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Investigation of the leaching behavior of lead in stabilized/solidified waste using a two-year semi-dynamic leaching test

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HIGHLIGHTS

- Leaching mechanisms during different periods turned out to be diffusion.
- The effective diffusivity of Pb has time-dependence.
- Successive leaching behavior were investigated under different occasions.
- S/S materials keep stable in weak acid or weak alkaline environment for two years.

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ABSTRACT

Long-term leaching behavior of contaminant from stabilization/solidification (S/S) treated waste stays unclear. For the purpose of studying long-term leaching behavior and leaching mechanism of lead from cement stabilized soil under different pH environment, semi-dynamic leaching test was extended to two years to investigate leaching behaviors of S/S treated lead contaminated soil. Effectiveness of S/S treatment in different scenarios was evaluated by leachability index (LX) and effective diffusion coefficient (D_e). In addition, the long-term leaching mechanism was investigated at different leaching periods. Results showed that no significant difference was observed among the values of the cumulative release of Pb, D_e and LX in weakly alkaline and weakly acidic environment (pH value varied from 5.00 to 10.00), and all the controlling leaching mechanisms of the samples immersed in weakly alkaline and weakly acidic environments turned out to be diffusion. Strong acid environment would significantly affect the leaching behavior and leaching mechanism of lead from S/S monolith. The two-year variation of D_e appeared to be time dependent, and D_e values increased after the 210th day in weakly alkaline and weakly acidic environment.

1. Introduction

Stabilization/solidification (S/S) technology is the best available technology for 57 regulated hazardous wastes and the most commonly applied technology in the past few years (US EPA, 2004; Shi and Spence, 2004). However, the fatal flaw of this technology is the hazardous waste remaining in the S/S monolith, and the future environmental risk caused by waste leaching is difficult to evaluate. The question asked in conjunction with S/S treatment is not so much “if the environment causes contaminants to be released from a stabilized material” but “in what form and what speed” does this occur (Hinsenveld, 1992). Furthermore, large amount of S/S treated waste that was dumped into sanitary landfills is a waste of resources, and recycling of S/S materials may become an important method for the disposal of S/S waste. The bottleneck problem for recycling of S/S materials involves quantifying long-term leaching behavior of hazardous waste and predicting the long-term environment risk.

The leachability of the S/S treated waste has always been evaluated by the toxicity characteristic leaching procedure (TCLP) (US EPA, 1992) and semi-dynamic leaching tests (ANS 16.1, 1986; NEN 7345, 1993). The TCLP can only provide a single leaching result at

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a specific time point after S/S treatment, which is usually conducted after short-term curing. Previous studies have failed to investigate the continuous leaching behaviors of waste from a S/S monolith. Although a few studies have assessed the leachability of S/S waste using TCLP after 5, 16 and 17 years of S/S treatment (Antemir et al., 2010; Al-Tabbaa and Boes, 2002; Wang et al., 2014), leaching result were only obtained from a single time point, and TCLP has been previously criticized (Kosson et al., 2002). Semi-dynamic leaching tests have been employed to provide much more information regarding the successive leaching behaviors of S/S waste and the leaching mechanism of waste. Unlike the flow-through dynamic leaching test, the semi-dynamic leaching test is a flow-around test where the leachate is replaced periodically to simulate leaching behavior of a S/S monolith with low permeability. However, most current studies using semi-dynamic leaching tests last less than 90 days (Guo et al., 2013; Moon and Dermatas, 2007; Song et al., 2013; Dermatas, 2004). A few studies have been correlated to long-term investigation of S/S treatments. Previous study of Wang et al. (2016) investigated leaching behaviors of lead under different pH conditions for 90 days. Liu et al. (2013) conducted a leaching test that lasted for 123 days to assess the leaching behaviors of phenol under different simulated situations. The Russian national analogue of the ISO testing standard (ISO, 1982) was performed by Ojovan et al. (2011) to evaluate the leaching rate of ^{137}Cs. However, the effectiveness and leaching mechanism were not evaluated. Jin et al. (2016) validated the 3-year effectiveness of S/S treated highly contaminant land with MgO based binders, which involved an evaluation at a specific time point after the S/S treatment. In general, all of the previously mentioned studies have not provided effective insight into the leaching behaviors and leaching mechanism of S/S waste during the entire process over a long period of time. As noted in previous studies, developing and evolving of crack in the S/S monolith would greatly change the leaching behaviors of waste components (Pabalan et al., 2009; Drace et al., 2012) and material integration does not last forever, which would affect the long-term leaching behaviors of waste. Besides, chemical reactions, such as sulphate erosions, carbonation and chloride attacks, would also affect long-term leaching behaviors of cementitious materials (Drace et al., 2012). Although problems with respect to long-term effectiveness of S/S treatment have been previously mentioned (Conner, 1990; Borns, 1997; Glasser, 1997; Loxham et al., 1997), satisfactory interpretations and direct evidence of the long-term performance of waste leaching from a S/S monolith both in experiment and in the field are lacking.

Once there is a proper understanding of the controlling leaching mechanism of contaminant release from S/S waste, the leaching behaviors can be predicted using a geochemical and transport model (Hinsenveld, 1992). Numerous calculations have been performed to predict the long-term leaching behaviors of heavy metals, and the diffusion model is the most popular model (Crack, 1975). Most existing diffusion theories usually model long-term leaching behaviors of waste through some parameters calculated from short-term tests (Godbee et al., 1980; Batchelor, 1990). A partial fixation model (i.e., diffusion model in conjunction with absorption Côté, 1986) or dissolution (Côté et al., 1987) has been established to predict the long-term metals release from a S/S monolith under different environment conditions. In general, the previously mentioned model may be effective and fit the short-term leaching data well but the accuracy of these models for long-term predictions remains unclear.

In particular, the effective diffusion coefficient in previously mentioned models is always taken as constant, but the effective diffusion coefficient would change when the S/S materials degraded and micro-cracks appeared in the S/S monolith. Typically, De has been considered time dependent. However, a diffusion model with a time-dependent D_e has not been previously reported (Huang et al., 2003).

Furthermore, acid rain is a common phenomenon in many countries and can be quite severe in some cities, such as Nan Jing, where the rain pH has been as low as 2.89 (Nanjing EPA, 2012). Moreover, once the S/S waste is recycled in the environment, different types of liquid will surround it including acid rain with different pH values or alkaline leachates. Therefore, deeper insight into the long-term leaching behaviors under different pH conditions is required.

Therefore, to obtain a better understanding of the long-term leaching behaviors, leaching mechanism and effectiveness of S/S waste, lead was chosen as the target contaminant in the S/S monolith. Semi-dynamic leaching test was extended to two years to investigate the difference in the long-term leaching behaviors of cement stabilized/solidified Pb contaminated soil in different environment. The leaching mechanisms were validated by two-year leaching data. In particular, a time-dependent D_e was evaluated to gain insight into the variation in the constant D_e used in diffusion models.

2. Materials and methods

The materials used in this study as well as the preparation of an artificially contaminated soil and S/S specimen have been documented in a previous study (Wang et al., 2016; Li et al., 2014, 2015). Except for the extended semi-dynamic leaching test, measurement of leachate pH and lead concentration were performed according to the protocols in the same previous study.

2.1. Materials

2.1.1. Preparation of artificially contaminated soil

Most of the materials used in this paper and the preparation are similar to the previous publication (Wang et al., 2016). Due to its easy accessibility and relatively low cost, silty clay prepared from a subway construction site in Wuhan was used to prepare the simulated contaminated soil. The basic properties of soil are presented in Table 1, and these properties were obtained through the "Standard for soil test method" in China. The light compaction experiment was used to acquire the maximum dry density and optimum moisture content. Portland cement was used to immobilize the lead contaminated soil due to its wider application in S/S technology and its low cost and effectiveness (Spence and Shi, 2004).

A 2 mm sieve was used to eliminate the large soil particles, and the fine screening soil were collected for experiment. A certain amount of the dry soil was weighed, and the amount of $Pb(NO_3)_2$ required to ensure that the lead concentration in this artificial contaminated soil kept 5000 mg/kg was calculated before the preparation of lead contaminated soil. The reason for choosing $Pb(NO_3)_2$ was because nitrate ions would not affect cement hydration (Cuisinier et al., 2011). Then, the amount of deionized water required ensuring that the moisture of the weighed contaminated soil kept at optimum moisture content 19.5%, was calculated. Lastly, the calculated $Pb(NO_3)_2$ and water was mixed in a container to yield a solution containing $Pb(NO_3)_2$, and the solution was evenly added to the prepared soil. The Pb-contaminated soil was evenly mixed and cured for 10 days to ensure that the reaction between $Pb(NO_3)_2$ and the clay reached equilibrium.

2.1.2. Sample preparation

The cement was bought from the China Huaxin Cement Co., Ltd. And the cement type was PO 42.5 N silicate cement, whose strength class was 42.5 and ordinary early strength indicated by N. More