

Flooding and Post-flooding Scenarios – Dynamics and Geochemistry

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Abstract. The geochemical and geohydraulic modeling of flooding processes is the scientific background for forecast and optimization of remediation. Basic principles are deduced from the dynamical description of flooding and post-flooding scenarios in different types of uranium mines. The time evolution is treated as a transition from dominant *oxidation* processes during flooding to long-term *reduction* processes influenced by reactive materials (organics, scrap iron). The spatial-time structure of the model depends on local conditions. The constructed models include advection, kinetics as well as equilibrium processes.

Box model (compartment model)

For the description of flooding processes in mining sites there is *no* standard software available. The well-known code families for reactive transport (Domenico and Schwartz 1998) are no adequate tools to describe the processes within a network of mine cavities because of the complicated – and in most cases unknown – combinations of flows through cavities, porous media, and/or fracture networks. Therefore a new approach for an *efficient* simulation of flooding dynamics has been developed and applied in several cases.

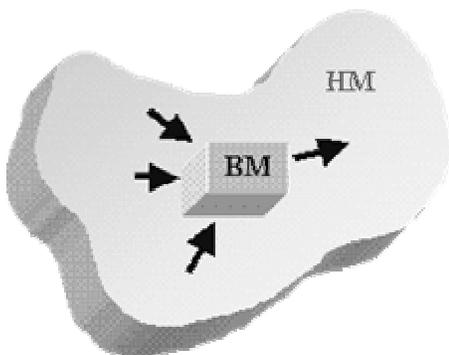
This paper reports some basic ideas and experiences obtained from modeling of flooding scenarios in uranium mines at different locations.

Main principles

To solve the problem numerically with adequate resolution, the mine site is decomposed in several boxes or compartments (up to about 10^3 boxes). The boxes are coupled by hydraulic flows (*internal* couplings to other boxes and *external* coupling to the outside). The structure of the box-system depends on the local conditions (cf. application cases in Figs. 4 - 6).

The main principles of the flooding models can be summarized as follows:

- Spatial-time structure: box model (synonym: compartment model)
- Dynamical description with high time resolution: $\Delta t \approx 1$ h
- Strict mass and charge conservation (local and global)
- Modular system including PHREEQC (Parkhurst and Appelo 1999) as a “subroutine”
- Combination of advection, kinetics and chemical equilibrium.



The box model (BM) represents a 3D distribution of cavity volumes in the mine. As shown in Fig.1, the BM is embedded in a large area hydrogeological model (HM) for groundwater flow simulations. The HM generates the time-dependent external flows as input for the box model.

Fig. 1. Box model (BM) of the mine embedded in a large-area hydrogeological model (HM).

An advantage of the box model is that any box can be equipped with (i) proper mechanisms of mass transformation (geochemistry) and (ii) technological devices like pumps, valves, pipelines, and reactive materials.

Basic equations

The model is based on a “transport equation” which describes the mass change within one box i due to advection and mass transformations (sink/source term):

$$\frac{dm_i}{dt} = \left(\frac{dm_i}{dt} \right)_{adv} + \left(\frac{dm_i}{dt} \right)_{src} \quad (1)$$

This equation holds for any chemical element or species with mass $m = cV$ (c – concentration, V – water volume) where $c(t)$ and $V(t)$ are time-dependent quantities. The advection term describes the hydraulic processes (without reactions) and is given by

$$\left(\frac{dm_i}{dt} \right)_{adv} = \sum_j (Q_{j \rightarrow i} c_j - Q_{i \rightarrow j} c_i) + Q_i^{in} c^{in} - Q_i^{out} c_i \quad (2)$$

where $Q_{i \rightarrow j}(t)$ denotes the flow rate (dV/dt) from box i to box j . $Q_i^{in}(t)$ and $Q_i^{out}(t)$ represent the external flow *from* and *to* the outside region, respectively. Whereas $Q_{i \rightarrow j}(t)$ are calculated from the hydraulic conditions at time t (using Darcy’s Law), the quantities $Q_i^{in}(t)$ and $Q_i^{out}(t)$ are input values (obtained from the hydrogeological model).

The source term in Eq.(1) summarizes all mass transformations between solid and aqueous phases discussed below in more detail.

Linkage between geohydraulics and geochemistry

During the flooding process, the hydraulic conditions (flow pattern, water volume in each box, water level which bisects the mine into a saturated and an unsaturated zone) changes from time step to time step. In this way, the geochemistry which do

strongly rely on the hydraulic conditions becomes also time-dependent.

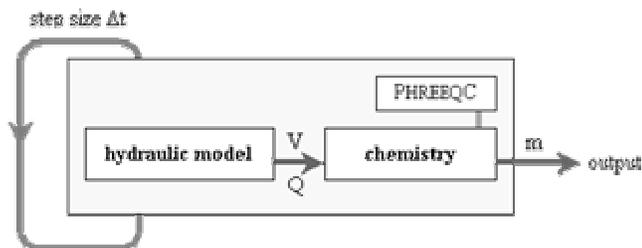


Fig. 2. Two-step-algorithm for geohydraulics and geochemistry.

From a numerical point of view, the flooding dynamics is simulated within a *two-step algorithm*: In each time step of size Δt (≈ 1 h) the hydraulic model calculates the internal flows $Q(t)$ and the water volume $V(t)$, which are the input quantities for the geochemical model (see Fig.2).

Geochemistry

The model predicts the composition $c = c(t)$ of mine water (10 - 20 elements, 100 - 200 species) and its changes in time.

Main processes

Using the geochemical code PHREEQC as a “subroutine” the flooding model includes the following standard processes:

- Mixing of water flows
- Speciation and complexation
- Precipitation and co-precipitation / dissolution
- Oxidation-reduction reactions

Additionally, in-situ water treatment (neutralization, incorporation of reactive materials) can be simulated in a given box, i.e. at any position in the mining site.

To calculate the chemical transformations between different phases (pore fluids, secondary phases, mine water and underground water) PHREEQC is called in the order of 10^6 times during a program run. The software codes have been optimized concerning computing time and accuracy (mass balance uncertainty $< 1\%$).

From oxidation to reduction processes

As shown in Fig.3 flooding is treated as a transition stage where *oxidation* processes gradually cease, and concurrently in the flooded mine, *reduction* processes influenced by reactive materials (wood, scrap iron etc.) become important.

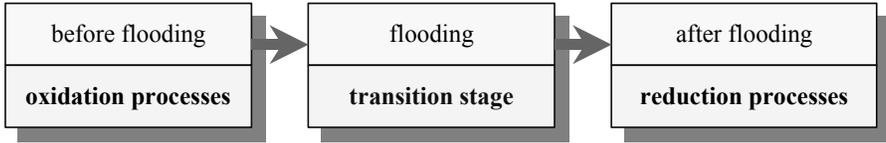


Fig. 3. Flooding as a transition stage between oxidation and reduction processes.

In this way, for large times ($t \rightarrow \infty$) the mine water in the upper levels of the flooded mine (near the land surface) does asymptotically approach a composition close to the water composition before mining.

In the model, the “switch-off” of the oxidation processes and the “switch-on” of the reduction processes automatically occur when the water level increases and the boxes are filled during flooding:

- Box empty – only oxidation processes (dissolution of primary minerals)
- Box partly filled – dissolution of secondary minerals
- Box full – reduction processes and dissolution of backfill materials; precipitation of hydroxides, carbonates and sulfides

Each of these processes is described by an individual source term in Eq.(1).

Source terms – the origin of mass and contaminants

To include chemical reactions and kinetic processes the source terms in Eq.(1) should be specified. Typical terms are given by (abbreviated notation):

$$\left(\frac{dm_i}{dt} \right)_{src} = rF_i \quad \text{and} \quad r = r_0 f(\text{pH}) \tag{3}$$

$$\left(\frac{dm_i}{dt} \right)_{src} = m_i^{sekm} \delta(t - t_i) \tag{4}$$

$$\left(\frac{dm_i}{dt} \right)_{src} = Q_i^{seep} c_i^{seep} \tag{5}$$

Eq.(3) describes the dissolution kinetics of a reactive material or primary mineral (r – specific reaction rate, F – reactive surface in box i). Here the reaction rate depends on pH value. Eq.(4) is used for the dissolution of secondary minerals at time t_i when box i is flooded (m_i^{sekm} – total mass of a secondary mineral in box i).

An alternative description of oxidation processes (pyrite oxidation etc.) is given by Eq.(5) where mass is “produced” by the seepage water inflow rate Q_i^{seep} enter-

ing box i with the concentration c_i^{seep} . Here, the seepage water composition c_i^{seep} can either be calculated within a so-called geochemical infiltration model (Kalka et al. 1998) or directly obtained from monitoring data sampled *before* flooding.

The advantage of the “effective kinetics” in Eq.(5) is that the seepage water (taken from monitoring data) already contains all information about the pyrite oxidation and other related reactions inside the rock matrix (dissolution of heavy metals and radionuclides, neutralization by calcite and dolomite minerals). Thus, using the composition of the seepage water as input, there is no data-need for the specific rates r and reactive surface F (which are unknown in most cases).

In summary, Eq.(3) can be used for the corrosion of backfill materials as well as for the dissolution of reactive materials like wood and scrap iron. The latter are responsible for the reduction processes under *saturated* conditions. On the other hand, Eq. (5) is a good candidate for an efficient description of the “pyrite oxidation” and all relating processes in the *unsaturated* zone.

Post-flooding processes

When flooding is completed the mine system is still in a physical and chemical non-equilibrium.

The origin of the *physical* instability is as follows: The mass production (due to oxidation processes and dissolution of secondary minerals) depends on the cavity volumes (and on the former mine activities) which are locally distributed. This pattern causes a layer structure in the flooding water where water of high density (salinity) might occur above low-density water. Hence, the instable state induces a density-driven *vertical convection*, which has been included in the box model. Calculations for the Ronneburg mine have shown that in view of these “density forces” effects arising from the geothermal gradient are negligible.

Due-to the presence of organic matter and scrap iron the flooding water is in a *chemical* non-equilibrium. To achieve equilibrium reduction processes take place (sulfate reduction). The description of such processes is difficult and lies beyond the “state of the art”. Nevertheless first attempts are made by using kinetics of zero order in form of Eq.(3) with the following rates:

- Dissolution of “ CH_2O ” $r_0 = 1.1 \cdot 10^{-10} \text{ mol/m}^2/\text{s}$
- Dissolution of metallic Fe $r_0 = 3.6 \cdot 10^{-7} \text{ mol/m}^2/\text{s}$
- Backfill corrosion (CaO) $r_0 = 2.0 \cdot 10^{-6} \text{ mol/m}^2/\text{s}$

Each rate is strongly pH-dependent, $r = r_0 \cdot f(\text{pH})$, where in the case of organic matter $f(\text{pH})$ has its maximum at $\text{pH} = 7$. Here, the backfill corrosion is included because of its influence on pH value of the flood water. In the calculations, quasi-equilibrium is assumed at each time step t for calling PhreeqC. As time elapses the sulfate reduction leads to the precipitation of the U(IV)-phase $\text{UO}_2(\text{a})$ and sulfides (FeS, Covellite, Millerite, Orpiment, $\text{ZnS}(\text{a})$ etc.). One open question is the “incubation period” for the microbiological processes. This parameter can only be deduced from observations in mines flooded a long time ago.

Examples for model application

The application of the box model to flooding simulations requires an adjustment to the local conditions at each uranium mine in two respects:

- (i) Spatial discretization of model space in relation to geohydraulics
- (ii) Specification of the inner box structure in relation to geochemistry

Ronneburg uranium mine complex (Thuringia)

The Ronneburg mining site is the most extensive *uranium mine complex* of WISMUT. Between 1952 and 1990, approximately 125 Mio. m³ uranium ore were recovered from underground and open-pit mines. Within the 164 square kilometer area (63 mi.²) of the Ronneburg Revier, 65 km² were developed for mining purposes. 113 000 t U were recovered in total. 2 926 km of mine works were built

underground. The open pit mine had a depth of 230 m. Roughly 60 % of the site has been contaminated by mining activities. This site is unique worldwide, particularly due to its location in a densely populated area.

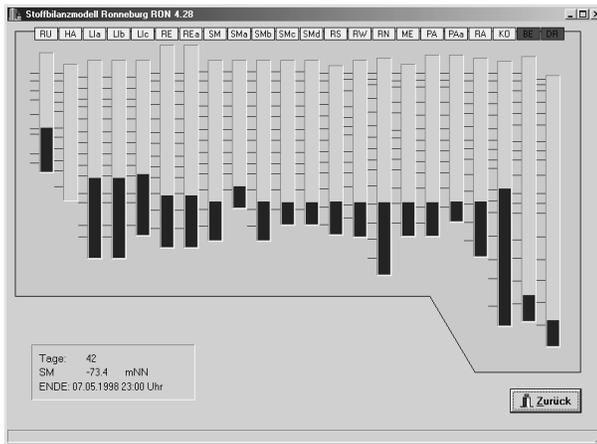
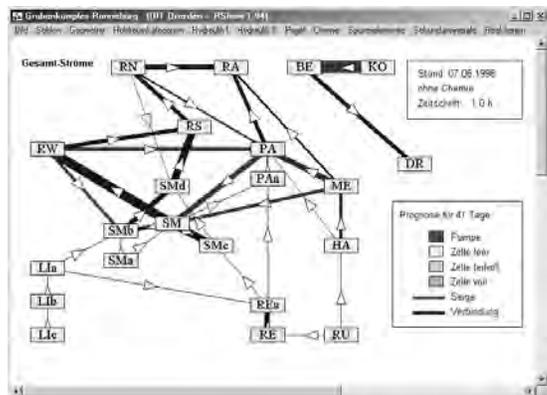


Fig. 4. Vertical cross section (online-graphic of the Ronneburg model)

Model space: The model considers the various types of dewatered cavity volumes for each mine level and each mining field (= 22 × 22 boxes) as well as the hydrogeological connections (vertical and horizontal) between the boxes. Devices like pumps and pipelines are also considered.

Fig. 5. Horizontal cross section through the Ronneburg complex



Königstein in-situ leaching mine (Saxony)

The sedimentary uranium deposit Königstein is located about 30 km SE of Dresden. Mining started in 1967. By 1984 leaching had replaced conventional mining entirely. Since 1991 the remediation of the under-ground mine works had been prepared. Mine flooding started in January 2001.

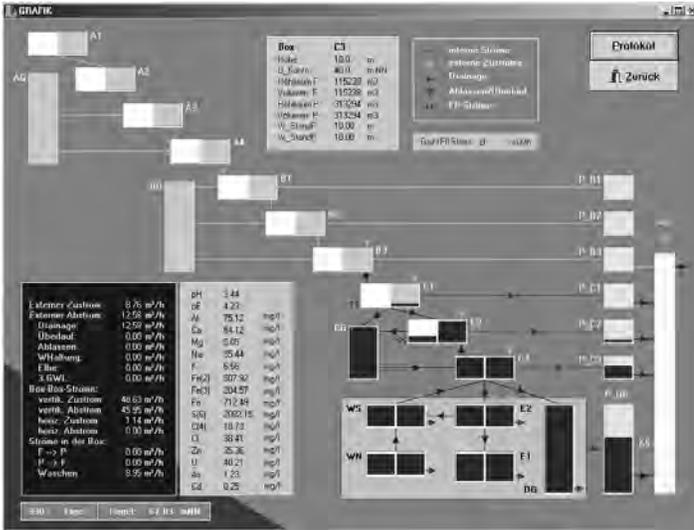


Fig. 6. Online-graphic of the Königstein model software

A “snapshot” of the flooding process is shown in Fig. 6. The model space consists of 28 boxes. The special feature of the Königstein mine is the following: Due to the in-situ leaching technology most of the contaminants are stored in the pore water of the sandstone. Therefore, each box had to be decomposed into two subspaces: (i) “pore volume” P (containing the highly contaminant pore water) and (ii) “cavity volume” F (with connections to other boxes). Both subspaces are coupled by a density driven mass exchange to simulate the washing-out process in the sandstone. Furthermore, “re-flooding”-scenarios have also been considered, i.e. intermediate decline of water level.

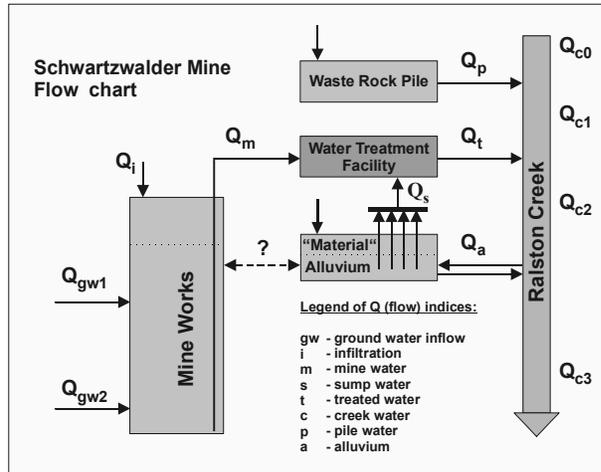
Schwartzwalder mine (Colorado)

From 1953 to the end of the nineties (with a standby from 1989 to 1995) about $1.9 \cdot 10^6$ tons ore with an average uranium grade of 0,47 % had been recovered from Schwartzwalder mine located near Golden, Colorado. The mine is a vein-type uranium deposit (the largest known in U.S.) located in Precambrian metamorphic rock of amphibolite grade. It has been developed from the steep canyon of Ralston Creek down to a level in 2 100 ft depth.

The box model similar to the Ronneburg application case was used to simulate different flooding scenarios in connection with proposed technical reclamation measures (bulkheads, hydraulic connections). Based on the calculations an optimum reclamation concept has been developed. As shown in Fig.7, the flooding

processes in the mine were combined with the total water balance at the site including the optional water treatment. Because of the spatial structure of the mine works a 2-column/21-level model space had been chosen, i.e. 42 boxes.

Fig. 7. Flow chart for combining flooding dynamics with total water balance



Conclusions

The dynamic box model combining the hydrogeological and geochemical description in a consistent manner represents a novel approach for the simulation of flooding scenarios. Various remediation measures can be considered explicitly.

A model complexity up to box numbers in the order of 10^3 are feasible, provided that reliable input data for the characterization of mine structure, hydrogeology and geochemistry are available. In general, the model has to be carefully calibrated on the basis of existing monitoring data, thus, reproducing the previous and present water balance and water quality data.

The dynamic 3D-compartment model is a powerful tool for the forecast of water quality in the flooded mine regions. It is the basis for:

- Evaluation of water quality development during mine flooding
- Development of water management strategies (in situ / ex situ)
- Permitting procedures governed by authorities

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