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An evaluation of lead leachability from stabilized/solidified soils under modified semi-dynamic leaching conditions

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Abstract

Semi-dynamic leaching tests were conducted for artificial soils contaminated with lead oxide (PbO) in order to assess the long-term leaching behavior of lead (Pb). In order to simulate "worst case" leaching conditions, the semi-dynamic leaching test was modified using 0.014 N acetic acid (pH = 3.25) instead of distilled water. Lead contaminated artificial soils were prepared by mixing kaolinite or montmorillonite with fine quartz sand. The contaminated soil was then subjected to stabilization/solidification (S/S) treatment using quicklime, fly ash, or quicklime–fly ash combination. Fly ash was added in order to provide sources of aluminum, calcium, or silicate for the formation of precipitates and pozzolanic reaction products. The effectiveness of S/S treatment was evaluated by determining diffusion coefficients (D_e) and leachability indices (LX). A model developed by de Groot and van der Sloot [de Groot, G.J., van der Sloot, H.A., 1992. Determination of leaching characteristics of waste materials leading to environmental product certification. Stabilization and Solidification of Hazardous, Radioactive, and Mixed Wastes, 2, STP 1123, T.M. Gilliam and C.C. Wiles, eds., ASTM, Philadelphia, 149–170.] was used in order to elucidate the controlling leaching mechanisms. Slurry tests were also performed by mixing PbO with quicklime and fly ash, in order to study the immobilization mechanisms of Pb. The resulting reaction products were identified using X-ray diffraction (XRD) analyses. Overall, the test results indicated that S/S treatment was effective in immobilizing Pb and the treated soils can be considered acceptable for "controlled utilization" based on LX values. The controlling leaching mechanism was found to be diffusion, in most quicklime-treated samples. Precipitation was identified as the most likely Pb immobilization mechanism in quicklime–fly ash-treated slurries. Lead silicate, Pb_2SiO_4 (which is highly insoluble) is the most probable precipitate that can be associated with the decrease in Pb leachability. No evidence of pozzolanic reaction products such as calcium silicate hydrates (CSH) and calcium aluminate hydrates (CAH) was identified by XRD.

Keywords: Semi-dynamic leaching test; Stabilization/solidification (S/S); Quicklime; Lead (Pb); Leachability index (LX); Precipitation

1. Introduction

Lead (Pb) has been identified as one of the most toxic elements to human health and is a widespread contaminant in many hazardous wastes sites (Lin et al., 1996). It has been reported that Pb can cause damage to the brain, red blood cells, blood vessels, kidneys and the nervous system (Lin et al., 1996; Long and Zhang, 1998). Human exposure to Pb has intensified due to industrialization. The transportation industry, which contributes leaded gasoline and Pb storage batteries, the paint industry with leaded paints, the defense industry using Pb in various

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ammunitions, the plumbing and electronics industries with leaded solder and the food industry with Pb-contaminated cans are all adding Pb to the environment. Due to the widespread use of Pb over a period of many years, the Pb loading rate in soil is approximately 20 times (or more) its natural removal rate (Nriagu, 1990). Therefore, the contamination risk to groundwater due to leaching of Pb from landfill areas and industrially contaminated land has received increasing attention (Gee et al., 1997).

In this study, a stabilization/solidification (S/S) process was used in order to remediate Pb contamination, because S/S techniques have been widely applied in order to treat soils with heavy metal contamination (Conner, 1990; Yukselen and Alpaslan, 2001). During S/S treatment, the hazardous waste potential of waste materials can be minimized by converting the contaminant into forms which are less-soluble, less mobile or less toxic and encapsulating the waste within a monolithic solid of high structural integrity (Conner, 1990). Various combinations of stabilizing agents have been used by numerous researchers in the treatment of soils contaminated with Pb. Jing et al. (2004) studied Pb leachability in cement, lime and fly ash stabilized/solidified soil samples. Dermatas and Meng (2003) used quicklime and fly ash in order to evaluate the degree of Pb immobilization using the Toxicity Characteristic Leaching Procedure (TCLP). Li et al. (2001) studied Pb immobilization using Portland cement (OPC) and pulverized fly ash (PFA). Long and Zhang (1998) used cement in combination with various additives such as lime, fly ash, clay, apatite and silicate for treating Pb contaminated soils. Wang and Vipulanandan (1996) used Type 1 Portland cement and class C fly ash as a stabilizing agent to evaluate Pb leachability.

In this study, quicklime was used as the main stabilizing agent rather than cement and hydrated lime because: a) of its economic advantage over cement and hydrated lime; b) its heat of hydration results in an accelerated rate of reaction; and c) there is limited information available to date regarding quicklime-based S/S.

Upon quicklime treatment of Pb-contaminated soils, there are three possible Pb immobilization mechanisms: precipitation, inclusion and sorption.

Fly ash was used as stabilizing agent alone or with quicklime in order to evaluate its effectiveness on Pb immobilization. Since fly ash contains silicon dioxide (SiO_2), aluminum oxide (Al_2O_3) and calcium oxide (CaO), it provides calcium, aluminum, or silicate sources for the formation of precipitates and pozzolanic reaction products at high pH conditions.

Leaching is known to be a complex phenomenon because many factors may influence the release of specific constituents from a waste over a period of time (van der Sloot et al., 1996). These factors include major element chemistry, pH, redox potential, complexation, liquid-to-solid ratio, contact time, etc. (van der Sloot et al., 1996). Moreover, since very little is known about the chemical species present in waste forms and their behavior with respect to time, the long-term performance of S/S waste forms has been difficult to predict.

In order to predict the long-term leaching behavior, a diffusion model is frequently used to evaluate the leaching kinetics. In this study, a diffusion theory-oriented test, the American Nuclear Society's semi-dynamic leaching test (ANS, 1986), was utilized in order to examine the mechanisms governing Pb leachability in quicklime-based solids.

The objectives of this study are: 1) to assess the effect of clay surface area and mineralogy on Pb leachability; 2) to evaluate the effectiveness of quicklime treatment; 3) to evaluate the importance of fly ash addition in increasing Pb immobilization; 4) to determine the controlling leaching mechanisms (diffusion vs. dissolution) of Pb in treated soils; and 5) to investigate the immobilization mechanisms of Pb by using X-ray diffraction (XRD) analyses.

2. Review of diffusion model

2.1. ANS model, diffusion coefficient (D_e) and leachability index (LX)

The long-term leachability of Pb from quicklime-treated soils was evaluated using the ANS 16.1 model (ANS, 1986). This model was established based on Fick's diffusion theory and standardized by ANS (ANS, 1986). This model can be used to determine the cumulative fraction of Pb leached against time. It has been widely reported that the leaching of the contaminant from cement-based waste forms is mostly a diffusion-controlled process (Godbee and Joy, 1974; Dutré and Vandecasteele, 1996). Due to the slow diffusion rate of contaminants, it can be assumed that a quicklime-based waste form is a semi-infinite medium, much like the cement-based waste forms examined in previous studies (Godbee and Joy, 1974; Côté et al., 1987; Andrés et al., 1995). This implies that the release of the contaminant from the waste form is negligible when compared to the contaminant's total mass. As a result of this implication, diffusion is expected to be the controlling leaching mechanism in soils treated with quicklime.