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Modelling the leaching of Pb, Cd, As, and Cr from cementitious waste using PHREEQC

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Abstract

A leaching model was developed using the United States Geological Survey public domain PHREEQC geochemical package to simulate the leaching of Pb, Cd, As, and Cr from cementitious wastes. The model utilises both kinetic terms and equilibrium thermodynamics of key compounds and provides information on leachate and precipitate speciation. The model was able to predict the leaching of Pb, Cd, As, and Cr from cement in the presence of both simple (0.1 and 0.6 M acetic acid) and complex municipal landfill leachates. Heavy metal complexation by the municipal landfill leachate was accounted for by the introduction of a monoprotic organic species into the model.

The model indicated Pb and As were predominantly incorporated within the calcium silicate hydrate matrix while a greater portion of Cd was seen to exist as discrete particles in the cement pores and Cr (VI) existed mostly as free CrO_4^{2-} ions. Precipitation was found to be the dominant mechanism controlling heavy metal solubility with carbonate and silicate species governing the solubility of Pb and carbonate, silicate and hydroxide species governing the solubility of Cd. In the presence of acetic acid, at low pH values Pb and Cd acetate complexes were predominant whereas, at high pH values, hydroxide species dominated. At high pH values, the concentration of As in the leachate was governed by the solubility of $Ca_3(AsO_4)_2$ with the presence of carbonate alkalinity competing with arsenate for Ca ions. In the presence of municipal landfill leachate, Pb and Cd organic complexes dominated the heavy metal species in solution. The reduction of As and Cr in municipal landfill leachate was crucial for determining aqueous speciation, with typical municipal landfill conditions providing the reduced forms of As and Cr.
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Keywords: Cementitious waste; PHREEQC; Geochemical modelling; Lead; Cadmium; Arsenic; Chromium; Municipal landfill; Leachate

1. Introduction

In recent years there have been increased concerns regarding the leaching of hazardous substances from landfills into local surroundings. As a result, an extensive array of leaching tests has been developed to assess the hazards of heavy metal-containing wastes prior to disposal. Failure to pass a regulatory leaching test typically necessitates some form of waste stabilisation, such as cement addition. Batch leaching tests are the preferred choice for regulatory assessment due to their simplicity, improved reproducibility, and shorter time requirements. However, as batch leaching tests are typically run over short time frames, it is debatable whether the compounds of interest behave similarly in the long term. Modelling can potentially predict the long term leaching of wastes providing a solution to problems inherent to batch procedures. A model capable of describing contaminant leaching from wastes can assist in improving the development of management options [1].

This study focuses on the leaching of Pb, Cd, As, and Cr from cement-stabilised waste. The leaching of heavy metal ions from cementitious waste has been extensively investi-

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gated [2–7], with many researchers using physical mechanistic leaching models [8–11]. Although physical mechanistic models can successfully describe the leaching of contaminants from cement, this approach does not give direct indication of chemical species controlling the release. According to van der Sloot et al. [1], the chemistry of elements and their interaction with substances present in the leaching fluid, including ligands and sorbing phases, are governing factors in element release. Geochemical modelling enables quantitative speciation of elements based on species stability constants. PHREEQC is a geochemical modelling package capable of describing chemical reactions and transport processes in water. It is based on the equilibrium chemistry of aqueous solutions with other components, such as minerals, gases, solid solutions, and sorbing surfaces. A solid solution forms when compounds containing similar properties, such as size and charge, precipitate to form a homogeneous solid compound. It is also capable of incorporating kinetic equations for chemical reactions and determining speciation at any designated time. Many models have been developed to describe the chemistry of natural waters [12] but at present no model of this type has been developed to describe the leaching of elemental contaminants from cement.

This paper presents work aimed at using PHREEQC to develop a model for simulating the leaching of Pb, Cd, As, and Cr from cementitious wastes and, where possible, identify species governing the release of these contaminants. Model outcomes were assessed by comparison with leaching profiles obtained from procedures recommended by AS 4439.3-1997. Leaching fluids studied included 0.1 and 0.6 M acetic acid and a municipal landfill (ML) leachate.

2. Experimental procedure

2.1. Preparation of cementitious waste

Cementitious wastes were prepared by mixing Type GP Ordinary Portland Cement (OPC), supplied by Australian Cement (in accordance with Australian Standard AS 3972-1991), with solutions of lead nitrate ($Pb(NO_3)_2$), cadmium nitrate tetrahydrate ($Cd(NO_3)_2 \cdot 4H_2O$), sodium arsenate ($Na_2HAsO_4 \cdot 7H_2O$), or sodium chromate (Na_2CrO_4) such that they contained 2.34% Pb, 1.3% Cd, 1.3% As, or 2.0% Cr by weight, respectively. The metal salts were dissolved in water, mechanically blended with the OPC and cured for 28 days at water to cement ratios of 0.38 (Pb-spiked cement), 0.44 (Cd-spiked cement), 0.38 (As-spiked cement), and 0.31 (Cr-spiked cement). Detailed procedures are further described in a previous publication [13]. The cured mixtures were crushed by three consecutive crushers (jaw crusher, cone crusher, and roller crusher), and passed through a 2.4-mm mesh sieve. The compositions of the cement samples were analysed using X-ray fluorescence (XRF) and are shown in Table 1.

Table 1
Composition of cementitious wastes (dry weight) containing Pb, Cd, As, and Cr as determined by X-ray fluorescence [49]

Element	Cement composition (mg g^{-1} of waste)			
	Pb cement	Cd cement	As cement	Cr cement
Pb	23	0.0	0.0	0.0
Cd	0.0	13	0.0	0.0
As	0.0	0.0	13	0.0
Cr	0.0	0.0	0.0	20
Al	16	15	16	15
C	15	20	11	9
Ca	340	340	340	340
Fe	23	22	23	23
K	6.6	7.5	6.6	6.6
Mg	10	10	10	10
Na	4.5	2.2	13	22
Si	74	73	74	73
S	8.8	8.4	8.8	8.8
Th	0.9	0.9	0.9	0.9
Ti	0.6	0.6	0.6	0.6

2.2. Leaching experiments

The leaching procedure used was that recommended by Australian Standard AS 4439.3-1997 [14]. AS 4439.3-1997 is a modified version of the toxicity characteristic leaching procedure (TCLP), designed to account for waste disposal scenarios other than and including codisposal in a municipal solid waste landfill [14]. Eighty grams of crushed waste was tumbled with 1600 g of leaching fluid at a speed of 30 rpm. Approximately ten 10-mL samples were taken at nominated time intervals between 0 and 7 days. The zero-hour value corresponds to sample mixed with the leaching fluid for a few seconds prior to tumbling. Leachate samples were filtered by a 0.8-micron membrane filter and analysed for metal ions using an Optima 3000 Inductively Coupled Plasma Atomic Emission Spectroscope (ICP AES).

Leaching fluids used included 0.1 and 0.6 M acetic acid solutions and a municipal landfill leachate. 0.1 M acetic acid is a leaching fluid recommended by AS 4439.3-1997 whereas 0.6 M acetic acid was utilised for comparison with ML leachate on the basis of final pH. That is, both leachants attained a final pH of between 10 and 12 after 7 days leaching. The ML leachate was collected in seven 20-L drums (no headspace) from a pipe leading to a leachate pond in a mature landfill (~20 years old). The landfill received both putrescible and non-putrescible wastes. Approximately 4 L of sample was taken from each drum for characteristic analyses, with the remainder stored at 4 °C. Leachate samples utilising acetic acid as the leaching fluid were preserved by 2% (v/v) HNO_3 addition while the ML leachate samples were stored at 4 °C to minimise microbial activity. Composition of the ML leachate is provided in Table 2. Detailed descriptions of landfill properties, leachate collection, and analytical procedures are described elsewhere [13].

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