GEOCHEMICAL AND HYDRAULIC PROCESSES IN THE VADOSE ZONE OF MINE TAILINGS

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Acid Rock Drainage is one of the main environmental problems in today's world. In most cases, the development of acid waters with pH values commonly in the range from 2 to 4 and high metal loads (Nordstrom and Ball, 1985) is caused by the interaction of fresh waters with sulfide minerals, mainly pyrite (FeS₂). In the presence of oxygen, pyrite dissolves oxidatively (Moses and Herman, 1991; Evangelou, 1995), releasing sulfate and acidity to the coexisting solutions. Although there are other sulfides (e.g. sphalerite) which do not require the presence of oxygen to dissolve (Abraitis *et al.*, 2004), their dissolution and the subsequent stability in solution of the released aqueous trace metals and metalloids is highly enhanced by the presence of the acidity associated to pyrite dissolution (Banks *et al.*, 1997).

One of the most common scenarios for ARD generation are mine tailings. These structures are usually simple ponds in which the powdered material resulting from milling operations is deposited and kept water-saturated during the operational time of the exploitations in order to avoid sulfide dissolution. Once the mine is abandoned, the cover of water over the waste disappears by evaporation, allowing the income of oxygen and starting the chemical reactions which lead to ARD production. These problems are especially dramatic in arid climate under which the tailings remain unsaturated for most of the year, which allows the access of oxygen deeper in the waste material.

The processes involved in the ARD generation in mine tailings under arid climate were studied in the abandoned

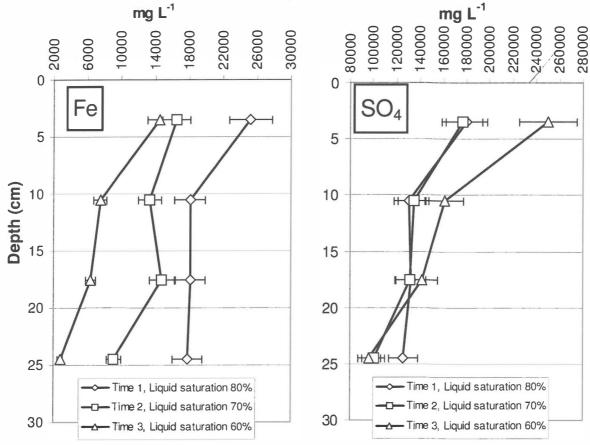


Figura 1: Evolution of the iron and sulfate concentrations throughout the column experiment, in which the evolution of a geochemical divide can be observed.

waste ponds of the Monte Romero mine (Pyritic Belt, Huelva, Spain) and under controlled laboratory conditions. For the field study, the main parameters involved in unsaturated flow (water content, suction, relative humidity, temperature) were monitored in a vertical profile throughout almost 5 years (from 2001 to 2006) by means of several automatic samplers and a datalogging system. Pore-waters at different depths were also extracted by suction lisymeters or by squeezing of borehole samples and characterized chemically. These waters show pH values close to 1 and very high iron and sulfate concentrations (up to 70 g L⁻¹ and 200 g L⁻¹, respectively).

Borehole samples of the tailings material were also studied by sequential extractions, X-Ray Diffraction, Scanning Electron Microscopy and conventional microscopy. Finally, the contents in oxygen gas in the pores at different depths in the monitored vertical profile were also measured at different times of the year, showing that this gas is exhausted by sulfide oxidation reactions in the first 50 cm depth of waste material.

For the laboratory experiments, ten PVC columns were filled with unoxidized, water-saturated tailings material from the Monte Romero mine. The columns were forced to dry at high rate under controlled conditions and the evolution of the mineralogy and of pore-waters and pore-gas were studied at different stages of evolution applying a methodology analogous to the field study and obtaining similar results. Owing to the extreme laboratory conditions, the geochemical processes within the waste material were accelerated compared to the processes observed in the field. Evaporation of the pore waters forced and upwards movement of the water and the solutes generated by the dissolution of sulfides and silicates. The combined effects of generation of new solutes by mineral dissolution and decrease in the mass of solvent by evaporation led to the precipitation of iron hidroxysulfates and to the evolution of a clear

geochemical divide (Fig. 1, evolution with increasing dryness from 80% to 60% of liquid saturation). Thus, whereas sulfate concentrations increased from 125 g L⁻¹ at the beginning of the experiment (water-saturated columns) to 250 g L⁻¹ at the end, iron concentrations followed the opposite trend, decreasing throughout the experiment from 18 at the beginning to less than 3 g L⁻¹ at the end of the column experiment.

Another interesting outcome observed in both in the column experiments and in the field is the precipitation of thick crusts of Fe(II), Al and Mg oxyhidroxysulfates (namely rozenite, szomolnokite, halotrichite, hexahidrite) over the surface of the waste. These crusts are potentially very important for the subsequent evolution of the waste material because 1) its presence causes a great decrease in the evaporation rate, and 2) the physical structure of the crust can act as barrier for the diffusion of oxygen.

The interpretation of the results from both parts of the study indicate that only with the coupled study of flow and reactive transport processes can the evolution of mine tailings be understood and their behavior and polluting potential be assessed.

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