10, and 0.01 mg L⁻¹ for Cd, Cu, Cr, Ni, Pb, and Zn, respectively. In this study, NPs (MgO, TiO₂, and ZnO) and clay minerals (bentonite and zeolite) were used to remediate ISWs. Industrial solid wastes without remediation were used as controls. Characteristics of the NPs and clay minerals were previously reported by Mahdavi *et al.* (2012, 2013) and Moharami and Jalali (2013).

Release of heavy metals from ISWs

Kinetics of heavy metal release were studied *via* successive extraction using extractants including two organic acids (10 mmol L⁻¹ citric acid and 10 mmol L⁻¹ oxalic acid), two inorganic acids (10 mmol L⁻¹ nitric acid and 10 mmol L⁻¹ sulfuric acid), and 10 mmol L⁻¹ CaCl₂. Citric and oxalic acids have different dissociation constants, ligand forms, and numbers of functional groups. Different organic acids have various influences on heavy metal release from ISWs, so these two organic acids were chosen as representatives of organic acids in the plant rhizosphere. Nitric and

sulfuric acids are produced from the decomposition of microbial metabolites in soils. In addition, 10 mmol L⁻¹ CaCl₂ has an ionic strength similar to soil solutions and thus was chosen as a contrastive extractant.

Industrial solid wastes were mixed with 1% of each NP and 5% of each clay mineral, separately. To determine heavy metal release from all treatments, 2 g of each sample was placed into a 50-mL centrifuge tube and mixed with 20 mL each of CaCl₂ and nitric, sulfuric, citric, and oxalic acids. All samples were equilibrated at 25 °C for intervals ranging from 0.5 to 90 h (0.5, 1, 4, 15, 30, 45, 57, 76, and 90 h). The suspensions were shaken for 0.5 h followed by centrifugation, and the solution was analyzed. Then, fresh solution was added to the solid residues and the contact process was repeated. The concentrations of Cd, Cu, Cr, Ni, Pb, and Zn in the supernatant were analyzed using an AAS in the same way as the total heavy metal concentrations. All extractions were run in duplicate.

Release kinetics of heavy metals from ISWs

The release of heavy metals from ISWs was described with pseudo-first-order (Eq. 1) or pseudo-second-order (Eq. 2) kinetic equations (Shirvani *et al.*, 2007; Chen and Li, 2010):

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

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

where q_t and q_e (mg kg⁻¹) are the amounts of the heavy metal released at time t (h) and cumulative heavy metal released at time t , respectively and k_1 (h⁻¹) and k_2 (kg mg⁻¹ h⁻¹) are the rate constants of the pseudo-first-order and pseudo-second-order equations, respectively.

Physiologically based extraction test

The bioaccessibilities of heavy metals in the ISWs were measured following a physiologically based extraction test (PBET) method (Kelley *et al.*, 2002). The PBET extraction solution was prepared using a 30 g L⁻¹ glycine (0.4 mol L⁻¹) solution adjusted to a pH of 2.3 using HNO₃ solution. This solution can be simulated as that of the human gastric system. In PBET, 0.1 g of each ISW was mixed with 10 mL of the extraction solution. Then, the samples were placed in an incubator at 37 ± 2 °C. After 1 h, the samples were centrifuged at 3000 r min⁻¹ for 10 min, and then analyzed for heavy metals using an AAS.

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