

Development of an in-situ mine water treatment approach for the flooded Königstein Uranium mine (Germany) – Field experiment

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Abstract Mine water of the partly flooded abandoned uranium mine at Königstein is acidic and oxidizing. In the frame work of the current licensing process, basic approaches for an in-situ water treatment technique have been investigated over the past decade. To test alkalinity injection into the flooded mine, a field-scale experiment using surface boreholes was carried out in 2010. The technological implementation was successful, and test tracer signals and geochemical reactions were observed at several monitoring points over a couple of months. The paper describes the general technological, hydraulic, and geochemical results of the field experiment.

Key Words in-situ mine water treatment, field experiment, alkalinity, Königstein mine

Introduction

Remediation of the Königstein mine near Dresden is a very special case within the WISMUT rehabilitation project in Germany. The mine is situated in an ecologically sensitive and densely populated area (Fig.1). From the early sixties through 1990, approximately 19,000 t of uranium were produced.

The ore body is located in the 4th sandstone aquifer, the deepest of four hydraulically isolated aquifers in a Cretaceous basin. The 3rd aquifer is an important water reservoir for the Dresden region and is environmentally very sensitive.

The uranium was extracted from the 4th sandstone aquifer initially using conventional mining

methods, but later an underground block leaching method using sulphuric acid supplanted conventional mining. Development of the mine site extended over a surface area of ca. 6.5 km² and comprised 4 main levels.

Considering the existing pollution potential and the importance of the overlying aquifer a concept of controlled flooding was developed. The major element of this approach is a control drift system, which allows collection of draining flooding water down-gradient of the deepest part of the mine.

To date, the mine is in a partly flooded state. Some 4.5 million m³ of a total volume of 11 million m³ of mine voids subject to flooding are currently



Figure 1 View of the Königstein mine site/Germany.

flooded. Hydraulic conditions of the mine are very complex, as rapid flow routes (drifts, mined sections) exist alongside less well permeable areas in the sandstone pore space. The water soluble contaminant potential is for the most part located within the pore space, from where it is released very slowly as indicated by findings from past flooding.

Because of the acid in-situ leach process, flood water at the Königstein mine is acidic and heavily polluted with uranium, heavy metals, and naturally occurring radionuclides. With a view to ensuring controlled flooding, mine water is specifically removed via two extraction wells connected to the control drift system and treated.

Experience from flooding so far as well as prognoses give reason to expect that the present chemical conditions will persist for decades to come and hence require comprehensive measures for water treatment and protection of the surrounding aquifers. In view of this fact and in order to comply with regulatory requirements, a series of approaches to immobilise contaminants in the mine were investigated. Following implementation of a procedure to immobilise leach blocks in parts of the accessible southern field of the mine from 2002 through 2005, focus of further R&D efforts shifted to the identification of procedures for the flooded mine.

Based on results of in-house studies to determine the effect of sodium sulphite as well as on acceleration of sulphate reduction processes under a project sponsored by the Ministry of Education and Research, and after clearance with regulatory authorities, a large-scale field experiment was initiated on August 9, 2010 at the Königstein site by injecting a reactive solution into the partly flooded mine.

The primary goal of the effort was to look for approaches to process engineering, which would allow to affect the floodwater body in the direction of restoring pre-mining groundwater conditions and minimise potential contaminant releases as flooding proceeds to the natural groundwater level. To this end, the acid and oxygen-rich flood water will have to be altered by a change in the direction of pre-mining groundwater conditions (pH-neutral, intermediate redox conditions), so that dissolved contaminants will be precipitated within the mine and natural, microbially-catalysed self-purification processes (sulphate reduction) will be enhanced. Large-scale implementation and the effects within the complex geometry of the flooded mine were priority investigation goals.

Methods

The tests were carried out while regular mine flooding continued. Ensuring the continued regular mine flooding was a fundamental require-

ment. The test concept provided for the identification of major areas subject to flooding in the Königstein mine such as drifts, blasted sandstone as well as blocks/pillars. To this end 3 partial tests were performed. The schematic diagram below depicts the spatial conditions and expected flow paths (Fig. 2).

Injection of reactive solutions was performed via existing boreholes from above ground. Measuring points established both above ground and underground to monitor mine flooding as well as specific process control units were available to ensure comprehensive test monitoring. Furthermore, the measuring points were to provide information both on the effect obtained within the contaminant source (the reaction space proper) and on the effect obtained within the relatively vast balance spaces.

Prior to the field experiment the subsequent parameters had to be established prior for every single partial test: flow path, reaction space, flow time, required amount of reactive solution. Prognosis of flow paths and required time frames were based on detailed investigations of the mine workings in their present state.

The 3 partial tests were performed successively. The reactive solution was produced by alkalinity injection (sodium hydroxide solution, potassium hydroxide solution) and sodium sulphite into a motive water flow (groundwater, treated flood water). A mobile dosing station moving from injection point to injection point was used to produce and inject the required volume of reactive solutions. During injection, the test unit was looked after round the clock (Fig. 3).

The test unit consisted of a dosing container, which housed the PC-based central control board, hose pumps, and electrical distribution box. At the container's rear were the metering unit, volume flow meter, potassium hydroxide and sodium sulphite feeder pipes, mixing vessel, pH probe and sampling station. The dosing unit also comprised a chemicals container, housing the moistening station, as well as two 10 m³ storage tanks to stock both the alkalinity carrier (potassium hydroxide solution) and the sodium sulphite solution. Injection of the reactive solution was by the respective boreholes.

As for the open drifts, one had to assume the presence of laminar pipe flow. As a matter of consequence, the rate of propagation is calculated as in fluid mechanics from the quotient of cross-section area and volumetric flow. Flow through the Cretaceous sandstone was modelled as flow in a porous medium. Given a corresponding size of the representative elementary volume, this approach also allowed to simulate fracture flows.

Table 1 depicts the key technological parameters of every single of the three partial tests.

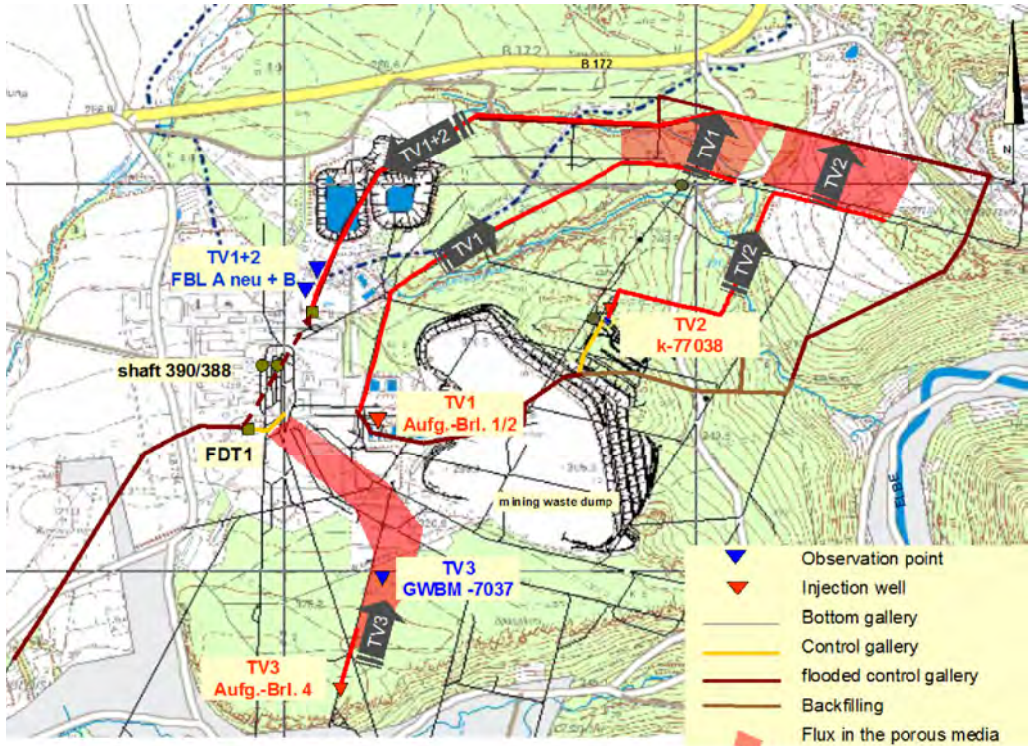


Figure 2 Schematic diagram of key spatial conditions, injection and monitoring points as well as of anticipated flow paths.



Figure 3 Mobile facility for producing and injection of reactive solution (left to right: mobile container for caustic soda, storage tanks, dose and station, moistening station).

Table 1 Key technological parameters of partial tests

	TV1	TV2	TV3
Reaction medium	Cretaceous sandstone	Cretaceous sandstone	Blasted sandstone
Flood water volume	82 500 m ³	74 250 m ³	104 650 m ³
Expected flow time value	114 days	112 days	47 days
Added volume of chemicals	34.4 t KOH 2.4 t Na ₂ SO ₃	34.6 t NaOH 2.3 t Na ₂ SO ₃	43.9 t NaOH 2.5 t Na ₂ SO ₃
Motive water	Groundwater	Water from water treatment plant	Groundwater
Motive water volume	ca. 24 200	ca. 24 700	ca. 48 300
Monitoring site	Extr. well B (k-08200)	Extr. well B (k-08200)	GWMP 7037

Together with the injection of potassium hydroxide solution during partial test #1 and of sodium hydroxide solution during partial test #2, two distinct types of cations were introduced. These cations served as tracer substances for the respective tests.

Acquisition of test results was based on analyses of flood water qualities performed at extraction wells (centralised flood water collection) as well as at selected groundwater monitoring points. Monitoring of partial test #3 involved pumped sampling at groundwater monitoring point GWMP #7037. The discharge volume was ca. 4 m³/h.

Injection of buffer fluids at the three feeder sites (partial tests 1 to 3) ran August 09 through October 04, 2010 and was followed by a monitoring period of ca. 7 months.

An accompanying full-scale column test was conducted in parallel to the field experiment. Among the primary objectives of the test was the acquisition of information on precipitation products and on rock matrix modifications to allow interpretation of long-term processes.

Main Results

Dosing alkalinity carriers into the motive water flow and injection of the generated buffer fluid via existing boreholes into the mine workings during all three partial tests were full-size actions with regard to potential large-scale applications. Smooth implementation of the three partial tests and injection of the buffer fluid demonstrated the full-scale technical feasibility of buffer fluid addition via boreholes (injection wells and groundwater measuring points). The mobile design of the unit has proved to be successful. Moving the unit to a different site was completed without any problem during one working week.

The approach to predicting flow paths, reaction spaces, and flow times has proved to be fundamentally appropriate and rewarding. Penetration patterns of Na and confirmed anticipated flow

path to a large extent.

Findings of the field experiment demonstrated major chemical processes of neutralisation as well as the induced immobilisation of contaminants. By the precipitation of iron hydroxides, neutralisation of iron-bearing flood water induces contaminant immobilisation in the mine.

Chemical effectiveness was demonstrated during all partial tests. The evolution of uranium and zink concentrations as a result of injection is exemplified by partial test #3 (Fig. 4). It became apparent that uranium and zink concentrations were significantly decreased, for a short time interrupted by a technologically conditioned inflow of acid flood water. Residual concentrations are in the order of 0.7–1.0 mg/L for uranium and ca. 0.5 mg/l for zink. This meant a concentration decrease of at least one order of magnitude. Similar significant concentration drops were established for the other heavy metals as well. It is also noteworthy that the 10-day injection of reactive solutions produced significant effects for more than 5 months.

Of particular significance is the reincrease of the pH-value (> 5) coinciding with the decrease of heavy metal levels following the inflow of acid flood water from the untreated surroundings as a consequence of interrupted water injection at injection well #4. Two conclusions may be derived with regard to long-term stability:

- 1 The newly formed precipitates, which induce contaminant demobilisation, are already relatively stable. Further stability increase may be expected in the course of advancing mineralisation.
- 2 To preserve long-term stability, the ingress of acid flood water must be averted.

During partial tests 1 and 2, the decrease in uranium and heavy metal concentrations at extraction wells was lower but still markedly ascertainable against the backdrop of significant mixing with other flood waters. Uranium concen-

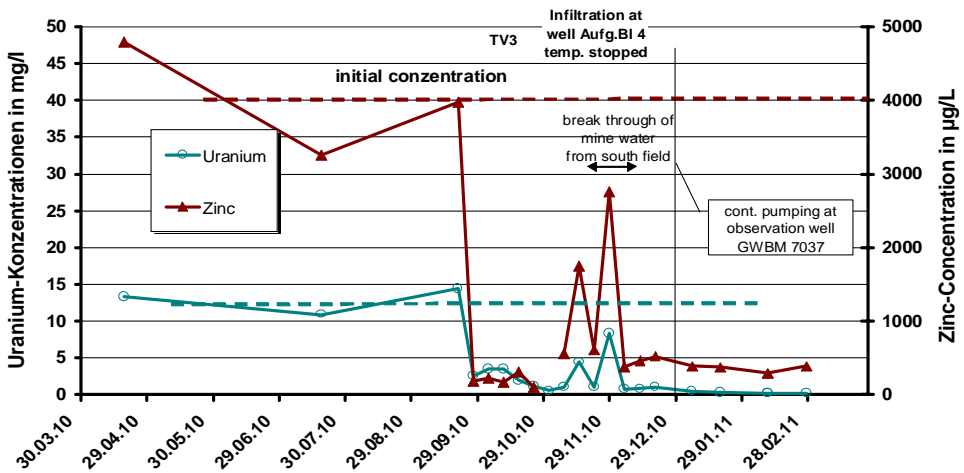


Figure 4 TV3: Concentration of Zinc and Uranium at monitoring point GWMP 7037

tration at extraction well B decreased from 5 mg/L to 3 mg/L. At an average discharge of 220 m³/d this implies a load reduction of ca. 8 kg/d.

There is fundamental concurrence between the findings of the field experiment and those of the accompanying column test. In the column, substantial quantities of sludge have collected, iron hydroxide sludge for the most part with some aluminosilicates (Si₁ Al_{0,46} K_{0,059}). This sludge contains significant levels of uranium and other heavy metals.

These findings are based on a data set established by March 2011. By then, concentration plots had not faded away, i.e. injection effects are going on.

By and large, the magnitude of the anticipated reaction spaces was confirmed. The total flood water volume within the reaction space (pore water and water in drifts, respectively) had been estimated at 260,000 m³, and that total was supplied with 46.3 t KOH and 71.6 t NaOH. This yields a dosage of ca. 10 mol of alkalinity per m³ flood water.

When scaling up the above-mentioned parameters to the mine as a whole one has to bear in mind that given the multitude of man-made per-

forations such as drifts or boreholes, flows through the mine will predominantly follow these preferential flow paths. For this reason, only part of the mine workings is directly subject to flood water flow. Contribution of the other mine voids is indirect, by density-driven contaminant release, on a much long time scale, though. This has to be taken into account for potential full-scale applications.

All in all, it may be said that continued process engineering will be rewarding as a supporting measure for the final flooding of the Königstein mine.

Acknowledgements

Significant contributions to scientific and planning preparations, implementation and interpretation of findings were made by GIP GmbH, Meraner Str. 10, D-01217 Dresden, Germany; IWSÖ GmbH, Coudray-Str. 4, D-99423 Weimar, Germany. Furthermore, the authors extend their gratitude to the staff of the Königstein Operations of Wismut GmbH for on-site implementation of the field experiment.

