Environmental Pollution 243 (2018) 929–939

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Graphite particle electrodes that enhance the detoxification of municipal solid waste incineration fly ashes in a three-dimensional electrokinetic platform and its mechanisms☆

Tao Huang [a, *], Shuwen Zhang [b, **], Longfei Liu [a], Jiaojiao Xu [a]

[a] School of Chemistry and Materials Engineering, Changshu Institute of Technology, 215500, China
[b] Nuclear Resources Engineering College, University of South China, 421001, China

ARTICLE INFO

Article history:
Received 23 July 2018
Received in revised form
31 August 2018
Accepted 31 August 2018
Available online 17 September 2018

Keywords:
Particle electrodes
A three-dimensional electrokinetic process
Municipal solid waste incineration fly ashes
Heavy metal removal
Enhancement

ABSTRACT

This paper investigated the application of graphite particle electrodes to the removal of Zn, Pb, Cu, and Cd from municipal solid waste incineration (MSWI) fly ashes in a three-dimensional (3D) electrokinetic reactor. The influences of the voltage gradient, mass ratio of graphite powers to fly ashes, nitric acid concentrations, proposing times, and liquid-solid (L-M) ratios on the remedial efficiencies of MSWI fly ashes were comprehensively studied in an orthogonal deign and a sequential double-factor setup. Significant analysis showed that changes in the mass ratios and nitric acid concentrations both had a statistically significant effect on the removals of Zn and Pb. Proposing times and L-M ratios both remarkably affected the removals of heavy metals (HMs) in a 3D electrochemical system. The graphite powers had a narrower distribution interval and slightly larger surface areas compared with MSWI fly ashes, which relented pH gradients over the time in the electrochemical experiments and minimized the bubble barricade caused by the hydrolysis. The particle electrode had increased the residue factions of Zn, Pb, Cu, and Cd in S1 region by approximately 216%, 136%, 309%, and 950%, respectively, compared with the raw MSWI fly ashes. The addition of graphite powders to a two-dimensional (2D) electrochemical process strengthened hydrolysis reactions, shortened time for the redistribution of pH balance, decreased the tortuosity of migration path, and increased the desorption concentrations of HMs in the sample area.

1. Introduction

Approximately 168.161 million tons of municipal solid wastes (MSW) generated in 244 large and medium-sized cities in China were needed to be appropriately treated in 2014(Huang et al., 2018b; Yu, 2018). Incineration has been adopted in priority for MSW treatment in more cities among the alternative techniques due to its own technical advantages including heat recovery and volume reduction. Correspondingly, a large amount of MSW incineration (MSWI) fly ash was generated for MSW disposal. MSWI fly ash is classified as a hazardous waste attributed to its high content of toxic substances, such as dioxins and heavy metals (HMs)(Huang et al., 2018a; Lederer et al., 2017). Free disposal of MSWI fly ashes has now attracted more environmental concerns in the local communities. Therefore, the detoxification of MSWI fly ashes mainly referring to the removal of HMs and the reduction to some dioxins is essential before the further treatment or utilization of MSWI fly ashes. Currently, the methods used to remediate MSWI fly ashes are typically divided into three categories, including chemical and electrochemical separation, thermal treatment (e.g., sintering and vitrification), and immobilization (e.g., solidification/stabilization) (Cioffi et al., 2009; De Boom and Degrez, 2015; Fedje et al., 2012; Kozakova et al., 2013; Meffre et al., 2015). However, the problems of low removal efficiency of HMs, the high consumption of chemical reagents, and secondary pollution persistently baffle the large-scale applications of these techniques for the harmless treatment and resource utilization of MSWI fly ashes(Huang et al., 2015).

Electrokinetic remediation (EKR), considered as a suitably practical method for the treatment of solid matrices with low permeability, has received attention and been widely conducted for the removal of several contaminants *in situ* and *ex situ*(Chowdhury

☆ This paper has been recommended for acceptance by Dr. Jorg Rinklebe.
* Corresponding author.
** Corresponding author.
E-mail addresses: ht1104705720@qq.com (T. Huang), tillg14536@yahoo.com (S. Zhang).

https://doi.org/10.1016/j.envpol.2018.08.101
0269-7491/

et al., 2017; Ferrucci et al., 2017; Vizcaino et al., 2018). An electric field is employed to the contaminated sample or media via a pair of embedded electrodes, which induces the reaction of water electrolysis and the mobilization of some dissolved substances including ionic species, interstitial fluid, and solubilized colloids through the porous matrices towards the opposite electrode (i.e., electromigration, electroosmosis, and electrophoresis)(Li et al., 2016). EKR technique can overcome the issue of secondary pollution more effectively and is easier to be implemented compared with other remediation techniques(Maturi and Reddy, 2008; Ricart et al., 2008; Rozas and Castellote, 2012). However, the elevation of HM removal via EKR process for the harmless treatment of solid wastes was restrained and affected by some factors such as the physicochemical characteristics of the media, changes of pH and zeta potential, mass transfer, etc. Generally, the soluble ions or complexes of HMs in the interstitial fluid can only be electromoved and concentrated in the specific area in the media(Jensen et al., 1994; Shiba and Hirata, 1998). The proton attacking caused by water hydrolysis and pH control in the electrode chambers for the matrices to facilitate the release of HM ions play a key role in determining HM mobility during EKR process(Kawachi and Kubo, 1999; Reddy and Chinthamreddy, 1999; Virkutyte et al., 2002).

To effectively cope with the high-buffering-capacity MSWI fly ashes with a smaller amount of acid added to the electrolyser, hydrolysis reaction happened on the anode must be intensified to generate more protons to decrease pH in the media. In recent years, a three-dimensional (3D) electrochemical process has been developed based on a two-dimensional (2D) platform, which also has been applied by many researchers to improve remedial efficiencies in the solid waste and wastewater treatments(Chmayssem et al., 2017; Huang et al., 2018a; Zhang et al., 2013). Some particles including granular activated carbon, graphite, carbon aerogel, metal, and modified kaolin are chosen to be packed between 2D electrodes to solve some intrinsic drawbacks caused by a 2D electrochemical process, such as mass transfer limitation and low area-volume ratio. These particles also named as bed electrodes or particle electrodes can be easily polarized by the external voltage to form the charged microelectrodes with one surface being charged as anode and the other surface being identified as the cathode in the sample region(Britto-Costa et al., 2014; Rosestolato et al., 2015; Yeung and Gu, 2011).

In this paper, graphite powers were selected as particle electrodes and incorporated into a traditional two-dimensional device. The application of an assembled 3D EKR platform to MSWI fly ashes was thoroughly investigated. Specifically, the influences of some factors including the voltage gradient (V/cm), mass ratio of particle electrodes to MSWI fly ashes (wt.%), nitric acid concentrations (mol/L), proposing times (d), and ratios of liquid to solid samples (mL/g) on the removal efficiencies of HMs from MSWI fly ash samples were comprehensively explored and tested. An orthogonal experimental design and a double-factor setup were sequentially used to achieve a reasonable arrangement for experimental factors. Analysis of variance (ANOVA) was conducted to obtain an optimal combination of factors. An appropriate neutral net model was chosen to predict fitting results in the set scope of parameters and to optimize the 3D EKR process for the remediation of MSWI fly ashes based on the training and simulating data coming from the orthogonal and double-factor experiments. The environmental toxicity and activity of MSWI fly ashes before and after the 3D treatments were comparatively characterized. The techniques of XRD, XRF, ICP-OES, SEM, and BET were applied to testify the suitability of some speculated theories in a 3D electrochemical process for wastewater treatment and to analyze the mechanisms of enhancing removal of inorganic contaminants from the solid wastes. This study not only explores the feasibility of graphite powders adopted as a particle electrode in a 3D electrochemical process also supplies an enhanced EKR method for practitioners to detoxify MSWI fly ashes more efficiently.

2. Materials and methods

2.1. MSWI fly ash and graphite powders

MSWI fly ash samples were captured and collected in the bag filters residing in a waste incineration power plant, Chongqing, China. The samples were dried in a thermostatic heater at 60 °C for 2 h (h) and sifted by a 200-mesh sieve. Graphite particles were purchased from Shanghai Xili Carbon Co., Ltd., China. The graphite powders used to the experiments were prepared by mechanically milling the graphite particles for 1 h at 150 rpm. The elementary composition of MSWI fly ashes was measured using X-ray fluorescence (XRF, 1800CCDE). The size distributions and the structure characteristics of both fly ash and graphite power samples were detected and analysed by a laser particle size analyzer (Microtrac, S3500) and a BET surface-area analyzer (Micromeritics, USA), respectively.

2.2. Experimental design

The electrokinetic experiments were conducted in the glass cells, which majorly consisted of a sample chamber and two electrode compartments. The chamber was evenly divided into three rectangular regions from the anode to cathode labeled as S1, S2, and S3, respectively. The other parameters on the experimental setup have been detailed in my previous work(Huang et al., 2015; Huang et al., 2018b; Liu et al., 2016). The particle electrodes were mixed with MSWI fly ashes in the sample chamber. The schematic diagram of a 3D EKR electrolyser used in this study has been specified in another work(Huang et al., 2018a). To investigate the effects of factors including the voltage gradients (V/cm) loaded over the sample chamber, mass ratio of graphite powers to fly ashes (wt.%), concentrations of nitric acid (mol/L) used for the pretreatment of mixtures of fly ashes and graphite powders (FA-GP) on the removal of HMs, an orthogonal layout was adopted and designed on the premise of fixing two variables (i.e., the proposing time of 7 d and L-M ratio of 1:1 ml/g). The levels of each factor, arrangements of electrochemical experiments, and removal rates of Zn, Pb, Cu, and Cd are specified in Table 1. Furthermore, the proposing times (d) of an EKR process and L-M ratios (mL/g) were changed in the form of double-factor setup based on the analytical results in the orthogonal tests to and were analysed using ANOVA to further optimize parameters for the remediation of contaminants considering the energy consumption and running cost. The double-factor experiments and removal rates of Zn, Pb, Cu, and Cd are shown in Table 2. Finally, a feed-forward back propping neural network was chosen and applied to train data acquired from the above tests and to predict the optimal combination of five factors for the 3D electrokinetic performance.

2.3. Analytical methods and calculations

The leaching toxicities of HMs were carried out according to a standardized procedure of TCLP in triplicate(Komilis et al., 2013; Tsang et al., 2013). The chemical speciation of HMs was analysed following a modified BCR procedure in triplicate, in which HMs are divided into four partitions including acid-soluble fraction (F1), reducible fraction (F2), oxidizable fraction (F3), and residue (F4)(Ahmadipour et al., 2014; Aydin et al., 2013; Huang et al., 2018b). The total concentration of HMs in the solid samples was determined by microwave digestion and concentration detection.