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Experimental percolation under intermittent conditions: Influence on pollutants emission from waste

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Abstract

As a precautionary measure, the re-use (or landfill) of waste requires an environmental assessment of its potential impact. This assessment is usually made by simulating the emission of pollutants with a predictive model based on laboratory tests (standardised batch leaching tests, up-flow percolation tests, acid neutralisation capacity tests [CEN, Characterisation of Waste – Leaching – Compliance Test for Leaching of Granular Waste Materials and Sludges, European Committee for Standardisation (ECS), Brussels, 2002 [1]; CEN, prCEN/TS 14405 Characterisation of Waste – Leaching Behaviour Tests – Up-flow Percolation Test (under specified conditions), ECS, Brussels, 2002 [2]; CEN, prCEN/TS 14429 Characterisation of Waste – Leaching Behaviour Test – Influence of pH on Leaching with Initial Acid/base Addition, ECS, Brussels, 2003 [3]]. These tests are performed with simpler conditions than those occurring in the scenario of re-use (saturated media, permanent inflow . . .).

In order to evaluate the relevance of these tests to be considered as a reference for predictive model, the purpose of this work is to determine how the intermittent hydrodynamic flow influences the pollutants release of unsaturated waste. As a result, we could estimate whether this parameter should be introduced in the model.

Keywords: Unsaturated porous media; Pollutant emission; Municipal solid waste inceneration ash; Fuzzy rule-based model

1. Introduction

The organisation of industrial societies binds its actors to manage wastes, from an environmental and socio-economic point of view. The assessment of different waste management strategies is necessary to minimise its potential impact on health and environment. For instance, the potential impact associated to the re-use of thermal process residues in road techniques is to be assessed in terms of emission of pollutants under various percolation conditions. The European methodology of evaluation EN 12-920, based on parametric laboratory tests and predictive model is usually used to assess this potential impact [4]. Nevertheless, the real emission of the waste in its re-use scenario is much more complex than what might result from these laboratory tests. In particular, the inflow intermittence modifies the water content, the repartition of the gas phase in the media, the chemical reactions and their kinetics . . ., in time and space. That is why there remain questions regarding how the results of such laboratory tests can be extrapolated in order to obtain predictions of potential impacts at different scales and for different percolation scenarios. We should investigate on the minimum level of complexity for tests and models in order to describe and predict properly the behaviour of the waste.

In this context, a multiple-scale leaching project ("LIMULE" [5]) was initiated to examine the influence of scale and flow regime on the mobilization of soluble constituents from municipal solid waste incineration boiler ash and to compare the applicability and efficiency of various models over scales.

The present study is a part of this project and deals more particularly with the influence of intermittent inflow regime on chemical emission of pollutants. To explore this problematic, four down-flow columns are performed in the laboratory. The outflow and pollutant mobilization curves are compared with the results of the standardised up-flow percolation test and two down-flow lysimeter cells performed for the LIMULE program. For these devices, the low eluate analysis frequency does not show the influence of intermittent inflow on chemical species release.

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In Section 2 are described the experiments and Section 3 is devoted to the obtained results.

2. Materials and methods

2.1. Characterisation of the waste

The thermal process residues are boiler ash from a fluidized-bed MSW incineration plant, located in Gien, France. The sampling was conducted only once (18 tonnes).

The ash was mixed with a clean Loire sand (0.1–1 mm) in equal weight proportions because it had a certain tendency to indurate in conditions of prolonged contact with water. The sand–ash mixing was performed thanks to two conveyor belts and a melting tube. The stability of the mixing process was controlled every 15 min. Dry density was measured for 20 different samples of mixture and shows a good coherence of values (medium: 1.32 g/cm^3, maximum: 1.39 g/cm^3, minimun: 1.27 g/cm^3) so that the mixture could be considered as homogeneous.

The chemical composition and grain size distribution of a representative sample of mixture are given in Tables 1 and 2, respectively.

Ca, Si, Al, Fe, K, Mg, Mn, Na, P, Si, Ti were determined by X-ray fluorescence (Philips PW2400). Ag, B, Ba, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb, Sn, Sr, V, W, Zn were determined by ICP-AES after high-sintering at 450 °C and attack with Na_2O_2/HCl. Uncertainties are about 5–10%. Loss on ignition was determined at 1000 °C. The total content of metallic aluminium was analysed by atomic absorption spectrometry after mild attack with brominated methanol. The chlorine content was determined by potentiometric analysis. Total sulfur, total organic and inorganic carbon contents were analysed with a Leco determinator. The BET surface was determined according to the French standard NF X 11-621.

Table 1
Global chemical analysis of the ash–sand mixture (from [5])

Major elements	%	Traces	mg/kg
Al_2O_3	11.5	Ag	4.3
CaO	7.6	As	46
Fe_2O_3	3.64	B	88
K_2O	3.00	Ba	955
MgO	1.3	Cd	4
MnO	0.07	Co	61
Na_2O	1.7	Cr	168
P_2O_5	1.25	Cr(VI)	<5
SiO_2	67.4	Cu	1021
TiO_2	0.69	Mo	8
Weight loss (105 °C)	0.54	Ni	63
Loss on ignition (1000 °C)	1.4	Pb	473
Cl	0.43	Sb	44
Organic carbon	<0.05	Sn	155
Total carbon	0.2	Sr	235
Total sulfur	0.26	V	24
		W	19
Al^0	1.4	Zn	2053

Major elements (total concentrations expressed as oxides in %) and trace elements in mg/kg.

Table 2
Grain size percentage distribution of boiler ash by dry screen analysis (from [5])

	Grain size (μm)					BET surface area (m^2/g)
	<63	63–125	125–250	250–500	>500	
Boiler ash	9	14	30	45	2	2.61

Chemical analysis shows that the sample is rich in Si-bearing phases and to a lesser extent in Al, Ca, Fe and K. The main traces are, in decreasing order, Zn, Cu, Ba, Pb, Sn and Sr. The ash is also rich in metallic aluminium (around 1.4 wt.%).

The granulometric distribution of the ash–sand mixture has a median of 0.4 mm with a 10% fraction lower than 80 μm.

XRD patterns were recorded on a diffractometer Bragg–Brentano (type Siemens D5000, monochromator with Co Kα1 radiation $\lambda = 17{,}890$ Å). X-ray diffraction analysis of the ash reveals the presence of crystalline phases corresponding to quartz, silicates – mainly microcline, plagioclase, diopside and gehlenite – anhydride sulfates ($CaSO_4$), carbonates ($CaCO_3$), iron oxides – hematite – and NaCl. An amorphous fraction is also detected.

2.2. Experimental device

Leaching of the waste in intermittent conditions is investigated using four down-flow columns. The size of the columns is chosen so as to have proportional height versus diameter ratios, in order to correlate fluid outflows and pollutants' concentrations over scales. A stired and initially dry sand–ash mixture sample is puddled uniformly in the columns with a pestle. The material is assume to be homogeneous in each column. The average densities vary from 1.27 to 1.42 g/cm^3 for the different columns. So, even with a strict filling protocol, the similarity of percolation experiments is difficult to obtain. This observation highlights the problematics of reproducibility and extrapolation from laboratory to field scales. A peristaltic pump controls inflow water supply. Water feed rate is equal for each column at a rate of 6.5 g/min and is distributed over one aspersion event of 60 min every 24 h. Given the hydraulic conductivity of the sand–ash mixture (on the order of 10^{-5} m/s), the water feed rate theoretically resulted in unsaturated downward flow conditions. However, a noticeable water accumulation at the head of columns A, B and C suggests that a part of the media is saturated in water (the volumetric saturated water content of the ash–sand mixture is of 47%).

To investigate the role of water ionic strength on the leaching of the waste, two identical columns are used, one fed with demineralised water (column A), and the other one with tap water (column B, as for columns C and D). As the accumulation of water at the head of column A is small (maximum 3 cm in height) and rapidly percolates in the material after the end of the injection, we can still consider input water in column A as demineralised.

Fluid outflow is monitored automatically by a fluid sampler. The eluates are then collected and conditioned for chemical analysis after a 0.45 μm filtration. Al, Ca, Cr, K, Na and Zn