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Release dynamic process identification for a cement based material in various leaching conditions. Part II. Modelling the release dynamics for different leaching conditions

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Abstract

This paper deals with process identification and model development for the case of a porous reference material leaching under certain hydrodynamic conditions. Four different dynamic leaching tests have been applied in order to take into account different types of solid/liquid contact conditions corresponding to various real leaching scenarios: monolithic and granular material with sequential eluate renewal, and granular material and continuously renewed eluate with different hydrodynamic conditions (dispersion, residence time). A coupled chemical-mass transfer model has been developed to describe the leaching behaviour under all experimental conditions. Diffusion has been considered as the mass transport mechanism inside the saturated porous material and dispersive convection as that in the leachate. Two specific phenomena have been identified and considered in the model: (i) the early surface dissolution of the material which results in high Ca concentration and (ii) the late weak dissolution of Na and K giving rise to a long-term residual release. The intrinsic material parameters such as the initial concentrations in the pore water and solid phases were determined by applying equilibrium leaching tests and geochemical modelling. Diffusion coefficients for different elements and the late solubility of alkalines have been found to reach the same values in the four tests. The estimated values of the surface dissolution kinetic constant have shown a dependence on leachate hydrodynamics when the thickness of the degraded layer is nearly the same in the four tests (intrinsic parameter of the material). The competition between the four main dynamic processes, i.e. diffusion, convection, late dissolution, and surface dissolution, has been emphasized and compared in the four leaching tests: the hydrodynamic dispersion and the residence time had no effect on the leaching behaviour of alkalines, which is controlled by diffusion, whereas the behaviour of calcium (a major element of the material) was strongly influenced. This has significant effects on eluate pH values and on the concentration of Pb (the monitored pollutant). The model was then applied to simulate a landfill scenario in the case of a stabilized/solidified incinerator residue containing heavy metals and chloride. A high rain infiltration level and the use of small blocs are favourable conditions for enhanced pollutant release.

Keywords: Mass transfer; Surface dissolution; Diffusion; Hydrodynamics; Residual flux

1. Introduction

Environmental assessment of waste and material storage or valorization scenarios involving contact with water requires knowledge concerning contact conditions and their influence on potential release of pollutants from such materials. Such information may be obtained from appropriate leaching tests at laboratory scale and behavioural models, which should allow the identification of the main chemical and mass transfer mechanisms and the competition between different dynamic processes and their relative importance over a given time scale. This second part of the study deals with the process identification and leaching model development for chemical species release under particular hydrodynamic conditions.

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Nomenclature

A, a	coefficients in B-Dot activity model
a_S	specific surface area solid/liquid, m^{-1}
A_{column}	column transversal area, m^2
$A_{specimen}$	specimen external surface area, m^2
B, B^0	coefficients in the B-Dot model
c	element concentration in pore water, mol m^{-3}
c'	element concentration in the lechate, mol m^{-3}
D	diffusion coefficient, $m^2\ s^{-1}$
D_e	effective diffusion coefficient, $m^2\ s^{-1}$
D_L	axial dispersion coefficient, $m^2\ s^{-1}$
E	void volume fraction
$2h$	monolith thickness, m
I	ionic strength, molal
k	dissolution/precipitation kinetic constant, s^{-1}
k_s	surface dissolution kinetic constant, s^{-1}
k_{SL}	solid-to-liquid mass transfer coefficient, m s^{-1}
K_s	solubility product
n	number of CSTR
$n_{particles}$	number of particles in the samples
Pe	Peclet number
Q	leachate flow rate, $m^3\ s^{-1}$
r	spherical coordinate, m
R	mean radius of particles, m
s	solid concentration, mol m^{-3} of pore water
s'	solid concentration, mol m^{-3} of leachate
t	time, s
t_m	mean residence time, s
$V_{leachate}$	leachate volume, m^3
$V_{specimen}$	specimen volume, m^3
x	space coordinate, m

Greek letters

δ	thickness of the degraded surface
ε	open porosity of the material
γ	activity coefficient

Superscripts

$'$	related to the leachate

Subscripts

0	initial state
exp	experimental
in	at the inlet of a CSTR
late	late dissolution of alkalines
s	surface
sat	saturation

In the first part of this study it has been shown that when characterizing leaching behaviour of the same material using different dynamic leaching tests, we obtain different results concerning the concentrations and quantities released of different chemical elements (mg/kg, mg/m^2). The reference porous material was obtained by solidification of PbO and CdO with Portland cement CEM I. The inflow leachate is demineralized water. The four tests presented use the material in solid granular or monolithic form, the solid–liquid contact surface and the renewal conditions being different. The four tests are: (1) Monolithic Leaching Test (MLT) with sequential total renewal of the eluate, (2) Granular material Leaching Test (GLT) with sequential total renewal of the eluate, (3) Column Leaching Test (CLT) of the granular material with continuous upflow, (4) Granular Leaching Test in a Series of 3 CSTR (GLTS) with continuous flow.

Based on the experimental results, a leaching behavioural model for solidified/stabilized porous materials was developed and applied for the conditions of the four laboratory tests. We have assumed that the mechanisms involved in pollutant transfer from the porous matrix towards the liquid are the same in all scenarios (laboratory tests). However, their order of magnitude and their relative significance depends on the type of liquid/solid contact. In order to demonstrate this fact, we based our model on an approach including: (i) the development of a physico-chemical model describing the major mass transfer phenomena of elements having typical behaviours (soluble, Na, K; weak soluble, Ca; amphoteric heavy metal, Pb), (ii) the identification of the characteristic model parameters, (iii) the analysis and comparison of results obtained in each case study with the aim to validate the model, to establish the parameter relevance and the phenomena hierarchy.

2. The leaching model

2.1. Principle

The model describes the leaching behaviour of the material on two linked levels: (1) chemical reactions and transport mechanisms in the porous system of the material, (2) chemical reactions and transport processes in the leachant (particularly leachant hydrodynamics). The two compartments, material and leachate, exchange flows of chemical species.

2.1.1. The material compartment

A large part of the experimental study was dedicated to the characterization of physico-chemical properties of the material, such as: acid-neutralization capacity, influence of pH on species solubility and qualitative and quantitative identification of the main solid phases involved in species dissolution in the porous system. The initial intrinsic physico-chemical parameters of the material are the same in the different leaching tests and the material compartment