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Application of hydrogeochemical modelling in simulating the transportation of elements in fly ash heap under different disposal systems in South Africa

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A R T I C L E   I N F O

A B S T R A C T

Ash heap modelling of South African fly ash from Tutuka was carried out and the duration of transportation projected for 20 years based on two disposal scenarios, namely; irrigation of ash with rainwater, and irrigation with brines. The hydrogeochemical modeling code, PHREEQC, was applied in the study which gave insights into the speciation, release and transport of elements from the water and brines–fly ash long term interactions. Tutuka ash–water heap model showed a general sharp decrease of total elemental concentrations released during the first 2.5 years simulation as the pH value dropped from 12.6 to 8.7, after which it remained constant and their concentration remained constant up to 20 years. The elements showing this trend included Ca, Mg, Al, Fe, Sr, Zn, Na, K, Li and C(4). Generally, brines caused sharp increase in released concentration of the elements Ca, Mg, S(6) and C(4) for the first 3 years of heap irrigation whereas with water irrigation an opposite trend was observed in which the elemental concentrations decreased. Much of the release chemistry of the elements was closely related to the phase dissolution/precipitation and formation as the major controlling factors. Generally therefore, the modelled leachate quality results revealed that many elements are mobile and move through the ash heap in a progressive leaching pathway. The model could therefore be used to provide reasonable leachate quality from the modelled Tutuka ash heap which may be reaching the ground water. Overall, the ash heap modelling enhanced the understanding of the environmental impacts of ash–water–brines interactions and demonstrated that leachate composition is determined by the following factors; (i) the mass flows from the pores of fly ash, (ii) the surface dissolution of the mineral phases, (iii) the various chemical reactions involved during the ash–brine and ash–water interactions, (iv) the interactions with a gas phase (atmospheric CO_2), (v) the composition of the initial fly ash, and (vi) the leachate flow and hydrodynamics as captured in the conceptual model. Further model validation is recommended with lysimeters to quantatively compare the simulated results against the experimental data and improve on the model.

1. Introduction

Fly ash heaps and dams are potential long-term sources of contamination to surface-water sources and groundwater systems due to their possible enrichment in major and trace elements relative to normal geological materials (Donahoe, 2004). This may occur if they are released into the environment in sufficient amounts, and therefore a long-standing need to assess the release and mobilisation of elements that result from weathering of fly ash,

is important. Geochemical reactions and the mineralogical changes that occur between fly ash components and the chemical species in the brine solutions have been reported as part of the larger collaborative ash–brine project work (Gitari et al., 2009a,b; Mbugua et al., 2013; Ojo, 2010; Ojo et al., 2011, 2007; Petrik et al., 2007, 2009a,b). The interactions between the various species in the fly ash and the brine may result either in neoformed phases (as secondary phases) or in dissolution of the primary phases. The speciation, release, transport and fate of the released mobile elements were investigated. Modelling reactive transport in fly ash–water–brines systems with a view to quantify and characterise the products formed and transport mechanisms involved

has been the focus of our study. Prediction of the leachate quality when fly ash heap is subjected to brines and water irrigation was carried out using PHREEQC as the modelling tool.

Reactive-transport modelling as an emerging research field, aims at a comprehensive, quantitative, and ultimately predictive treatment of chemical transformations and mass transfers within the earth system. The field of modern geosciences is one of the fields in which reactive-transport models have had significant contribution. In their work, Regnier and co-workers (Regnier et al., 2003) noted that reactive-transport models (RTMs) provide platforms for testing concepts and hypotheses, and for integrating new experimental, observational, and theoretical knowledge about geochemical, biological and transport processes. Through numerical computation and simulation, RTMs provide the most valuable diagnostic and prognostic tools available for elucidating the inherently complex dynamics of natural and engineered environments such as our ash heap scenario. Furthermore, RTMs bridge the gap between fundamental, process-oriented research and applied research in the fields of operational modelling, environmental engineering and global change. Reactive-transport models are a recent development and modelers do not have a large body of work from which to draw. The combined capability to model flow, transport, and chemical reactions provides a systematic approach for studying ground-water processes (Parkhurst et al., 2003). For a process-based interpretation of test results and their translation to field situations, sufficient understanding is required of the geochemical and mass transfer processes that control the leaching of contaminants in a percolation regime. This understanding will form the basis of our ash heap modelling in order to determine the leaching and transport mechanism during fly ash–water and fly ash–brine interactions as well as the quality of the leachates.

Even though large uncertainties are associated with the modelling results (Ligia et al., 2004) a reactive-transport model is the only systematic method available to estimate the time dependency of the loads and fate of major and trace elements in a complex ash–brine disposal system transported down an ash heap and to assess the sensitivity of the load estimate to various chemical and physical processes.

This study therefore seeks to model the Tutuka ash heap and demonstrate the application of PHREEQC as an analytical-hydrogeochemical tool in predicting the interaction of water and brines respectively, with fly ash during their co-disposal from Eskom coal-utility plant, Tutuka.

2. Modelling methodology

A description of a one-dimensional advective–dispersive–reactive-transport model which is used to simulate transport of various elements down gradient of an ash heap disposal beds at Tutuka disposal facilities is presented. Model definitions include geometry and boundary conditions, initial conditions, and selection of chemical reactions. Conceptual models were developed and mechanisms involved were used as the input parameters for the PHREEQC program using a modified Lawrence and Livermore National Laboratory (LLNL) database for inorganics. A description of the conceptual model and the PHREEQC input data code used for the simulations are provided in the subsequent Sections 2.1 and 2.2 respectively. Both fly ash–water and fly ash–brine models had common input parameters except the infilling solutions. The fly ash–water model had water equilibrated with atmospheric CO2 and O2 gases where as the fly ash–brine model had the infilling solution of brines whose composition was as given in the ash–brine interim reports (Mbugua et al. 2009, 2010). The results for both systems are presented in a combined format and given in Section 3.

Previous ash–brine project work (Gitari et al., 2009a,b; Mbugua et al., 2013; Ojo, 2010; Ojo et al., 2011, 2007; Petrik et al., 2007, 2009a,b). provided relevant experimental data for acid neutralization capacity, (ANC), ash and brines characterisation, column dynamic leaching data, water flux and composition data, porosity and permeability data and conceptual model of brines flow in the heaps, all of which were used for the modelling as part of the initial modelling conditions. The flow rates, the volumes of the leachates and the specific solid/liquid (S/L ratio) were imposed at the laboratory scale. In the aforesaid project work, fly ash is reported as the dust-like material that results from the combustion of either hard (bituminous) coal or brown coal (lignite) in a wide variety of combustion processes. Fly ash particles are generally very fine (silt size, 0.074–0.005 mm) and spherical in shape (Ferguson, 1993; Berry et al., 1990; Mehta, 1985). Coal fly ash is a dark grey fine-grained material which is mostly made up of spherical, glassy particles. Mineralogical characterisation of two South African fresh fly ashes (Secunda and Tutuka ashes) indicated the presence of mullite, quartz, hematite, magnetite, maghemite, anhydrite, portlandite, lime, periclase and titanium oxides as the major phases (Hareeparsad et al., 2010; Petrik et al., 2009a,b; Matjie et al., 2005; Mahlaba et al., 2011).

2.1. Conceptual model

Initial conceptual model of Tutuka ash heap entailed capturing the wet disposal method involving irrigating the heap with water to mimic rainwater as well as irrigation with brines as a method for co-disposal of ash and brines. The Tutuka fly ash composition and that of brines (Tables 1 and 2) were based on the work of Hareeparsad et al. (2010) and Ojo (2007) respectively.

The water was equilibrated with atmospheric CO_2 and O_2 gases. In our model a heap height of 12 m was considered. For simplicity, the ash heap was considered as a 12 m column which was cylindrical in section and represents an element of volume within the heap. The column was discretized into 10 cells of equal lengths (1.2 m each) with 4000 shifts and a time step of 157,680 s. It was assumed that there was no diffusion (diffusion coefficient = 0) while a dispersivity of 0.8 m was imposed as adapted from Appelo and Postma estimates) (Appelo and Postma, 2005); the general trend is that macrodispersivity is about 10% of the travelled distance. These assumptions and estimates which gave a more realistic and reasonable values of pH and total elemental concentrations which compared well with some results of the core

Table 1
Modelled mineralogical composition of Tutuka fly ash (am or A = amorphous, mC = microcrystalline, Csh = C–S–H).

Mineral phase	Moles (L)	Mineral phase	Moles (L)
Al(OH)$_3$(mC)	0.00E+00	Kaolinite	5.32E−04
Anhydrite	6.90E−03	Lime	4.41E−02
Brucite	0.00E+00	Magnesite	0.00E+00
Bunsenite	0.00E+00	Millerite	0.00E+00
CaCrO$_4$	3.00E−05	Mullite	5.00E−03
Calcite	6.90E−03	Ni(OH)$_2$	0.00E+00
CaMoO$_4$	1.80E−07	Ni$_2$SiO$_4$	3.00E−06
Celestite	0.00E+00	NiCO$_3$	0.00E+00
Cr(OH)$_3$(A)	0.00E+00	Periclase	2.10E−02
CSH_1.4	0.00E+00	Portlandite	0.00E+00
Csh_gel_0.8	0.00E+00	Pyrite	3.00E−04
Csh_gel_1.1	0.00E+00	SiO$_2$(am)	0.00E+00
Csh_gel_1.8	0.00E+00	Sr(OH)$_2$	0.00E+00
Diaspore	0.00E+00	SrSiO$_3$	4.59E−04
Ettringite	0.00E+00	Zincite	0.00E+00
Fe(OH)$_3$(am)-CF	0.00E+00	Zn(OH)$_2$(gamma)	0.00E+00
Gypsum	0.00E+00	Zn$_2$TiO$_4$	2.00E−06
Hematite	3.00E−02		