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Multiphase flow and reactive transport model in vadose tailings

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ABSTRACT

Weathering processes affecting pyritic wastes may generate huge amounts of acid waters with high concentrations of potentially toxic contaminants (acid mine drainage). Acid mine drainage is mainly produced in the vadose zone. In the present study, a coupled non-isothermal multiphase flow and reactive transport model of the vadose zone of sulfide mine tailings was developed. The geochemical model included kinetically controlled reactions for Fe(II)-oxidation and for the dissolution of sulfide and aluminosilicate phases and the Pitzer ion-interaction model to describe the behavior of the pore-water solutions. Model results were compared with experimental observations in unsaturated column experiments under strongly evaporative conditions similar to arid or semiarid climates. Evolution trends for temperature, water saturation, evaporation rates, pore-water hydrochemistry and mineral phases observed during the drying experiment were adequately reproduced. The coupled model reproduced the increase of solute concentrations in the column top and the precipitation of a crust of secondary mineral phases. This crust became a barrier for water vapour diffusion to the atmosphere and modified the thermohydraulic behavior of the tailings. Enhanced downward migration of water vapour and its condensation in this colder end of the column were correctly taken into account by the model, which reproduced the dilution observed in the lower part of the column during the experiments.

1. Introduction

Weathering of sulfidic mine wastes and the resulting generation of acid waters with high concentrations of potentially toxic contaminants (acid mine drainage; AMD) is a severe environmental problem in many regions. Its importance has motivated much research aimed at studying weathering processes in pyritic mine tailings. Most of this work has been devoted to the identification of the geochemical processes determining the evolution of tailings (Blowes and Jambor, 1990; Blowes et al., 1991; Al et al., 1997; Holmström and Ohlander, 1999; Dold and Fontboté, 2001, 2002; Moncur et al., 2005; Gunsinger et al., 2006). Typically, the interactions between pore-water and mineral phases have been characterized by combining multiple analytical techniques with geochemical speciation calculations. These studies have led to the identification and description of geochemical processes. Oxygen diffusion into the tailings promotes pyrite oxidation and the oxidation of Fe(II) to Fe(III), which in turn drives further dissolution of sulfides. In general, if carbonates and aluminosilicates are not abundant enough, the dissolution of sulfides lowers pH and releases potential pollutant elements to pore-water. The geochemical evolution of tailings also is determined by the precipitation of secondary phases, which may act as permanent or temporary sinks for the released elements and may produce even more acidity than the dissolution of sulfides. This type of work, though critical for understanding these systems, is essentially qualitative.

Quantitative evaluation of tailings oxidation requires reactive transport modeling because the occurrence, location and rate of reactions are controlled by both chemistry and transport. Therefore, it is not surprising that many recent studies have developed and applied reactive transport models of tailings (for exhaustive reviews of this topic, refer to Gerke et al. (1998, 2001), Mayer et al. (2003)). Reactive transport modeling has helped determine the relative importance of the various processes taking place in tailings impoundments (e.g. rates of sulfide and aluminosilicate dissolution at the field scale, Fe(II)-oxidation rates and the catalytic role of microorganisms, precipitation of secondary phases, role of O_2 diffusion, effect of oxidation coatings on the dissolution rates of sulfides). This modeling has also underscored the need to carry out on-site calibration of reaction rates, which are often orders of magnitude smaller than those derived from laboratory tests. However, these studies have mainly focused on the evolution of the saturated zone of tailings because they were developed for humid areas (e.g. Canada, Sweden).

Since many sulfide oxidation rates are enhanced by O_2 availability, the above studies have concluded that diffusion of O_2 is the limiting factor for sulfide weathering in nearly-saturated tailings.

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As a result, the importance and magnitude of geochemical processes (that is, the generation of AMD) should be greatly enhanced in unsaturated zones. Yet, few studies have performed reactive transport modeling in the vadose zone. This scarcity may be attributed to the lack of codes to deal with multiphase flow coupled to geochemical reactions, and also to the difficulty in obtaining reliable sets of field or experimental data to compare with model results. Therefore, the majority of earlier studies developing reactive transport models for the vadose zone of mine tailings have assumed time-invariant boundary conditions and thermohydraulic properties and have approached it either as a single reactor (Strömberg and Banwart, 1994; Salmon and Malmström, 2004) or as a profile or section (Wunderly et al., 1996; Gerke et al., 1998, 2001; Bain et al., 2000; Mayer et al., 2002; Saaltink et al., 2002; Jurjovec et al., 2004; Linklater et al., 2005; Molson et al., 2005; Molins and Mayer, 2007). In the studies in which fluctuating boundary conditions or thermohydraulic properties have been taken into account, a simplified reactive transport problem has been considered (Xu et al., 2000; Lefebvre et al., 2001a,b). In some cases, the developed models have been used as a tool for prediction of the long-term evolution of sulfide wastes.

These approaches may be adequate for humid areas, where pore-water evolution is controlled by infiltration and changes in the vadose zone can be comparatively slow. In subarid areas, sparse rainfall favours evaporative conditions, O_2 access into the tailings, faster sulfide dissolution rates and enhanced precipitation of secondary phases. Since mineral precipitation and dissolution modify the properties of tailings, a full coupling between thermohydraulic and geochemical processes is required. Understanding these interactions motivated a series of laboratory experiments, which have been detailed in Acero et al. (2007a). However, the processes involved are complex and their full interpretation requires the implementation of a global conceptual model. Therefore, the objective of this paper is to present a coupled transient thermohydraulic and reactive transport model. This model is used for interpreting the evaporative column experiments presented in Acero et al. (2007a), in the hope of shedding light on the processes determining the depth distribution and time evolution of hydraulic properties, mineral phases and pore-water geochemistry.

2. Model description

2.1. Conceptual model

The column experiments are described in detail in Acero et al. (2007a). In the experiments, 10 columns with inner diameters of 14.2 cm, and lengths of 30 cm were filled with fresh and water-saturated mine waste from a tailings impoundment. Each column was placed with the top surface 60 cm below an infrared bulb, which acted as a constant heat source throughout the 125-day experiment. Water loss by evaporation was determined by weighing. The columns were dismantled at four sequential stages (after 16, 30, 70 and 125 days from the beginning of the experiment) and vertical profiles of moisture content, temperature, mineralogy and pore-water concentrations were investigated.

Water saturation was almost constant with depth but decreased with time from 1 to around 0.50. Temperatures increased with water desaturation, especially in the top portion of the column. Thus, the top centimeter reached almost 47 °C by the end of the experiment, whereas the bottom surface of the column remained at a nearly constant temperature of 38 °C. This steep thermal gradient played a key role in the evolution of the column experiments, as discussed below.

The column experiments are shown schematically in Fig. 1. Lamp radiation heated the top surface, causing evaporation and decreasing water pressure, which caused water to flow upwards. Solutes carried in the upflowing water precipitated upon evaporation, thus forming a mineral crust that grew upwards. As evaporation progressed, liquid saturation was reduced, which inhibited liquid flux towards the surface. Therefore, evaporation and mineral precipitation started taking place further below the surface and forced the crust to grow downwards. This caused the column top to become increasingly dense and less permeable. Since the reduction in the crust permeability and diffusivity inhibited upwards flow, water vapour was forced to flow downwards. Vapour condensed at depth, thus diluting concentrations of the solutes. In parallel to the reduction of liquid saturation, gaseous species (notably O_2) started flowing into the column, activating oxidation reactions.

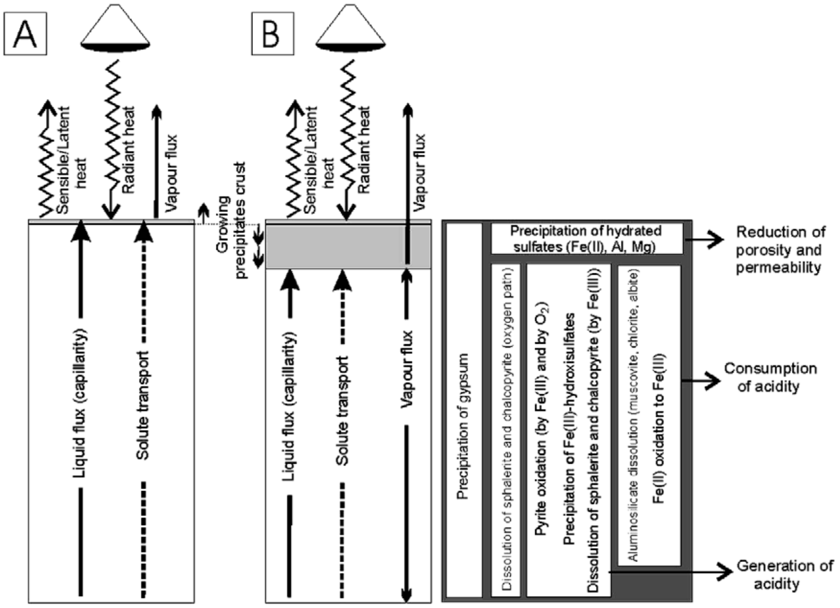

Fig. 1. Conceptual model for the main thermohydraulic and geochemical processes considered in the model. A corresponds to the initial stages and B to the advanced stages in the column experiments.