Diavik Waste Rock Project: A Mechanistic Approach To The Prediction Of The Geochemical Evolution Of Sulfidic Waste Rock

David Wilson[1], Richard T. Amos[2], David W. Blowes[1], Jeff B. Langman[3],
Leslie Smith[4], David C. Sego[5]

[1]Department of Earth and Environmental Sciences, University of Waterloo, Waterloo, Canada.

[2]Deptartment of Earth Sciences, Carleton University, Ottawa, Canada.

[3]Department of Geological Sciences, University of Idaho, Moscow, U.S.A.

[4]Department of Earth, Ocean and Atmospheric Sciences, University of British Columbia, Vancouver, Canada.

[5]Department of Civil and Environmental Engineering, University of Alberta, Edmonton, Canada.

Abstract

Prediction of the geochemical evolution of mine-waste rock is of critical importance to mining companies and regulators because the impacts of mining activities on the local environment must be considered as part of mine-life planning. Prediction is often achieved by using the results of small-scale kinetic tests in conjunction with scale factors in a process commonly referred to as scale-up. One of the overall goals of the Diavik Waste Rock Project was the development of a mechanistic scale-up technique for the prediction of the geochemistry of mine-waste effluent. The project consisted of laboratory and field experiments to investigate the geochemical evolution of waste rock at different scales in support of scale-up research. The laboratory study consisted of long-term (9 years) humidity cell (0.1 m) experiments; the field studies consisted of medium scale (2 m) lysimeter experiments and large-scale test pile (15 m) experiments.

A conceptual model of the geochemical evolution of the Diavik waste rock was implemented at the humidity cell scale using the multicomponent reactive transport model MIN3P. The humidity cell experiments were conducted with replicate cells under three different sulfide mineral contents and two temperature conditions (5 and 22 °C). Mass- and temperature-dependent rate parameters for reactions involving sulfide content, host mineralogy, and secondary minerals were calibrated by simulating the effluent geochemistry of the humidity cell experiments. The resulting calibrated model of the geochemical evolution of Diavik waste rock was then implemented for the field-scale experiments, using site measured temperature and hydrological parameters that provided conditions representative of the experiments. Only the sulfide and carbonate mineral contents of the field simulations were changed from the humidity cell simulations; initially, no other calibration was conducted.

Results of the simulations at the three experiment scales indicated that carbonate mineral availability was likely more limited in the field experiments when compared to the humidity cell experiments. Otherwise, the geochemical evolution of the field experiments was captured well by the scaled model.

These simulations indicated that a reactive transport model, calibrated using results of humidity cell experiments, has the potential to predict the geochemical evolution of mine-waste rock at the large scale without the use of scaling factors. Site specific measurements of physical parameters including precipitation, temperature, hydraulic conductivity, and mineralogy would be required to conduct the appropriate simulations at other sites.

Keywords: scale-up, reactive transport, sulfide oxidation, waste rock

Introduction

The assessment of environmental impacts due to waste rock stockpiling is a critical component of the mine planning process. The potential for acidity and elevated solute concentrations from waste-rock are often assessed by conducting laboratory-scale humidity cell experiments (Lapakko, 2003; Ardau, Blowes & Ptacek, 2009; Sapsford et al., 2009). Solute release rates from the small-scale experiments are typically extrapolated using scale factors to make predictions about waste-rock effluent chemistry (a process commonly referred to as scale-up). Parameters understood to influence effluent chemistry, including pH, particle size, moisture content, temperature, oxygen availability and mineral surface area, (Kempton, 2012; Amos et al., 2015) are some of the scale factors that are typically considered for a scale-up assessment. These empirical scale factors often do not take in to consideration site specific heterogeneities and variability in flow and temperature regimes that can make it very difficult to approximate the geochemical and physical behaviour of a waste-rock pile in a realistic manner. Mechanistic approaches to scale-up, specifically the use of reactive transport models, may be able to address these complexities and provide a quantitative approach to scale-up that more closely approximates the geochemical conditions at a waste-rock pile.

The Diavik Waste Rock Project (DWRP) consists of small-scale laboratory (0.1 m), and medium-scale (2 m) and large-scale (15 m) field experiments conducted to address issues related to scale-up. The Diavik Diamond Mine (Diavik) is located approximately 300 km northeast of Yellowknife, NT, Canada. Simulations of the geochemical evolution of low-sulfide waste rock produced at Diavik were conducted using the reactive transport code MIN3P (Mayer, Frind & Blowes, 2002). This article presents the implementation of a conceptual model of sulfide mineral weathering, developed at the laboratory scale, to simulate the geochemical evolution at the two field-scale experiments. Further detail on each stage of the modelling can be found in Wilson et al., (2018a), Wilson et al., (2018b), and Wilson et al., (in prep). The simulations are intended to demonstrate a mechanistic method of scale-up of laboratory-scale experiments for the assessment of the geochemical evolution of waste rock at the large scale.

Methods

Waste rock used in the experiments exhibited similar mineralogy consisting of approximately 75% granite (primarily quartz, K-feldspar, and albite), 14% pegmatitic granite, 10% biotite schist, and 1 % diabase (Blowes & Logsdon, 1998; Langman et al., 2014). The biotite schist was composed primarily of albite (35–55%), quartz (20–50%), and biotite (10–25%). The dominant sulfide mineral, pyrrhotite with minor substitution of Ni and Co for Fe following an approximate ratio of $[Fe_{0.852}Ni_{0.004}Co_{0.001}S]$ (Jambor, 1997), was contained primarily in the biotite schist component of the waste rock. Other sulfide minerals present in the waste rock included minor amounts of chalcopyrite $[CuFeS_2]$, sphalerite $[ZnS]$, and pentlandite $[(Ni,Fe)_9S_8]$. The waste rock at Diavik is sorted into three streams according to S content (Type I < 0.04 wt.%, Type II 0.04 to 0.08 wt.%, and Type III > 0.08 wt.% S) (Smith et al., 2013a).

Each of the 4 humidity cell experiments (small-scale laboratory) discussed here was conducted over a 9 year period. The humidity cells were each constructed with 1 kg of Type III waste rock with mean S content of 0.18 wt.%. The weathering cycle for the humidity cells was based on a weekly schedule of flooding the cells with 500 mL deionized water (first week of the experiment included three flooding events then twice weekly flooding events for the second and third weeks), dry air, and wet air per the ASTM D5744 (ASTM, 1996) protocol and the Lapakko and White (2000) modification of the protocol (Langman et al. 2014). The humidity cell experiments were conducted in duplicate at two temperatures (22 °C and 5 °C).

The Type III active-zone lysimeter (AZL) experiments (medium-scale field) were conducted over 11+ years at Diavik and consisted of two HDPE vessels (2.2 m in diameter × 1.7 m in height). Duplicate experiments were constructed with Type III waste rock with mean S content of 0.053 wt.%. Flow volume from the AZL experiments was measured using rain gauge tipping buckets connected to drains. Samples for geochemi-

cal analysis were collected from the drains every 2-3 days during periods of flow. Temperature was measured at the AZL experiments at depths of 0.3, 0.6, and 1.0 m below ground surface.

The Type III test pile experiments (large-scale field) were conducted over a period of 11+ years at Diavik and included a waste-rock pile constructed of Type III material measuring 50 m × 60 m × 15 m high with mean S content of 0.053 w.t.% (Smith et al., 2013b). Flow volume from the interior of the test pile experiment was measured using rain gauge tipping buckets connected to lysimeter drains at the base of the pile. Samples for geochemical analysis were collected from the drains every 2-3 days during periods of active flow. Temperature was measured at the interior of the test pile at intervals of ≈0.5 m through the top 11 m of the experiment and below the liner of the test pile (≈14-16 m below the pile crest).

Samples collected from each of the experiments were analysed for a suite of 42 inorganic and general parameters, which included Fe, Ni, Co, Cu, Zn, Al, Si, Ca, Mg, K, Na, and SO_4, pH, and alkalinity. Thorough descriptions of the humidity cell, AZL, and test pile experiment sampling procedures are provided by Langman et al. (2014), Bailey et al. (2015), and Smith et al. (2013c), respectively.

Conceptual Model

An integrated conceptual model for sulfidic mine waste-rock weathering developed to simulate the geochemical evolution of Diavik waste rock in a humidity cell experiment (Wilson et al., 2018a) was scaled to simulate the geochemical evolution at the Diavik field experiments. Similar mineralogy was used for the simulations with the exception of S and C mineral contents which differed between the laboratory and field experiments. Temperature and infiltration rates were constant for the humidity cell simulations. The temperature for the field experiment simulations were based on the measured daily values at the AZLs and test pile experiments. Infiltration of water through the field experiments was calculated on a daily basis from measured precipitation data using the FAO Penman-Monteith (FAO P-M) method described by Neuner et al. (2013).

Simulations of the Type III humidity cell effluent were conducted to facilitate calibration of the model using S and C mineralogy specific to the laboratory experiments. Specific calibration parameters included diffusion coefficients for the sulfide minerals, surface area for the host minerals and equilibrium constants for the precipitation and dissolution of secondary minerals. Calibration of the influence of temperature on rate constants was also conducted for the humidity cell simulations. Simulation of the Type III AZL and Type III test pile experiments were conducted by adjusting the S and C content to match the measured S and C content of the field experiments. Physical parameters including hydraulic conductivity and soil hydraulic values were also adjusted to represent the field experiments. Only the matrix portion (material < 5 mm diameter) of the field experiments was considered in the simulations. The temperature of the AZL simulation was uniform throughout the simulation domain; the temperature for the test pile simulation varied with depth within the simulation domain consistent with measured values. Temperature within simulations of the field experiments fluctuated daily and flow and reactions within the test pile simulation stopped when T<0°C.

The integrated conceptual model of sulfidic mineral weathering is based on oxidants $O_{2(aq)}$ and Fe^{3+} being dominant drivers of sulfide mineral oxidation (Nordstrom & Southam, 1997; Blowes et al., 2003). The stoichiometry for generic pyrrhotite can be used to describe the overall reaction of oxidation by $O_{2(aq)}$ (Nicholson and Scharer, 1998; Janzen et al., 2000; Belzile et al., 2004):
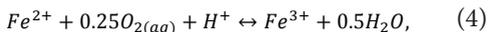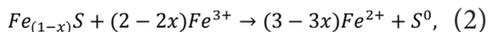
$$Fe_{(1-x)}S + (2 - 0.5x)O_{2(aq)} + xH_2O \rightarrow (1-x)Fe^{2+} + SO_4^{2-} + 2xH^+,$$
(1)
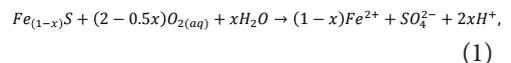
Versions of equation 1 for $CuFeS_2$, ZnS, and $(Fe,Ni)_9S_8$ were included in the simulations to approximate oxidation of the remaining sulfide minerals by $O_{2(aq)}$.

To simulate the oxidation of sulfide minerals by Fe^{3+}, three parallel reactions were used, following a reaction mechanism proposed by Schippers and Sand (1999). The oxidation of Fe^{2+} was kinetically controlled and was included because it of its role in the supply of Fe^{3+} to the geochemical system (Singer

and Stumm, 1970; Schippers and Sand, 1999; Rohwerder et al., 2003).

$$Fe_{(1-x)}S + (2-2x)Fe^{3+} \rightarrow (3-3x)Fe^{2+} + S^0, \quad (2)$$

$$S^0 + 1.5O_{2(aq)} + H_2O \rightarrow SO_4^{2-} + 2H^+, \quad (3)$$

$$Fe^{2+} + 0.25O_{2(aq)} + H^+ \leftrightarrow Fe^{3+} + 0.5H_2O, \quad (4)$$

Site specific host minerals were included based on host rock mineralogy. Host minerals biotite, muscovite, albite, calcite, and dolomite were represented by mineral specific rate expressions coupled with reaction stoichiometry. Secondary minerals were included based on the results of speciation modelling which indicated that Fe(III)(oxy)hydroxide, jarosite, gypsum, siderite, gibbsite, and amorphous silica could all be present at one or more of the experimental scales. Secondary minerals were allowed to precipitate or dissolve according to mineral specific equilibrium-controlled rate expressions coupled with reaction stoichiometry.

All simulations were conducted using 1-D domains sized to the experiment (e.g. the test pile simulation domain was a 1 m × 1 m × 12.5 m column). Physical parameters such as hydraulic conductivity and van Genuchten soil hydraulic parameters were obtained from Neuner et al. (2013).

Results and Discussion

The simulated effluent concentrations for the humidity cell experiments indicated that the conceptual model as implemented captured the measured effluent concentrations well for parameters SO_4, Fe, Ni, and pH over the course of the 9 year simulation period (fig. 1). The simulated effluent concentrations for the AZL and test pile experiments (fig. 1) indicated that the calibrated model captured the measured effluent concentrations well for parameters SO_4, Fe, Ni, and pH over the course of the 5 year simulation period; however, pH was slightly over-estimated in later years in both cases. Additional simulations for the AZL and test pile experiments indicated that carbonate mineral availability may be reduced at the field-experiment scale (calibrated pH shown in fig. 1).

The effluent concentrations of Fe, Ni, Co, Cu, Zn, Al, Si, Ca, Mg, K, Na, and SO_4 were influenced in the simulations by multiple processes including primary and secondary mineral precipitation and dissolution, sorption, and temperature. For example: i) SO_4 concentrations were influenced by sulfide mineral oxidation and precipitation and dissolution of jarosite and gypsum, ii) Ni, Co, Cu, and Zn concentrations were influenced by sulfide mineral oxidation and sorption to Fe(III)(oxy)hydroxide minerals, and iii) Fe and Al concentrations were influenced by the dissolution of multiple host minerals (biotite, muscovite, sulfides (Fe only)) and precipitation and dissolution of Fe(III)(oxy)hydroxide and gibbsite respectively.

Conclusions

This reactive transport code implementation of a conceptual model of sulfide oxidation developed at the laboratory scale indicated that a mechanistic approach to scale-up can result in a reasonable assessment of expected waste-rock effluent. The core scale-up parameters used in these simulations are routinely measured at mine sites and include mineral content, temperature, and rainfall. It is suggested that there is good potential for implementation of this approach at other sites because of the use of the relatively small number of readily available parameters required for the simulations.

Acknowledgements

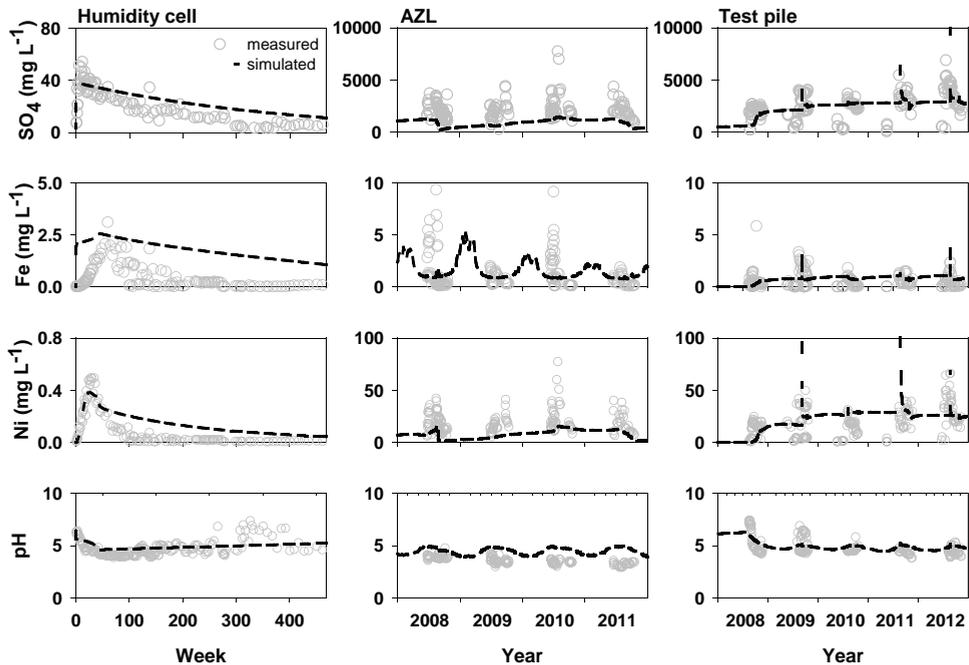*Figure 1* Concentrations of mineral weathering products SO4, Fe Ni, [mg L-1] and pH versus time [weeks, year] measured in Type III warm temperature humidity cell, Type III AZL, and Type III test pile effluents compared to aqueous concentrations exiting the simulation domains.

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