

Application of 1d reactive transport modelling to evaluate remediation measures at abandoned uranium mines

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INTRODUCTION

Although uranium concentrations are often elevated in granites and gneiss, ground water from such host rocks often shows low to intermediate uranium concentrations, whereas some ground waters derived from sandstone and carbonate aquifers show naturally elevated uranium concentrations up to more than hundred μg per litre. In other situations surface waters may have anthropogenically increased uranium concentrations due to the intensive use of phosphate fertilizers in agricultural areas. However, the most highly elevated concentrations of uranium in ground water are related to mining and milling activities. Saxony and Thuringia, two states of the reunified Germany, are areas where past uranium mining activities have impacted the environment more severely than probably in any other part of the world. In situ leaching by means of alkaline and especially acid leaching agents has endangered ground water dramatically.

This paper demonstrates the prospects of reactive transport modelling in risk assessment studies and rehabilitation measures using the in situ leaching mine Königstein as an example. This mine is situated about 30 km southeast of the city of Dresden close to the Elbe river. Underground operations started at the Königstein mine in 1967 by conventional deep mining in the Cretaceous sandstone ore body. Since 1984 a special in situ leaching technique was applied using sulfuric acid as leaching agent. Uranium mining was stopped at the end of 1991 after reunification of Germany (Zimmermann and Schreyer 1995). The multilayer aquifer consists of four interlayered double porosity aquifers and siltstones units. Aquifers 1 and 2 are unconfined and only of local interest, aquifer 3 is unconfined as well and used downstream of the mine for extracting potable water for the city of Pirna. Aquifer 4 is confined and the lowermost strata with the uranium ore body in it.

METHODOLOGY

Distinct potential reactions were modelled by means of PHREEQC using its 1d reactive transport tool. One principle problem associated with 1d transport modelling is that dilution by the surrounding uncontaminated ground water is not taken into account. However, within PHREEQC this dilution can be mastered in the 1d reactive transport tool. The aquifer was assumed to contain 1% of calcite and pyrite respectively and 2% organic matter. Kinetic rate constants for calcite and pyrite precipitation and dissolution and decay rates for organic matter were taken from Parkhurst & Apello (1999) and partly modified. Since laboratory tests of permeability show k_f -values of approximately 10^{-7} m/s and pumping test data in contrast are approximately 10^{-5} m/s it can be concluded that the Cretaceous sandstone is a double porosity aquifer. For the 1d transport simulation a primary (fracture) porosity of 0.01 was coupled with 0.15 secondary sandstone porosity. Diffusion was assumed to be the major process of exchange between fracture and pore volume.

RESULTS

Cation and anion exchange as well as pyrite dissolution have only minor impacts on concentrations in the contamination plume. However, calcite dissolution, precipitation of iron

hydroxides and consequently surface complexation of uranium and arsenic as well as dilution by uncontaminated ground water are important factors. Assuming that the peak flush of major contaminants will be pumped and treated until the concentrations are decreased by one order of magnitude, the estimated amount of 1% calcite is sufficient to buffer the mine water to a pH between 6.7 and 7.3 (Fig.1a).

A completely different scenario results if it is assumed that easily degradable organic matter is available in addition to calcite and pyrite. The addition of organic matter results in the development of reducing conditions. With decreasing pe, redox sensitive elements like iron, arsenic, copper, and uranium are also reduced. After approximately 520 days no iron(III) remains in the system. Roughly 10 days later uranium(VI) is transformed to uranium(IV) and consequently precipitated due to the low solubility product of uraninite. After about 800 days the pe drops from 17.6 to 4 and reduction of sulfate to H₂S starts.

Figure 1b displays the changes in concentrations of Fe, As and U species. Concentrations of As(V) and Fe(III) decrease; and an increase in As(III) concentration is inversely correlated to As(V). If the thermodynamic data used are reliable and the reduction reactions for Fe(III) to Fe(II) and U(VI) to U(IV) are not kinetically limited, it is likely that sorption of uranium on Fe(OH)₃ and simultaneous precipitation of UO₂ or Uraninite will not likely occur.

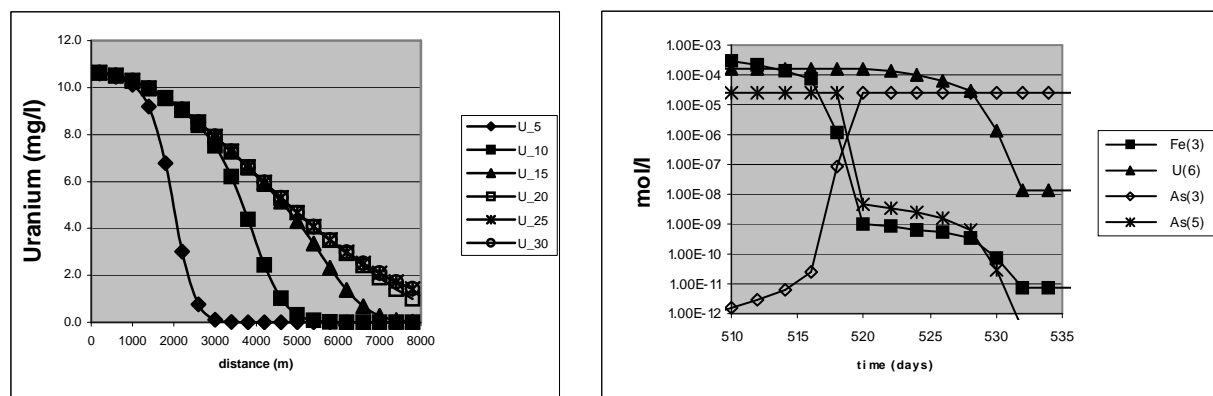


Fig. 1. a) Breakthrough curve for Uranium for time intervals of 5 ... 30 years. Due to iron hydroxide precipitation in the first cell of the model and consequent surface complexation of uranium onto Fe(OH)₃ surfaces, uranium concentrations decrease from 20 to 10.5 mg/l in the first cell. Further decreases in uranium concentrations are due to dilution of the plume.

b) Distribution of redox sensitive species of iron, arsenic and uranium versus time due to kinetically controlled degradation of organic matter

REFERENCES

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