ÉCOLE POLYTECHNIQUE
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Modelling migration and dissolution of mineral particles in saturated porous media

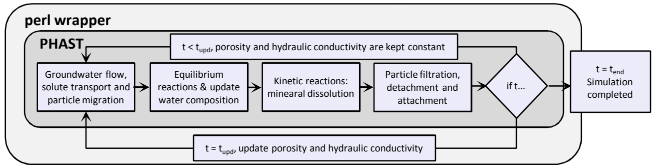
A. Brovelli(1), E. Lacroix(1, 2), C. Holliger(2) and D. A. Barry(1)

(1) Ecological Engineering Laboratory, Environmental Engineering Institute, Ecole Polytechnique Fédérale de Lausanne, Lausanne CH-1015, Switzerland, http://ecol.epfl.ch. Emails: alessandro.brovelli@epfl.ch, andrew.barry@epfl.ch.
(2) Laboratory for Environmental Biotechnology, Environmental Engineering Institute, Ecole Polytechnique Fédérale de Lausanne, Lausanne CH-1015, Switzerland, http://lbe.epfl.ch. Emails: elsa.lacroix@epfl.ch, christof.holliger@epfl.ch.

1. Introduction and motivations

- Migration of solid mineral particles (the so-called *'fines'*) in soils and porous substrates (including geological formations) affects the operation of civil and environmental engineering applications (such as clean-up of contaminated soils, wastewater treatment, water and hydrocarbon production).
- In particular, the dynamics of particles with a diameter in the range 0.1 to 10 um can substantially modify the hydrodynamic properties of porous media.
- The dynamics of particles in the micrometer range is governed by a number of non-linear interrelated processes, including detachment, deposition or filtration and dissolution/precipitation.
- These processes are controlled by the pore-water composition and velocity, and by the chemical/physical properties and mineralogy of the solid phase and are difficult to predict.
- The aim of this work is to develop a simulator to study the dynamics of fines (at the continuum scale) and conduct a sensitivity analysis to identify the parameters that control the distance of migration and the particle's fate.
- The work is motivated by a research project focused on the use of silicate particles to buffer groundwater pH during soil remediation (Robinson et al., 2009; Lacroix et al., 2012).

2. Methodology



- The 3D reactive transport model PHAST is used to simulate groundwater flow, transport and mineral reactions and rate-dependent particle filtration.
- After a user specified period of time (t_{upd}), an external script ('wrapper') is used to modify hydraulic conductivity and porosity and restart PHAST.
- Mineral dissolution is modelled as a rate-dependent process, while precipitation occurs when the saturation index Ω exceeds a user specified treshold (normally $\Omega > 0$). Dissolution rates are computed as

$$r_S = k_{H^+}\frac{\left(10^{-pH}\right)^{-n_{H^+}}}{f_{H^+}} + \frac{k_W}{f_W} + k_{OH^-}\left(10^{-pH}\right)^{-n_{OH^-}},$$

Silicates (Lacroix et al., 2012)

$$r_C = k_1\left[H^+\right] + k_2\left[H_2CO_3\right] + k_3\left[H_2O\right] - k_4\left[Ca^{2+}\right]\left[HCO_3^-\right]$$

Carbonates(Brovelli et al., 2012)

3. Deep-bed filtration

- The three-stage ripening model is used to simulate deep-bed filtration o solid particles.
  (1) Ripening stage: irreversible formation of a deposit monolayer.
  (2) Operable stage: consequent reversible deposit growth.
  (3) Breakthrough stage: halt of further accumulation upon reaching a certain amount of the deposited material.

$$\frac{dX_p}{dt} = \begin{cases} k_r u C_P & 0 \le X_P \le X_P^r \\ k_a u C_P - k_d u^f X_P & X_P^r \le X_P < X_P^u \\ 0 & X_P = X_P^u \end{cases}$$

X_P is deposited particle concentration, $X^r{}_P$ and $X^u{}_P$ are the threshold concentrations for the ripening and breakthrough stages, and C_P is the particle concentration in the liquid

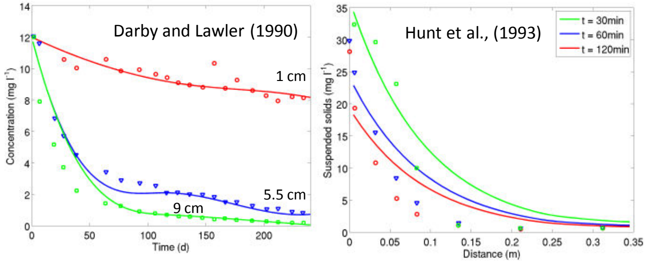
- Porosity (n) is modified using the macroscopic approach, considering the concentration X of c solid (immobile) components, minerals and particles (Brovelli et al., 2009)

$$n(t) = n_0 - \sum_{i=1}^{c}\frac{X_i(t)\rho_b}{\rho_i}$$

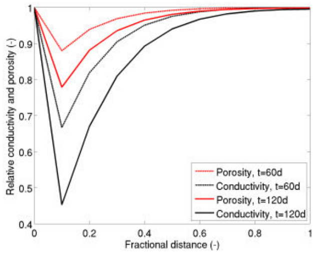
- A power law is used to model hydraulic conductivity (K) changes as a function of porosity, with exponent $p > 1$ (normally $2/3<p<7/2$, e.g. Brovelli et al., 2009)

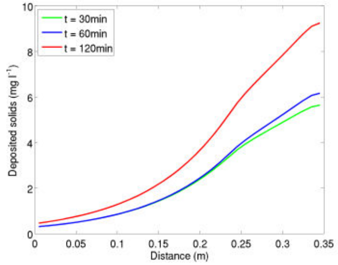
$$K(t) = K_0\left(n_0 - n(t)\right)^p$$

4. Model testing

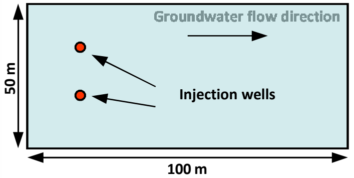


Model application to the experimental data of Darby and Lawler (1990, left) and Hunt et al., (1993, right). Experiments were conducted to study particle retention in sand filters, and ultimately to evaluate the purification efficiency. The diameter of the suspended particles was 2.2 µm.

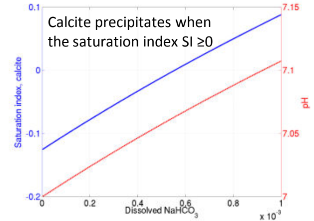




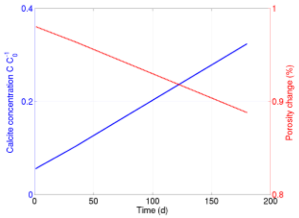
5. 2D application: buffering of acid mine leachate



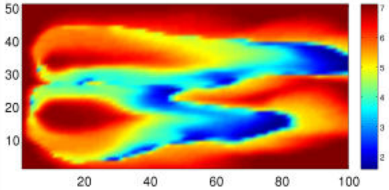
The model was applied to study buffering of acid mine leachate using calcium bicarbonate in a realistic setting. A 2D setup was considered, and a heterogeneous distribution of the hydraulic conductivity was included.



Calcite saturation index for the influent solution as a function of the amount of bicarbonate dissolved.



Calcite precipitation and corresponding porosity change near the injection well.



Spatial distribution of pH after 2 months of treatment. The areas in blue (low pH) are still significantly affected by the acid mine leachate, while around the wells the injected mineral powder was able to buffer groundwater and pH returned to near-neutral values

6. Summary and discussion

- The model seems able to reproduce the results of deep-bed filtration experiments, at least concerning the concentration of suspended and deposited particles. Model ability to reproduce hydraulic conductivity changes is currently tested.
- The model is sensitive to pore-water geochemistry, in particular pH and dissolved ions.
- On the basis of the DLVO theory, attachment (k_r and k_a) and detachment (k_d) coefficients depend on pH and ionic strength of the solution, as well on pore water velocity u. Suitable relationships to describe this dependency are not yet considered.
- Simulation results show that injection of buffer in an acidified aquifer may result in poor performance of the remediation scheme. This is due primarily to calcite precipitation near the inlet, with subsequent reduction of the alkalinity of the injected solution
- While our results show only limited pore-clogging, depending on the amount of dissolved bicarbonate, these process can become a further limitation of this approach

8. References and acknowledgements

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