Predicting natural attenuation for flooding of an ISL-uranium mine – potentials and limitations

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Abstract

Flooding of a former ISL underground uranium mine situated in a sandstone aquifer inevitably results in mine waters entering the downstream aquifers influencing the groundwater quality. The mine water contains apart from elevated content of salts (e.g., sulfate, iron) also high concentrations of uranium exceeding the WHO guidance value for drinking waters by several orders of magnitude.

Especially uranium and other metals are of concern in terms of an impact on downstream groundwater quality. Even with high efforts to reduce the contaminant concentrations in the mine water by in-situ hydrochemical measures concentrations in the order of geogenic background conditions are to be achieved only in a time frame of decades. Complex technical efforts and consequently funding have to be assured. Effective and reasonable remediation solutions must consider groundwater-rock-gas interactions leading i) to a specific spatial and temporal development of pH/EH-conditions and ii) consequently to the immobilization and potential subsequent re-mobilization of contaminants (e.g., uranium) in the downstream aquifers. In combination with active remediation measures the contaminant retardation in the downstream aquifer could act as additional safety factor for a sustainable solution.

The paper outlines the approach followed to determine crucial processes of aqueous uranium mobility and related geohydraulic, geochemical, and hydrochemical parameters theoretically (by hydrogeochemical mass transport modeling) as well as in lab-scale experiments for a case study. Mineralogical and geochemical conditions within the affected aquifers were determined based on sampling from a number of boreholes. Using the findings of these studies a hydraulic and geochemical modelling concept was developed allowing predictions as scenario analysis. The concept for the hydrogeochemical model predictions is described with a number of limitations and constraints which mainly arise from limited data gathered on the field scale. The paper therefore especially focuses on the identification of representative parameter sets describing the potentially impacted part of the regional sandstone aquifer.

Keywords: mine flooding, geochemical modelling, parameterisation, contaminant retardation

Introduction

Uranium mining started at the Königstein mining site in 1964. The ore was first mined conventionally while in 1984 the winning technology was fully changed to an underground in-situ-leaching (ISL). The mined uranium ore is partly found in a 300 m deep sandstone aquifer with a considerable hydraulic conductivity. Ore in adjacent less permeable rocks was prepared for leaching by increasing the permeability by mining out compensation volumes followed by blasting of the ore containing rock. This generated additional flow paths for the drainage of the lixiviant. Sulfuric acid was used to leach the uranium from the sandstone and the ambient loosened host rocks. As a result the rocks in the underground mine contain pore water with high salt and metal concentrations and a low pH (tab. 1).

Presently the mine is flooded to a water level hydraulically not affecting the adjacent aquifers. A control drift surrounds the deepest downstream part of the mine. This control drift is separated by a safety pillar of sandstone rock from the flooded open mine workings.

Table 1 *Pore water concentration in leached sandstones of the Königstein mine.*

pH	EC [mV]	SO_4^{-2} [g/L]	Fe [g/L]	Zn [mg/Ll]	U [mg/L]	Al [mg/L]
2.0	700	10	3	200	200	300

Pumping water from the control drift creates a hydraulic depression and thereby assures that the mine water decant is fully collected, pumped out and treated in a water treatment plant. As a consequence the flooded mine is continuously washed resulting in decreasing contaminant concentrations and an increase of pH.

The remediation concept for the mine considers flooding to a natural level. The mine is mainly situated in a permeable sandstone aquifer which is the deepest in an interlayered system of sandstone aquifers and aquitards. Flooding the mine results in groundwater flowing through the mine workings and the leached sandstone block with finally entering the downstream surrounding aquifers. Especially the upper sandstone aquifer is a potential drinking water resource and needs to be protected against adverse effects from the mine flooding. Continuing flooding will lead to an outflow of mine water into this aquifer.

General modelling approach

The general modelling concept for the Königstein site consists of a number of separate modelling tools. A regional groundwater flow model was implemented based on the program code SPRING. It is used to describe the regional groundwater flow conditions in terms of potentiometric surfaces and water flow in the various model domains. Calibration and validation of the model was done using the extensive data set of groundwater monitoring results and mine water pumping. Modelling the different hydraulic states such as mine development, active mining and remediation the model uncertainty in predicting the regional hydraulic conditions was considerably reduced. Using this model the regional hydraulic conditions are described for various remediation options providing the boundary conditions for further modelling tools (Metschies et.al 2013). These additional tools comprise of reactive hydrogeochemical mass transport models describing the chemical conditions in different compartments such as the flooded mine or in the sandstone aquifers downstream of the mine.

The scope of the geochemical modelling is to evaluate the potential impact of mine water entering the aquifer. Even active remediation measures will not lead to contaminant concentrations at the natural background level for a fairly long time. Technical measures will be required as long as either a significantly elevated contaminant level in the surrounding aquifers compared to the natural conditions is accepted or potential retardation effects within a limited aquifer zone are taken into account. In any case model predictions of the dispersion of a contaminant plume resulting from the mine flooding are necessary.

Geochemical modelling approach and parameterisation

Without pumping out mine water balancing the natural water inflow to the mine contaminated water would enter the downstream aquifer which itself contains the remaining not mined ore body. This ore body forms a potential source for a groundwater contamination under specific milieu conditions. Apart from this flow path mine water could also enter the upper sandstone aquifer depending on the hydraulic conditions. In this case contaminants could spread in this at present hydrochemically unaffected aquifer. These two downstream paths are considered individually because the general flow direction in the two aquifers differs and there is no interaction of the potential plumes along these flow paths expected. Each of these two paths could be described in separate geochemical models.

The followed general approach in predicting the geochemical conditions focuses on identification of relevant geochemical processes influencing the fate of potential contaminants downstream of the mine. Due to the fact that the available baseline information concerning the geochemical and mineralogical conditions is limited the predictions are made as scenario analysis with a variation of several parameters based on their identified individual parameter ranges. The geochemical models are implemented using the transport option of the reactive modelling code PHREEQC (Parkhurst et. al. 1999). The thermodynamic model approach is used including the options of surface complexiation and ion-exchange. Such calculations are constraint by the principle laws of mass and charge conservation.

The identification of relevant processes is supported by laboratory experiments conducted in the conception phase. A step wise approach was followed with experiments on various scales including batch and column tests. The batch experiments represented single and combined reactions of the fluid solid mixtures containing elements and minerals relevant for the natural system to be modelled under the milieu conditions in the mine and the downstream aquifers. These batch experiments were reproduced and retraced by respective PHREEQC models to validate the general modelling concept.

For the parameterisation of the model mineralogical and geochemical analysis of rock samples from the relevant sandstone strata are available. Due to the depth of the modelled aquifers only a limited number of exploration drillings with relevant geochemical information are existent. Back in the 1960ies a vast number of drillings were made to explore the uranium ore body. Analysis of samples however were mainly focussed on the metal content with none or just limited data characterising the general mineralogical and geochemical conditions. In addition with the limited mineral content in the sandstones most relevant analysis data is below the detection limit at that time.

The respective sandstone aquifers have a thickness of several 10 metres. Due to the changing sedimentation conditions the characteristics of the rocks is variable over the depth. With saturated aquifer thickness of up to 50 m it is difficult to determine the zones of preferential groundwater flow. The variation of the vertical and lateral hydrogeological conditions, such as porosity and permeability, gives a high variety of the hydraulic and geochemical characteristics, as well. While average groundwater flow conditions could be well estimated using the calibrated and validated regional flow model the geochemical parameterisation is far more complex.

The mineralogical composition of the sandstone is dominated by the geochemically stable quartz but calcite, pyrite and goethite are to be considered as primary mineral phases even with their low concentrations influencing the geochemical conditions in the downstream of the mine outside of the ore body. Within the ore body uraninite, coffinite, sphalerite, orpiment and otavite are additional relevant mineral phases.

The mineral phase assemblage is defined based on the limited information from several new boreholes in the downstream area where more detailed analysis were made to gather a data set representative for the reactive transport modelling (tab. 2). The geochemical and mineralogical data show mostly local characteristics resulting in a high variability of the physical and geochemical properties. The values range several order of magnitude in vertical and lateral direction over short distances including the hydraulic conductivities, grain size distribution and mineral contents.

Apart from a plausible definition of the mineral content it has to be considered which share of the total content is reactive in terms of potential mass conversion in the natural system. This depends on preferential flow conditions as well as coating of surfaces or agglomeration inhibiting the access of potential reactants to the reactive minerals. It is practically impossible to derive values characterising the conditions in the regional flow system simply from the analysis of a limited number of rock samples. As an alternative approach just scenario calculations with varying shares of the mineral content considered as reac-

***Table 2** Range of measured geochemical content of sandstone relevant for geochemical transport modelling.*

Range	Carbonate-carbon [mass %]	Disulfid-S (pyrite) [mass %]	Non-sulfidic iron [mass %]	Potential cation exchange capacity [meq/100g]
min	<0.005	0.02	0.028	<0.02
max	0.02	2.6	0.65	3.4
mean (model)	0.01	0.4	0.19	1.5

tive are feasible. Thereby the reactive mineral content is changed over orders of magnitude in relation to an assumed mean concentration (tab. 2).

The uncertainty of the geochemical model parameterisation could not be simply reduced by additional sampling and analysis of sandstone material because under the given hydrogeological and sedimentological conditions a statistically safe parameter distribution could not be derived. Therefore, looking at the wide range of uncertainties in geochemical model parameterisation a full-scale 3D-reactive modelling seeming to give a biunique prediction is considered inadequate. In case of the Königstein mine it is instead preferred to make calculation under less complex geometrical assumptions to be able to investigate relevant effects and derive conclusion concerning the effect of specific geochemical parameter combinations. Here the 1D transport option of PHREEQC offers a volume based approach with a specified ratio of reacting pore water and solid. Implementing a simple transport model for a 1D-flow path allows a robust and reproducible scenario analysis for the wide range of parameter combinations. By negligence of effects on the groundwater flow due to spatial effects the focus can be put on the relevant processes. Otherwise PHREEQC allows to include diffusion und dispersion effects which can be characterised based on the results of the regional hydraulic modelling and are included accordingly in the geochemical transport model. This holds also for water balance considerations and groundwater flow velocities, as well.

Using this more generic modelling approach the geochemical conditions along the downstream flow path are predicted in close combination with the assumptions made concerning the availability of reactive minerals. Systematic calculation of various scenarios give valuable generalised information concerning the extent of a potential plume in the aquifer and the expected effect of retardation of mainly uranium along the flow path under the considered boundary conditions. It could be shown that the hydrochemical impact of mine flooding in terms of pH, uranium and other metal concentration is limited to a flow distance of a few 100 m downstream of the mine in the upper aquifer even under conservative parameter assumptions. As consequence, in case of the outflow of mine water during the final flooding a limited extend of the aquifer needs to be sacrificed for the retardation of the contaminants. In this part of the aquifer the use of the groundwater has to be restricted. However, with regard to the prohibition of deterioration defined by the EU water framework directive such a concept is difficult to be followed by the authorities in general under the given uncertainties.

Given the data base and the uncertainties of the various model parameters which could not be significantly further decreased by additional analysis or lab experiments a further improvement of the prediction accuracy is impossible unless first real data from the field scale is available. Field scale data would implicitly provide indications helping to better estimate the amounts of reactive minerals present along the groundwater flow path. It thereby leads to integral effective parameters which are more representative for the hydrogeological and chemical setting downstream of the mine than small drill core-derived data from local analysis.

It was agreed with the permitting authorities to make a test flooding of the mine by temporarily increasing the mine water level by another 10 m followed by an immediate lowering to the base level. As consequence, a limited volume of mine water enters the upper aquifer. By intensive monitoring of a number of groundwater observation boreholes along the expected flow path the propagation of the limited plume should be monitored. In a first step the conceptual model should be validated whereupon the low pH of the mine water is buffered and uranium is retarded in the aquifer. It is expected to detect only an increase of the salt content in the observation boreholes as signal for the breakthrough of the mine water.

Results

As figure 1 shows the limited outflow of mine water during the hydraulic test is expected to temporarily slightly decrease the pH-value in the groundwater close to the inflow. Depending on the acidity introduced and the available neutralisation potential the changes in the milieu conditions will be significant enough to be measured. However, an increase of the salt content as e.g. chloride should more clearly show the impact of the mine water. On the other hand uranium is expected to be retarded in any case. Even when the load of mine water entering the aquifer is doubled no measurable changes in concentration are expected. Based on such kind of predictions the mining and water authorities issued a permit for such a hydraulic test.

Presently the hydraulic test is ongoing accompanied by an intensified monitoring of the mine and groundwater composition in the mine and the downstream area. It is expected that first monitoring results showing the impact of the test on the surrounding aquifer will be available in 2019.

Conclusions

Predicting the impact of mine flooding on an adjacent aquifer is clearly constraint by baseline information concerning the geochemical and mineralogical composition. The mineralogical composition strongly affects the efficiency of retardation of certain contaminants. Due to the physical and chemical heterogeneity of the aquifer a representative parameterisation based on chemical and mineralogical analysis of selected drill-core based samples seems to be difficult to achieve. As a workaround scenario analysis allows to derive conclusions concerning general behaviour of the system allowing to make predictions considering the parameter uncertainties. Experiments on a laboratory scale allow to set up a

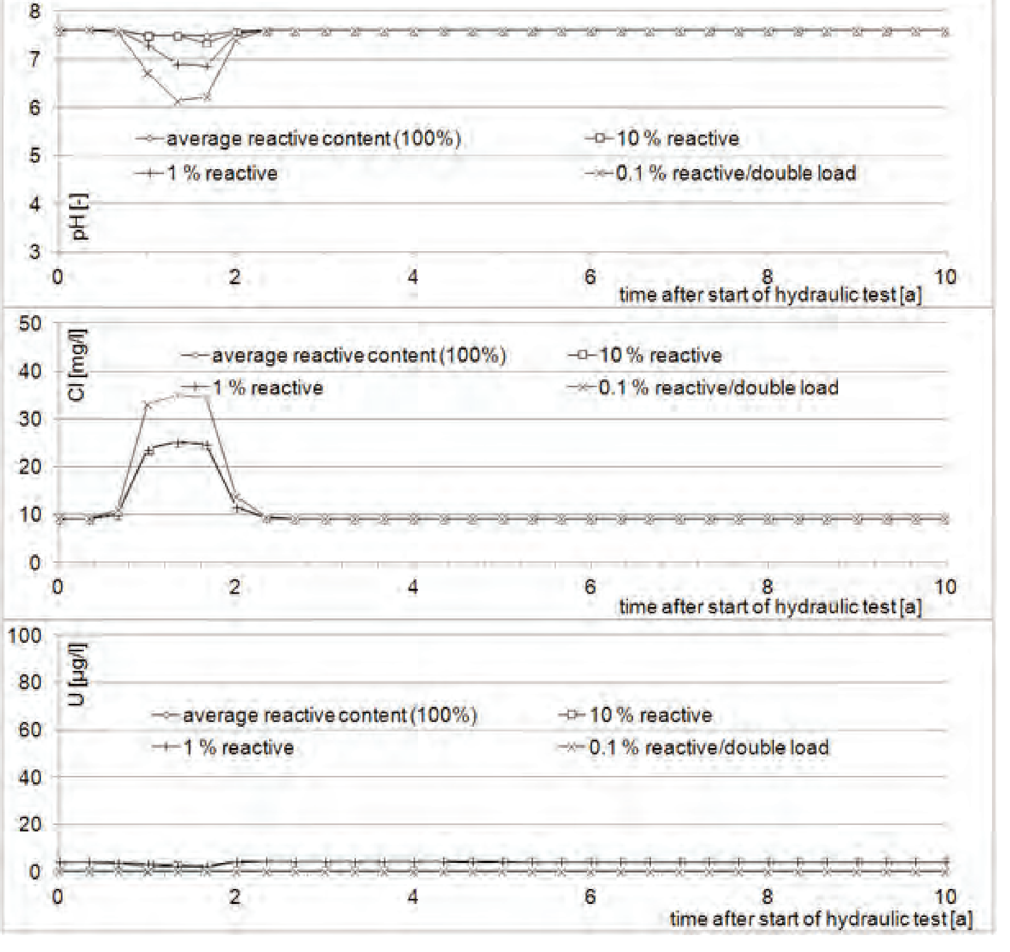

Figure 1 *Predicted concentration curve in an observation borehole about 50 m downstream of inflow of mine water into the sandstone aquifer for selected scenarios varying the reactive mineral content and the load introduced during the hydraulic test.*

conceptual model of the relevant geochemical processes and to validate the respective concept. For a reproducible prediction of the conditions downstream of a mine integral information on the field scale is essential to be able to up-scale the conceptual model and to derive a representative integral data set describing the reactive share of the available mineral content relevant for the retardation and other geochemical processes. This is a precondition to comprehensibly predict the effect of the natural attenuation of contaminants and thereby to include this effect in a sustainable remediation concept.

Outlook

Validating the conceptual geochemical model using the results of the hydraulic test in the Königstein mine will be fundamental to further decrease the uncertainty for predictions of the effect of mine water entering the surrounding aquifers. Using this confirmed and further improved understanding of the main geochemical processes and their interactions with groundwater flow an extension of the model from the column along the flow path to a model with a higher spatial resolution is the next step to improve the predictions for arbitrary points in the downstream flow path by better describing the influence of lateral dilution.

With regard to the final flooding of the Königstein mine a stepwise flooding with continuous monitoring will be necessary. This allows to further adapt the geochemical transport model to the effective conditions in the aquifer downstream of the mine. Only such an approach ensures to reduce the prediction uncertainties of the extent of a retardation of contaminants entering the aquifer with the mine water sufficiently.

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