SIMULATING TRANSPORT AND GEOCHEMICAL EVOLUTION OF ACID MINE DRAINAGE THROUGH DISCRETELY FRACTURED POROUS MEDIA

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ABSTRACT
A modelling study is performed to assess the evolution of acid mine drainage (AMD) in fractured subsurface environments using two simulation methods: the equivalent porous medium (EPM) and the discrete-fracture network (DFN) approach. The 2D simulations are conducted using the numerical flow and reactive transport model POLYMIN (Molson et al. 2005) based on a mixed silicate-carbonate host rock overlain by reactive tailings. The POLYMIN model uses the local equilibrium assumption (LEA) for geochemical reactions and embeds discrete 1D or 2D planar elements into the porous matrix blocks to represent the discrete fracture network. Multi-component advective-dispersive transport, geochemical speciation and water-rock interactions are accounted for. The conceptual modelling approaches are compared using predicted pH and concentrations of aqueous oxidation products including SO$_4^{2-}$ and Fe. The EPM vs. DFN comparisons show that even for a densely-fractured network, AMD plume evolution can be significantly different from that assuming an equivalent porous medium approach. The results have implications for future coupling of hydro-geochemical and geo-mechanical models for predicting environmental impacts of acid mine drainage in complex mining environments.

RÉSUMÉ
L'évolution du drainage minier acide (DMA) dans des milieux fracturés est étudiée ici à l'aide de deux méthodes de simulation : le milieu poreux équivalent (MPE) et l'approche par réseau de fractures discrètes (RFD). Les simulations en 2D ont été réalisées en utilisant le code numérique d’écoulement et de transport réactif POLYMIN (Molson et al. 2005). Le modèle conceptuel étudié consiste en une couche de résidus miniers placée au-dessus d'un massif de roches fracturées constituées de silicates et de carbonates. Le modèle POLYMIN utilise l'hypothèse d'équilibre locale (LEA) pour les réactions géochimiques et inclut les éléments 1D ou 2D dans la matrice poreuse pour représenter le réseau de fractures. Le transport advectif-dispersif de divers composés, la spéciation géochimique et les interactions eau-roche sont pris en compte. Les différents modèles conceptuels sont comparés entre eux en utilisant, entre autres, le pH et les concentrations des produits d'oxydation en solution, tels les SO$_4^{2-}$ et le Fe. Les comparaisons montrent que, pour un réseau densément fracturé, l'évolution du drainage minier acide (DMA) obtenue avec le RFD peut être sensiblement différente de celle définie par l’approche utilisant le MPE. Les résultats ont des implications pour les travaux futurs visant à développer des modèles couplés combinant l'hydrogéochimie et la géo-mécanique afin de prédire les impacts du drainage minier acide sur les environnements miniers complexes.

1 INTRODUCTION
Predicting the evolution of acid mine drainage (AMD) is of increasing interest to the mining industry due to its potential for long-term environmental damage. AMD is characterized by its low-pH water and typically by high concentrations of sulphate, iron and dissolved metals (e.g. Aubertin et al., 2002; Ptacek & Blowes, 2003; Price, 2003). The behaviour of this acidic water is controlled by the hydro-geochemical environment in which water-rock interactions play an important role.

To date, research into the evolution of AMD in the environment has focussed on transport through porous (unfractured) media (e.g. Wunderly et al. 1996; Bain et al., 2001; Mayer et al. 2003). In many mine tailings environments, however, there is a high potential for transport through fractured rock. For example, in northern Quebec the fractured rock of the Canadian Shield underlies many tailings deposits. Also, there is increasing world-wide use of paste tailings for backfill in stopes excavated in rock mass (e.g. Aubertin et al. 2002; Martin et al. 2006). Backfill and tailings may then be in direct contact with the host rock which can be fractured naturally or by blast-induced damage.

Numerical modelling of contaminant transport through fractured media is challenging due to complex system geometry, demand for high computational resources, and spatial scales which vary from fracture apertures on the
order of tens of microns to flow system scales of hundreds of metres or even kilometres. The addition of geochemical reactions to these types of systems is currently an active field of research. Ghogmu & Therrien (2000), Graf & Therrien (2005) and Lipson et al. (2007), for example, present modelling approaches for different types of fractured systems. MacQuarrie and Mayer (2005) and Neuman (2005) provide excellent reviews of current issues and challenges related to fracture flow and transport modelling.

In this paper, a numerical model for flow and reactive transport using a discrete fracture network (DFN) approach is presented and tested using a conceptual model of acid mine drainage migrating through a fractured porous medium and reacting with the host rock.

The purpose of this paper is i: to test the performance of the DFN POLYMIN model for acid mine drainage through complex fracture networks, ii: to evaluate the effect of fracture density on evolution of the reactive plumes (including comparisons with an equivalent porous medium – or EPM approach), and iii: to provide some preliminary results on the sensitivity to dispersion and fraction of buffer minerals.

3 CONCEPTUAL MODEL

The conceptual model for the numerical simulations is a 2-D vertical section of fractured porous rock overlain by a source of reactive tailings (Figure 1). The system is 100m long and 20m deep and is discretized using 500x100 rectangular elements in the horizontal and vertical dimensions, respectively, giving a spatial resolution of 20 cm in each direction.

The system is assumed initially gypsum-free but gypsum fraction of buffer minerals. Preliminary results on the sensitivity to dispersion and 2/3 of the total execution time, respectively. The final matrix equations for both the flow and geochemical transport problems are solved using an efficient preconditioned conjugate gradient (PCG) solution for symmetric matrices (Schmidt & Braess, 1988).

Typically, the transport and chemical steps account for approximately 1/3rd and 2/3rd of the total execution time, respectively. The primary assumptions in the model applications herein include isothermal, saturated and steady state flow, equilibrium speciation, an orthogonal fracture system and a non-deforming 2-D porous medium. Changes in mineral volume fractions are assumed small and thus do not influence the porosity, permeability or fracture apertures.

Full details on the model including governing equations, solution scheme and other examples, can be found in Molson et al. (2005, 2006). Other versions of the model include density-dependent thermal transport and temperature-dependent geochemistry (Molson et al. 1992, 1994; Molson & Yang, 2007).

2 POLYMIN NUMERICAL MODEL

POLYMIN is a finite element numerical model for simulating groundwater flow and multi-component reactive transport in porous and fractured porous media (Molson et al. 2005; 2006). The model includes modules for transient or steady state flow, geochemical speciation using the MINTEQ/A2 code (Allison et al. 1991) and oxygen diffusion with kinetic sulphide mineral oxidation. Model domains can be discretized using either triangular or rectangular elements for 2-D systems, or by using rectangular prisms for 3-D systems.

POLYMIN uses a two-step sequential physical-chemical coupling method to solve the reactive transport equations (Walter et al. 1994a; Steefel and MacQuarrie 1996). In this approach, the transport equation is split into a physical step, and a chemical step. The equilibrium chemical step is completed independently for each grid node.

The primary assumptions in the model applications herein include isothermal, saturated and steady state flow, equilibrium speciation, an orthogonal fracture system and a non-deforming 2-D porous medium. Changes in mineral volume fractions are assumed small and thus do not influence the porosity, permeability or fracture apertures.

The initial background and source concentrations for the aqueous components are provided in Table 1. The source of acid mine drainage was based on that given in Walter et al. (1994b) for the Elliot Lake tailings deposit. Thus, the tailings are assumed oxidizing at a constant rate with the AMD source concentrations applied as a transport boundary condition (direct sulphide oxidation using the shrinking core model (e.g. Davis & Ritchie, 1986) is also available in POLYMIN, but for simplicity was not included herein).

Background mineralogy was assumed dominated by silicate minerals (99%vol.; represented by a single pseudo-component of silica) with small amounts of calcite, siderite, gibbsite, and ferricydrite (each < 0.1%). The system is assumed initially gypsum-free but gypsum
is allowed to precipitate. Equilibrium constants from the standard MINTEQ database were used. Jurjovec et al. (2002) use a similar conceptual geochemical model in a porous medium mine tailings column experiment.

In the transport simulations, the longitudinal and transverse dispersivities were 1.0m and 0.005m, respectively (after Gelhar et al. 1992 and Schulze-Makuch 2005), and the aqueous diffusion coefficient for all components was assumed $1 \times 10^{-10}$ m$^2$/s. Time steps varied from 0.1 to 5 days.

Execution times for a 12-component, 4000-day, 50,000-node simulation using a Pentium IV, 3.3Ghz machine were on the order of 48 hours.

<table>
<thead>
<tr>
<th>Component</th>
<th>Background mol/L</th>
<th>Source Conc. mol/L</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca$^{2+}$</td>
<td>5.39E-03</td>
<td>1.08E-02</td>
</tr>
<tr>
<td>Mg$^{2+}$</td>
<td>1.94E-03</td>
<td>9.69E-04</td>
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<tr>
<td>Na$^+$</td>
<td>1.31E-03</td>
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<tr>
<td>K$^+$</td>
<td>6.65E-05</td>
<td>8.14E-04</td>
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<tr>
<td>Cl$^-$</td>
<td>1.03E-03</td>
<td>1.58E-02</td>
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<tr>
<td>CO$_3^{2-}$</td>
<td>3.38E-03</td>
<td>4.92E-04</td>
</tr>
<tr>
<td>SO$_4^{2-}$</td>
<td>7.48E-03</td>
<td>5.00E-02</td>
</tr>
<tr>
<td>Mn$^{2+}$</td>
<td>4.73E-05</td>
<td>7.84E-03</td>
</tr>
<tr>
<td>H$_2$SiO$_4$</td>
<td>3.35E-04</td>
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</tr>
<tr>
<td>Fe$^{2+}$</td>
<td>1.02E-03</td>
<td>3.06E-02</td>
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<td>Fe$^{3+}$</td>
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<tr>
<td>Al$^{3+}$</td>
<td>1.10E-05</td>
<td>4.30E-03</td>
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<tr>
<td>pH</td>
<td>7.2</td>
<td>4</td>
</tr>
</tbody>
</table>

4 FRACTURE NETWORKS AND FLOW FIELD

Three discrete fracture networks were generated for the current study (Figure 2). The fracture systems were generated using the NETWORK model (Molson, 2006) which creates random orthogonal networks of 2-D planar fractures and embeds them seamlessly onto faces of the rectangular prism elements in the 3-D mesh. Thus, hydraulic heads, aqueous and solid concentrations are computed simultaneously for the fractures and porous blocks without iteration.

Fracture density decreases from Case I to Case III and each set assumes a log-normal distribution of fracture apertures varying from 400-600 µm (Figure 2). For each case, the mean fracture lengths were 10m in x, and 5m in z, with a standard deviation of 2m. Mean fracture spacings are 1m, 2m and 4m for Cases I, II and III, respectively.

In each simulation, the steady state flow field was simulated concurrently with the transport simulations. Once steady state had been reached after the first time step, the flow module was automatically de-activated and the reactive transport simulation continued to 4000 days.

Figure 2. Random fracture networks used in the reactive transport simulations, showing Case I (high density), Case II (intermediate density), and Case III (low density) networks. The log-normal aperture distributions for the vertical and horizontal fracture sets are also shown (N is the number of fractures within each aperture range).

Figure 3 shows the steady state hydraulic head distribution and flow velocity vectors for the Case II system of intermediate fracture density. The hydraulic heads show a dominantly horizontal flow gradient with several local perturbations due to the more conductive fractures or fracture zones. Fracture velocity vectors are dominantly horizontal, and increase toward the downgradient boundary at the right. Flow velocities within the porous medium matrix blocks are highest in the upper 2m non-fractured porous layer, and are three orders of magnitude lower within the less permeable rock matrix. Thus, transport of the aqueous components from the tailings source is controlled by flow through the fractures. Similar conditions apply for Cases I and III.
TRANSPORT SIMULATION RESULTS

Each simulation was run to a maximum time of 4000 days, or about 11 years. The results from Case II – of intermediate fracture density – will be treated as the base case, then compared to the higher density (Case I) and lower density (Case III) systems using selected components. For each fracture network, comparisons are also included with the equivalent porous medium cases.

The simulated results for the selected aqueous component and mineral concentrations are shown in Figures 4 and 5, respectively, after 4000 days for the fracture system of Case II. The aqueous component plumes are characterized by a complex web of concentrations which follow the more conductive fracture network. The individual fractures are clearly discernable from the matrix by their higher concentrations which shows that numerical dispersion has been controlled.

After 4000 days, the chloride plume (which is non-reactive) has reached the right outflow boundary and shows significant vertical dispersion. Sulphate also behaves as an essentially conservative component since the mass lost to gypsum precipitation is relatively low (see Figure 5). Relatively more aqueous iron precipitates as siderite (Figure 5) and small amounts of ferrihydrite (not shown), therefore the advance of the iron plume is somewhat retarded. Higher carbonate concentrations within the fractures are the result of calcite and siderite dissolution from the advance of the low-pH plume.

Figure 5 also shows the zone of dissolved calcite which correlates well with the low-pH plume throughout the fracture network. By 4000 days, siderite has dissolved from within the source area where calcite has already been completely depleted. However, siderite also precipitates due to higher carbonate concentrations at the leading edge of the calcite dissolution front (i.e. within the fracture network) and due to high iron concentrations from the AMD source. Gypsum precipitates near the source due to higher sulphate concentrations within the source, and due to higher calcium concentrations from calcite dissolution.

The evolution of the low-pH plume in Case II is shown in Figure 6 at 1000, 2000 and 3000 days (the 4000 day plume is shown in Figure 4). Although representing only a single fracture network realization, the sequence shows a somewhat irregular evolution (in this case through two main lobes) before reaching a more uniformly distributed plume at 4000 days.
Differences in plume behaviour through the three fracture networks of Cases I, II, and III are highlighted in Figures 7, 8 and 9 for chloride, pH, and sulphate, respectively. In each plot, the corresponding simulated plume assuming a homogeneous equivalent porous media approach is also provided.

The hydraulic conductivities $K_x$ and $K_z$ for the EPM approach were determined by running numerical permeability tests in the horizontal and vertical directions for each network. By applying a fixed Darcy flux at the upgradient boundary (i.e. at the left boundary for determining $K_x$ and at the top boundary for determining $K_z$), and a constant head at the downgradient boundary, the POLYMIN model was used to calculate the total head gradient. A thin conductive porous strip at each boundary allowed the water flux to naturally infiltrate the fracture network. Applying Darcy’s law then provides the bulk EPM $K$ for each direction and each fracture network.

In all cases, the DFN model plumes are longer and wider than the corresponding EPM plumes. This was not unexpected, at least for the less dense Case II and III networks, since conservation of mass and mass flux must be maintained. With the same surface recharge in each case, and with low fracture porosities, the fracture flow velocities are significantly higher than those of the EPM model (see Figure 2) thus the plumes in the fractured systems travel further.

Note that the EPM plumes are roughly identical for each case since the surface boundary recharge for contaminant influx was kept uniform. Thus, as the bulk hydraulic conductivity changes, the hydraulic gradient adjusts to maintaining the same total flux.

Some interesting trends emerge from the results shown in Figures 7-9. First, the correlation between the fracture network plumes and the EPM plumes increases as the fracture density increases. In particular, the DFN and EPM pH plumes for Case I agree very well (Figure 8). The fit for chloride and sulphate in Case I is not as good but acceptable nevertheless. The poor correlations for Case II and III clearly show that an EPM approach for mass transport would not be justifiable for these cases.

Secondly, dispersion of the plumes in the fracture networks does not seem strongly correlated to the fracture density. For example, although transverse (vertical) dispersion is weakest in the low density network of Case III, transverse dispersion appears greatest in Case II of intermediate density. This behaviour is likely dependent on the connectivity of these specific fracture network realizations; many more networks would need to be simulated to confirm any definite trends. As in porous
media, however, these fractured cases clearly show the inverse correlation of longer plume distances with lower transverse dispersion.

It is also interesting to note that the 4000-day plumes formed by the network of AMD-impacted fractures have reasonably well-behaved outlines, even for the low-density network of Case III. This suggests that the connectivity in Case III is high enough (i.e. above a critical threshold) such that the plumes are still influenced by the larger scale flow gradients. The fact that the matrix blocks in these cases have a significant (non-negligible) permeability may also play a role.

It must be acknowledged here that some of the discrepancy between the DFN and EPM plumes, especially in the vertical direction for the low density network of Case III, is likely due to difficulty in defining a uniform bulk hydraulic conductivity. Because of the sparse networks and relatively short spatial scale, the simulated hydraulic gradients were not uniform across the domain, thus there was an inherent uncertainty in defining bulk Kx and Kz values. This could be alleviated by running more fracture networks and calculating more representative average bulk K values.

Figure 9. Comparison of sulphate at 4000 days within the fracture network of Case I, II and III, superimposed with the $[SO_4]=0.01$ mol/L contour from the equivalent porous medium (homogeneous bulk Kx,Kz) simulation.

6 EFFECT OF CALCITE FRACTION AND HYDRODYNAMIC DISPERSION

Two sensitivity analysis simulations are presented wherein which show the effects of a higher volume fraction of calcite (Figure 10) and higher dispersivities (Figure 11).

Increasing the calcite fraction from 0.02 to 0.2 mol/L significantly attenuates the low-pH plume thereby limiting the calcite dissolution zone to the local source area (Figure 10). In this case, since the pH is still being buffered by calcite dissolution near the source even after 4000 days, the advance of the low-pH plume is significantly limited and hence there is no siderite.

Figure 10. Simulated pH, calcite and siderite concentrations from the high-calcite scenario (calcite increased from 0.02 to 0.2 mol/Lw), Case II fracture network, 4000 days (compare with Figures 4 & 5).

Figure 11. Simulated aqueous plume concentrations from the high-dispersion scenario (longitudinal and transverse dispersivities increased from 1.0 to 5.0m and from 0.005 to 0.025, respectively. Case II fracture network, 4000 days (compare with Figure 4).
precipitation within the fracture network as was seen in the base case with less calcite (Figure 5). Instead, there is a siderite precipitation zone within the source area which persists until the calcite is dissolved.

Increasing the longitudinal and transverse dispersivities of the porous medium and fractures by a factor of 5 most notably increases the transverse (vertical) spreading within the fractured system. Note that a longitudinal dispersivity of 5m is still within the range of observed field data at scales on the order of 100m for sandstones, carbonates, basalts and granites (Schulze-Makuch, 2005). In this case, plume lengths are not significantly affected and the high concentration gradients along the fracture interfaces are still preserved.

7 CONCLUSIONS

The POLYMIN discrete fracture model has been shown useful for predicting the behaviour of acid mine drainage as it migrates through fractured porous media. The simulated infiltration of a field-derived source of AMD into three different fracture networks showed the effects of carbonate pH-buffering, gypsum precipitation and diffusion-controlled dispersion into the porous matrix blocks. The model is limited, however, to assuming equilibrium reactions and requires a fine spatial discretization to control numerical dispersion.

The reactive transport simulations of acid mine drainage through discretely fractured porous media have shown that in most cases, such systems cannot be simplified using an equivalent porous medium approach. The DFN-derived plumes were consistently longer and more dispersed than those simulated using an EPM approach. Nevertheless, after sufficient time, the AMD-impacted fracture networks formed general plume shapes typical of those in a porous medium – suggesting all cases had an above-threshold fracture density, even for the low density network of Case III simulated herein.

Research is continuing within the authors’ group in the field of reactive transport modelling, and on coupling these types of hydro-geochemical models (including Hydrogeosphere; see Therrien & Sudicky 1996) to geo-mechanical models in order to account for stress-induced changes in fracture apertures.

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