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TECHNICAL ARTICLE

Field Test to Restore Original Geochemical Conditions in a Flooded Mine Area-An Essential Milestone for the Complete Remediation of the Königstein Uranium Mine

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Abstract

Uranium was extracted in the Königstein uranium mine by in-situ leaching with sulfuric acid until 1990. The originally anoxic conditions in the ore body became oxic due to the leaching process and the dewatering of the mine workings. Under these conditions, acid and metals continue to be mobilized. A field test was performed on how to restore reductive conditions in an area of the mine workings with low flow velocities. The aim was to test the immobilisation of contaminants as a prerequisite for further flooding of the mine. The field test showed that it is possible to re-establish and maintain rather reductive geochemical in-situ-conditions, which is necessary to precipitate sulphides as well as reduce acidity and metal concentrations.

Keywords Uranium mining · Mine closure · Sulphate reduction · In situ remediation

Introduction

The Königstein mine was developed in a uranium deposit, which is hosted in four stratigraphic levels of Cretaceous sandstones. The uranium deposit is distributed irregularly and is linked in the main to fine-grained sand layers that include carbonized organic substances next to sandstones with higher permeability. The host rocks are located in the lowermost of four aquifers. It is separated from the overlying 3rd aquifer by slightly permeable silty limestones and claystone.

The deposit was discovered in the early sixties. The underground mine was under production from 1967 until 1990. The mine extended over an area of ≈ 6 km^2 (Fig. 1) at a depth of 200 m below ground level (b.g.l). in the south and 300 m b.g.l. in the north.

12,250 t of uranium were mined from 1967 to 1983 by conventional room and pillar mining. Another 5750 t of uranium were extracted by in-situ leaching between 1984 and 1990 using sulfuric acid. The infiltrated leaching solution and the acid mine drainage lowered the pH, increased the redox potential, and thus changed the hydrogeochemical conditions in the effected 4th aquifer completely. Injecting sulfuric acid into the underground triggered the processes shown in Fig. 2, which are thought to be responsible for the present mine water quality:

- Increased concentrations of dissolved oxygen accelerated pyrite weathering. This contributed to the release of iron and sulphate. Due to the absence of buffering minerals like calcite, the pH decreased.
- pH values between 2.5 and 3.5 increased the weathering rates of mafic minerals, feldspars, and clay minerals and thus enhanced the release of various metals.
- Under acidic conditions, metal sulphates like jarosite and melanterite are stable (Bigham and Nordstrom 2000). They precipitated and lowered the metal and sulphate concentrations of the pore solutions. These minerals also bind acidity (which can be released again if the minerals

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Fig. 1 Location of the Königstein uranium mine

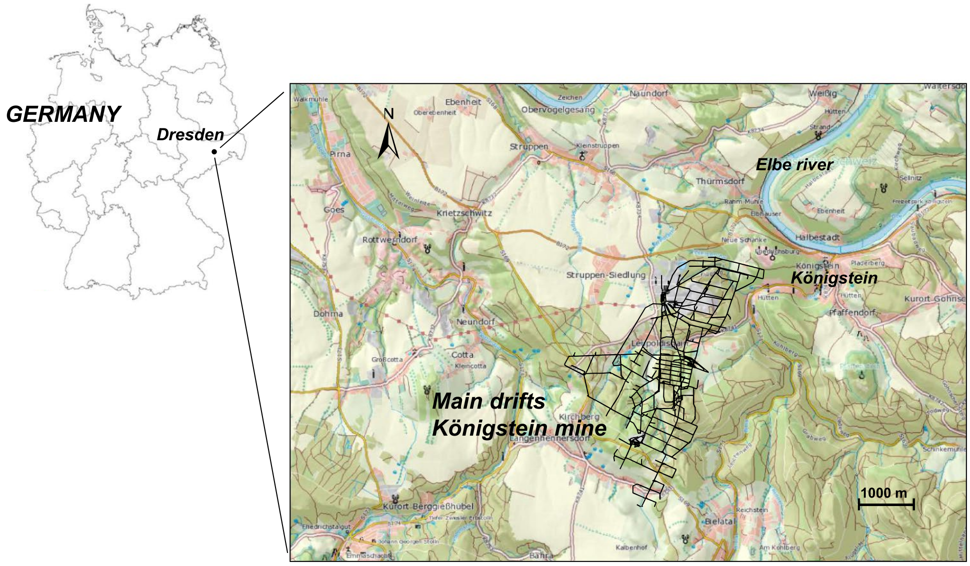
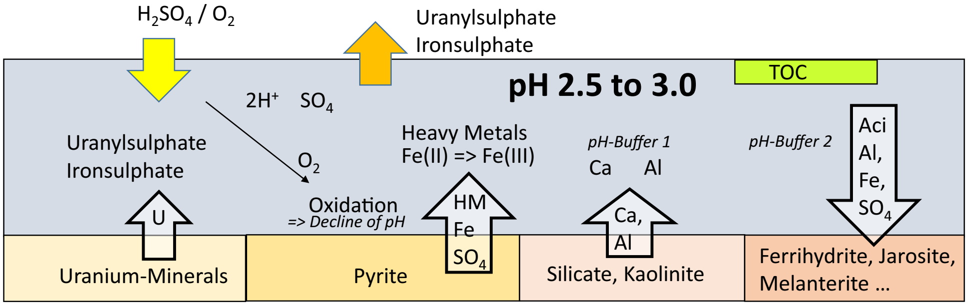


Fig. 2 Geochemical reactions in the mine, which still control the mine water quality



become unstable and redissolve). These minerals thus serve as a buffer for the metals and sulphate.

- Iron, derived mainly from the oxidizing sulphides, was precipitated as hydroxides, which are enriched with sulphate and adsorb other metals.
- Reductive microbial processes take place in the mine but are limited by the low availability of organic material.

Due to these processes, the present quality of the mine water:

- Is dominated by iron, aluminium, and sulphate;
- Shows high Ca and Mg concentrations compared to background concentrations of undisturbed adjacent aquifers;
- Shows high uranium, arsenic, and zinc concentrations;
- Shows pH values as low as pH 3.5, which indicate a lack of carbonate buffers in the aquatic phase; and
- Has a redox potential that is controlled by the Fe(II)/Fe(III) system and redox values that are mildly anoxic.

Remediation Concept and Current Status of the Königstein Uranium Mine

Remediation of the mine has been in progress since 1991. It is the overall intention to completely flood the mine and to re-establish a near-natural groundwater flow regime. The rehabilitation of the Königstein mine had to address both the specific boundary conditions in terms of the mining method and the requirements of a densely populated region within the Saxon Switzerland (a protected landscape area).

Flooding of the Königstein mine was preceded by 10 years of comprehensive preliminary studies, multi-year flooding experiments, and consideration of several remediation alternatives (Jenk and Luckner 1998; Jenk et al. 2003, 2006, 2014a; Merkel and Helling 1998). As a result of this intensive preliminary scientific work, permits to proceed with the flooding of the mine up to a water level of 140 m above sea level (a.s.l.) were issued in January 2001. The maximum allowable level was established based on predictions that excluded mine water discharges into the overlying 3rd aquifer (Jenk et al. 2014b).

At the beginning of flooding in 2001, mine water qualities varied heavily from location to location with rapidly increasing contaminant levels at low pH values. With progressive flooding, concentrations in the flooded area levelled out and contaminant levels peaked in 2002 (sulphate 3800 mg/L, zinc 56 mg/L, uranium ≈ 260 mg/L). As flooding continued, contaminant levels declined. The concentration gradient followed a typical asymptotic wash-out curve with more contaminants being delivered from the pore space of the sandstones in the mine. According to model-based predictions, further decline of contaminant concentrations down to discharge limits will continue for decades.

Until now, mobile substances and the remnants of the sulfuric acid are being dissolved in the groundwater. To prevent spreading of these contaminants to the downstream groundwater (especially aquifers 3 and 4) and surface waters, a depression cone is being maintained by constant pumping of the mine water. This was made possible by the establishment of a system of control drifts (downstream horizontal wells) with two pumping wells and a safety pillar between the control drift and the flooded mine workings (Fig. 3). The contaminated mine water is collected and treated in a treatment facility. Iron and uranium are removed in the treatment plant by neutralizing the water with lime. The purified water is then discharged into the Elbe River.

However, if the mine water level is maintained at < 140 m a.s.l. it will be impossible to achieve the overriding public interest of a sustainable rehabilitation and restoring pre-existing groundwater flow conditions. While a controlled complete flooding would admittedly involve the hydraulically inevitable local contamination of the 3rd and 4th aquifer, the gradual and controlled rise of the mine water level in conjunction with contaminant fixation in the mine should considerably minimise the spread of contaminants from the former mine.

Supporting the Flooding Process–In situ Contaminant Fixation by Injection of Reactive Solutions

In the long term, the goal is to re-establish a self-regulating water balance in the mine area and to stop pumping of mine water. The full restoration of the natural groundwater flux is considered to be a prerequisite for the complete rehabilitation of the mine site. To achieve this, dissolved contaminants such as iron, uranium and other metals must be immobilized, e.g. by precipitating them as sulphides. Furthermore, their redissolution must be prevented.

As a result of five years of intensive consultation with licensing authorities, further stepwise flooding (in ≈ 10 m steps) up to the natural groundwater level, supported by a measure for in situ neutralization and contaminant precipitation, was accepted. A first experimental step for in situ remediation, accompanied by a supporting monitoring campaign, has been agreed to. We intend to accelerate the remediation process by reducing the acid potential and supporting the geochemical fixation of contaminants in their source region by injecting reactive solutions. Long-term bench scale tests using solids and water from the mine to immobilize contaminants by increasing the pH and lowering the redox potential by injection of alkaline and organic solutions have been performed from 2013 to 2017 (Bilek et al. 2019). Dissolved contaminants such as iron, uranium and other metals were immobilized in iron hydroxides or precipitated as sulphides.

Based on these small-scale investigations, a field test at a monitoring well placed in the mine was planned and commissioned by Wismut. GmbH. Detailed planning and performance were realized by GIP/GFI Dresden as a periodic pull–push test where mine water was enriched with butanol and sodium hydroxide in a mine sector with low flow velocities. The main goal of this test was to answer these questions:

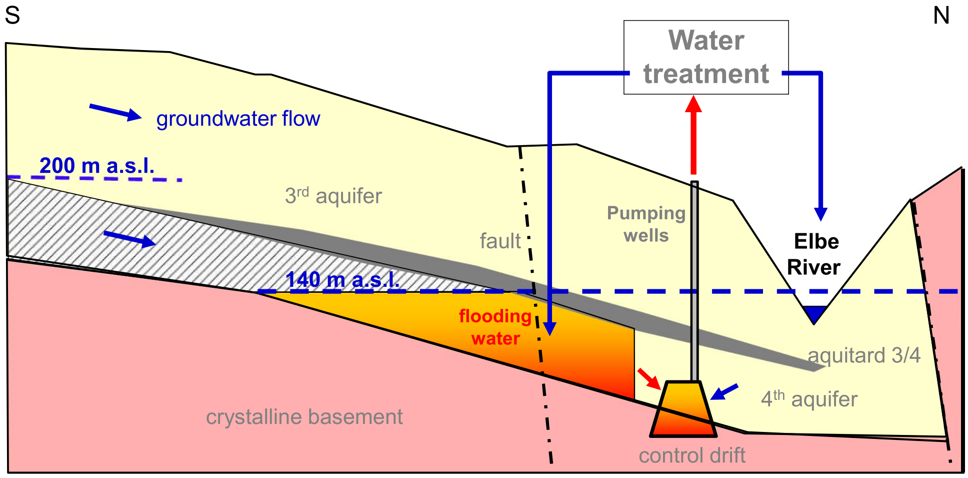


Fig. 3 Schematic representing mine water management by means of flooded control drifts and pumping wells, status 2022

- Is it possible to establish and maintain the favourable chemical conditions generated during the long-term bench-scale tests on the mine scale?
- What effects are to be expected if the hydrogeochemical environment in the oxidized mine workings is significantly changed?
- How will the secondary minerals, formed over decades under oxic conditions in the mine, react to the sudden change in geochemical conditions?

Hydrogeochemical Basics

Butanol readily serves as a substrate for microbial sulphate reduction (SR) (Giese and Hache 2016). SR works best under neutral pH conditions (Widdel 1988) and in the complete absence of oxygen (Kaksonen and Puhakka 2007). Nevertheless, it can be initiated under slightly acidic to neutral pH-values (pH 3 to 7) and even at a low partial pressure of oxygen in the bulk water phase (Elliot et al. 1998). This is possible because the microorganisms that perform SR can create micro-niches with favourable geochemical conditions (Jong and Parry 2006), especially in porous media where transport of oxygen is limited.

Oxidation of butanol serves to remove oxygen and thus prepares the conditions for SR. The SR (Eq. 1a–c) lowers the sulphate concentration and generates sulphide, which forms solids with iron (Eq. 2) and many other metals in the mine water by pH-dependent, purely inorganic precipitation (Peiffer 1994). These sulphidic solids can also sequester other aquatic species. A pH increase due to SR is expected only if the initial pH is low (Eq. 1a). When the pH rises above 5, FeS precipitation becomes dominant and will cause the pH rise to slow down or stop (Eq. 1b + Eq. 2). FeS is formed until Fe(II) is used up. Bicarbonate formation will start at pH of about 5 and the buffer capacity of the mine water increases. The acidity of the mine water decreases as the iron concentrations decrease and bicarbonate forms.

$$\mathrm{SO_4^{2-} + 1/3\ C_4^{(-II)}\ H_{10}O + 2\ H^+ \rightarrow H_2S + 4/3\ H_2CO_3 + 1/3\ H_2O} \quad \text{(acidic conditions : pH < 6.4)} \tag{1a}$$

$$\mathrm{SO_4^{2-} + 1/3\ C_4^{(-II)}H_{10}O + 2/3\ H^+ \rightarrow H_2S + 4/3\ HCO_3^- + 1/3\ H_2O} \quad \text{(slightly acidic conditions : pH < 6.4 – 7.0)} \tag{1b}$$

$$\mathrm{SO_4^{2-} + 1/3\ C_4^{(-II)}H_{10}O \rightarrow HS^- + 1/3\ H^+ + 4/3\ HCO_3^- + 1/3\ H_2O} \quad \text{(circumneutral conditions : pH > 7.0)} \tag{1c}$$

$$\mathrm{Fe^{2+} + HS^- \rightarrow FeS_{(s)} + H^+} \tag{2}$$

Execution of the Field-Test

To investigate the effects of SR under field conditions, a seven month field test was planned and performed between fall 2020 and summer 2021 (Fig. 4). The mine

Fig. 4 Experimental plant–view from air and inside the container

workings of Königstein mine can only be sampled in monitoring wells with filter screens about 200 m b.g.l. and about 60 m below the groundwater table. Due to the large distances between the monitoring wells, it was decided to perform a pull–push test using a single bore hole. This means that all reactions that might be initiated in the mine water would have to be detected by sampling a single well. It was thus necessary to select a monitoring well that is filtered in a zone of low flow velocity to avoid drifting out of the "reaction zone" too fast. By performing a couple of infiltration and tracer tests, one sampling monitoring well was selected to perform the test. The test itself consisted of these steps:

1. 9 m^3 of mine water were pumped within two hours and stored in a tank next to the monitoring well. The pumped water was sampled after about 5 m^3 were pumped. The pH, c(Fe(II)), and c(K) were analysed repeatedly during the pumping process to get information about the concentration distribution in the reaction zone around the filter screen. While pumping, butanol was evenly mixed into the mine water.
2. 1 m^3 was pumped without spiking it with butanol. This volume was stored separately.
3. Immediately afterwards, the butanol-enriched mine water was reinjected. A DOC concentration of 1650 to 1800 mg/L was measured in the mine water immediately before reinjection. Four times during the whole test, KCl was mixed as a tracer into the mine water during reinjection. The initial K concentration in the KCl-spiked mine water ranged between 200 and 250 mg/L.
4. 1 m^3 of the untreated mine water was reinjected to push the butanol-enriched mine water away from the well screen and the well filter zone, into the aquifer around the well. This was done to prevent precipitation processes, which might have clogged the well screen.
5. After complete reinjection of the 9 + 1 m^3 of reactive fluid, a "reaction zone" was established around the well screen.
6. During a reaction period of 2 weeks, no pumping occurred. Various reactions and mixing/dilution processes were expected to take place in the underground reaction zone.
7. Steps 1 to 6 were repeated for 217 days. In total, 17 pull-and-push-cycles were performed.

Peristaltic pumps were used to dose the exact volumes of butanol and KCl solution (Fig. 5). The pumps and other technical devises were installed in a container to avoid freezing. The pumped mine water was temporarily stored at the surface in two glass fibre-reinforced plastic tanks.

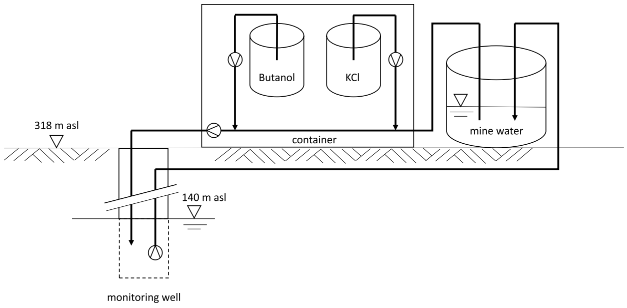


Fig. 5 Scheme of the experimental plant

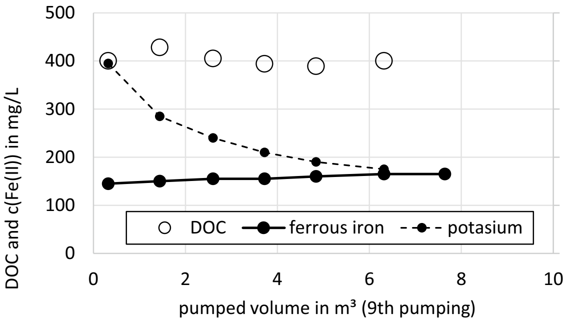


Fig. 6 Change of c(DOC), c(Fe(II)) and c(K) in the course of pumping

Results

Figure 6 shows the concentration of ferrous iron, potassium (tracer), and DOC during the 9th pumping cycle, which in fact is representative of all the other pumping cycles as well. The data basically shows that there was nearly no change in the concentration of these three parameters during the pumping process. This shows that:

(a) The reaction zone was of a rather homogeneous water quality,

(b) The reaction zone had a higher volume than the 10 m^3 that were pumped (otherwise, the concentrations would have approached the background concentration at the end of the pumping process). Only a slight decrease in the K concentration hints at the decline of the concentrations at the outer part of the reaction zone. The background concentrations of 18–20 mg_K/L were not reached.

(c) No significant drift of the DOC-spiked water body away from the well screen occurred (otherwise, the first samples would show concentrations closer to the background concentrations of about 5 mg_{DOC}/L due to the offset of the spiked water body and its replacement by original mine water).

(d) The Fe concentration increased slightly during the pumping process. This is interpreted as being due to an iron-releasing process, which might be more active in the iron-rich pore space of the adjacent aquifer.

Figure 7 shows the concentration of chloride, which is used as tracer. Its background concentration of about 45 mg/L was raised five times to 200 to 250 mg/L by adding KCl and then diluted by the groundwater flow regime in the vicinity of the well screen. Its remaining concentration after the first two weeks was about 80 to 100 mg/L and then slowly approached the background level within the next 8 weeks. The background level was never reached in this time. After each tracer injection, the concentration level in the reaction zone remained higher than before.

Figure 7 also shows the development of the sulphate concentration during the field test. After about 30 days, the sulphate concentration (background concentration of about 1500 mg/L) started to drop. The sulphate concentration decline was fastest after 50 days; after that, it levelled off, until the concentration reached about 200 mg/L after 170 days.

Figure 8 shows the concentrations of measured and modelled DOC (see also Discussion). While the organic carbon concentration was about 5 mg/L in the original mine water, a constant DOC concentration of about 400 to 500 mg/L was measured each time after a 2 week period during the tests. This concentration is the result:

- Of a mixing/dilution process of 9 m^3 of DOC spiked water (1500–1900 mg/L) with the remaining water in the reaction zone;
- The drifting off and replacement of the volume by the original mine water; and
- The oxidation of butanol by sulphate reduction

These values show that there was always enough reductive substrate in the reaction zone present to support SR and that about two thirds of the butanol was needed to replace losses due to the mixing/dilution processes and drifting off (see also Fig. 8).

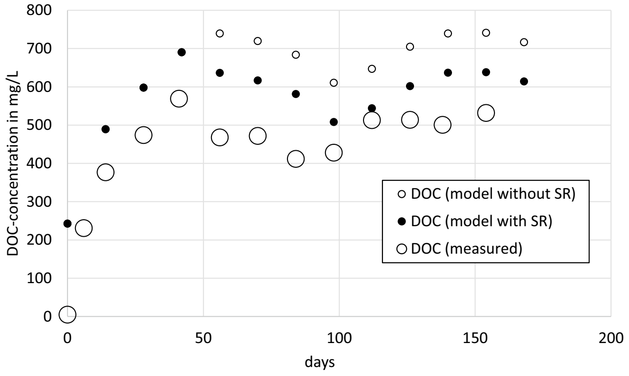


Fig. 8 Measured and modelled c(DOC)

Figure 9 shows the sulphide and oxygen concentrations and the pH. Sulphide, which was just above the detection limit (0.01 mg/L), with concentrations between 0.02 and 0.05 mg/L, rose to 6 mg/L at day 70 shortly after initiation of the SR. From then on, a decline to about 2 mg/L was observed. The c(O_2) was constantly below 2 mg/L during the field test. This remaining oxygen concentration was considered a remnant of the pumping process and apparently did not impair the microbial reduction process. The pH stayed constantly between 5.7 and 6.0 throughout the test. A slight depression of the pH was observed during the period of fast c(SO_4) decline from day 41 to 98.

Figure 10 shows the concentrations of Fe(II), TIC, Ni, and Co measured in the mine water during the test. The c(Fe(II)) stayed at its background level of about 230 mg/L until SR started. From then on, it dropped below 150 mg/l at day 84 and then rose slowly until it reached 180 mg/L again. The c(TIC) stayed at ≈ 70 mg/L (background-concentration) and started to rise from day 41 on to > 100 mg/L. The c(Co) and c(Ni) showed a decline in concentration from about 50 to 25 µg/L and 90 to 50 µg/L, respectively.

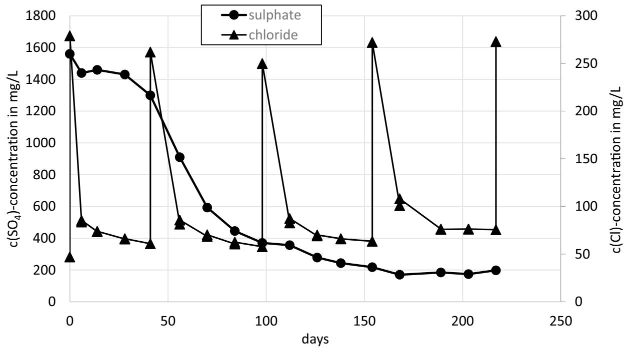


Fig. 7 Sulphate and chloride concentration in the course of the field test

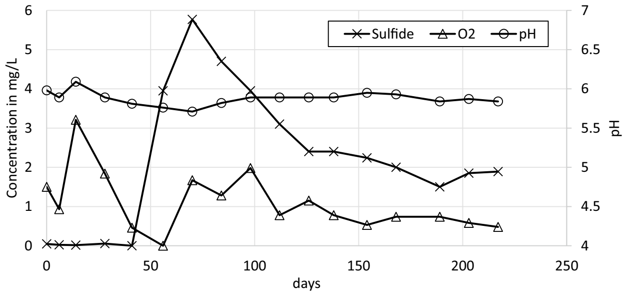


Fig. 9 Sulphide and O_2 concentration as well as pH in the course of the field test

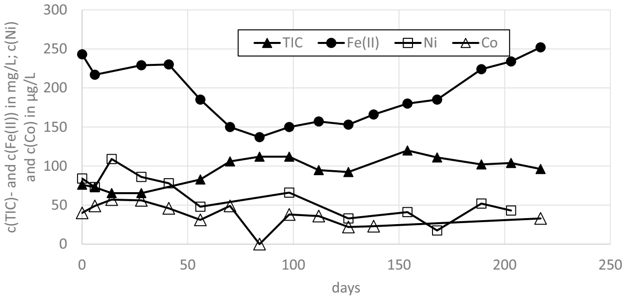


Fig. 10 c(TIC), c(Fe(II)), c(Co) and c(Ni) in the course of the field test

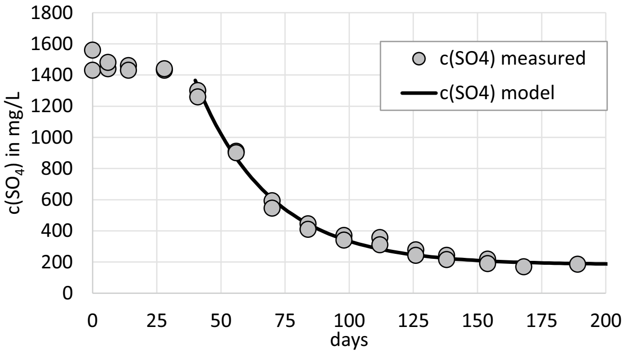


Fig. 12 Measured and modelled sulphate concentration decline with the mixing model taking into account SR

Discussion

Mixing Model

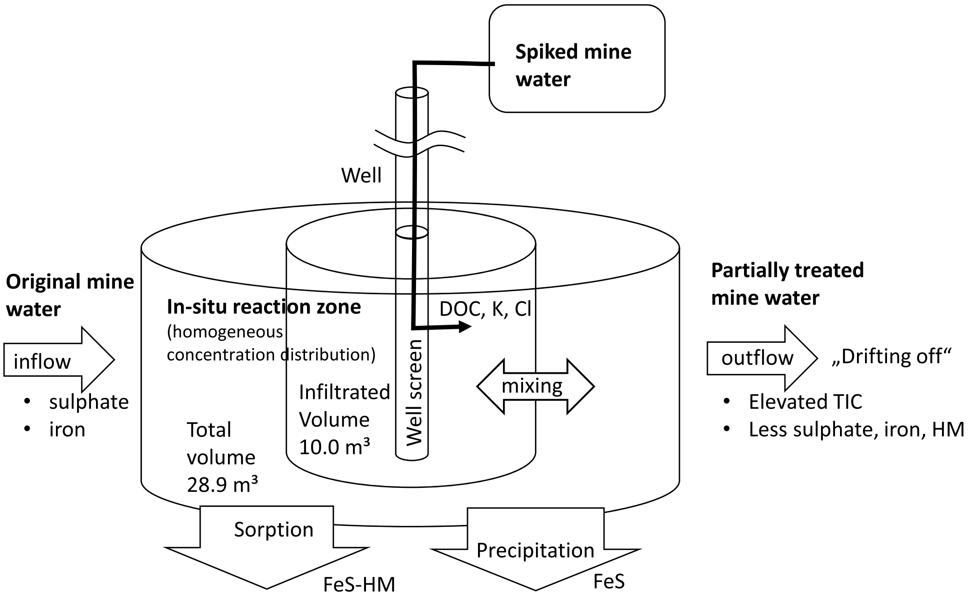
A mixing model was set up for the in-situ reaction zone to better understand the observed concentration changes. The spiked volume of mine water did not behave like a plume, which constantly drifts away from the well. Instead, the concentration development during the pumping phases is best explained by a 'mixing model' (Fig. 11), in which the water volume, which is subject to a changed water quality (increase in DOC, K …), amounts to significantly more than 10 m^3.

The concentrations of DOC and sulphate were calculated with this model and compared in Fig. 8 and Fig. 12 with the measured values. To fit modelled and measured data, a) the volume of the reaction zone, b) the mine water flux "inflow" with background concentration into the in-situ-reaction zone, and c) the sulphate reduction rate, were all used as fitting parameters. The curve fitting provides the following estimates:

(a) The reaction volume in the vicinity of the well was 28.9 m^3;
(b) The inflow of original mine water from "outside" was ≈ 0.04 m^3/h. The same amount of water leaves the reaction zone at the same time; and.
(c) The sulphate reduction rate was ≈ 45 mg/(L d).

The measured data can be explained without assuming a decline of SR with time. This would also explain the c(TIC) as an indicator of ongoing butanol oxidation, which stays elevated until the end of the test. According to this model, the slowing down of sulphate decline and its levelling off is purely a result of a mixing process.

Fig. 11 Mixing model of the in-situ-reaction zone according to the measured timelines of the concentrations



This also holds for the DOC concentration in the reaction zone, which levels off because of the dilution process. Nevertheless, comparing the measured data and the model data suggest that the beginning of SR contributed to the levelling off of the DOC.

Sulphate Reduction and Subsequent Reactions

Trace concentrations of sulphide in the initial mine water, only slightly acidic pH values of about 5, and a declining sulphate concentration in the last years made it likely that sulphate-reducing bacteria (SRB) were already active in the vicinity of the monitoring well. The initial 40 days with no significant change in water quality (except for a rise of DOC) are interpreted as a lag phase, which was necessary for the adaption and growth of the in situ SRB population.

The development of the presented parameters after day 50 is interpreted as the successful initiation of SR in that part of the Königstein mine. The most visible result of SR is the strong decline in sulphate concentration (Eq. 1a–c), which levels off and asymptotically approaches a concentration of ≈ 200 mg/L.

According to the mixing model, microbial sulphate reduction and the mixing of the original mine water with the treated mine water explain the observed decline of the sulphate concentration. At the same time, c(TIC) rose, probably due to the oxidation of butanol (Eq. 1a–c).

The decline in the concentration of iron is also considered to be the direct consequence of sulphide generation and its precipitation with iron (Eq. 2). However, comparing the rate of iron decline with the rate of sulphate reduction, it is obvious that precipitation of aqueous Fe(II) is not the only sink for sulphide unless one assumes a constant release of Fe(II) from Fe(III) hydroxides, which might be present in the mining-affected environment within the mine. Iron(III) hydroxides were likely there due to the oxic and suboxic conditions, which had been prevailing there for many decades. Re-establishing a reductive milieu might have the consequence of redissolving iron hydroxides (Eq. 3) and causing the pH to rise.

$$C_4H_{10}O + 24\,Fe(OH)_{3(s)} + 48\,H^+ \rightarrow 24\,Fe^{2+} + 4\,CO_2 + 65\,H_2O \quad (3)$$

Also, the reaction of aqueous sulphide with the precipitated iron hydroxides (Eq. 4) is described to be a dominant sink for sulphide (dos Santos Afonso and Stumm 1992; Pyzik and Sommer 1981).

$$2\,FeOOH + 3\,HS^- \Rightarrow 2\,FeS + S^0 + 3\,OH^- + H_2O \quad (4)$$

It is assumed that all three processes are responsible for the strong control of sulphide concentrations. Basically, all the sulphide that is released by SR is precipitated as iron sulphide. Also, it can be concluded that ferric hydroxides were present in the in-situ reaction zone and must be considered when estimating the amount of reactive material in the aquifer.

The pH of the mine water at the beginning of the test was in the range of 5.8 and in the range of Eq. 1a and 1b. Thus, it can be assumed that SR in combination with FeS precipitation (Eq. 2) did not have a strong effect on pH. The rather stable pH could have been a result of a combination of the Eq. 1 and 2. Reductive redissolution of ferric hydroxides according to Eq. 3 could contribute to a pH increase, but the extent of this reaction is unknown.

In summary, all the parameters suggest that SR was intensified successfully in the underground reaction zone. The concentrations of some of the metals, such as Ni and Co, were reduced during the field test. Other metals like U did not yet show a significant decline. There was an increase in the Al concentrations, which is thought to be due to the reductive redissolution of iron hydroxides. Al is not redox sensitive, but it might have been included in the iron hydroxide sludge present in the mine workings. Also, c(Ba) rose, probably due to SR, which consumes sulphate. Ba is known to been precipitated with sulphate as barite in the Königstein mine (Jenk and Ziegenbalg 2006). Therefore, redissolution of sulphates by SR might have liberated sulphate as well as some Ba.

The elevated sulphide concentration until the end of the test implies that SR continued as long as there was butanol infiltration. The sulphate reduction rate of 45 mg/(L d), or 0.02 mmol/(L h), is comparable to other SR rates measured in in-situ reaction zones spiked with easily available organic carbon (Walko and Hildmann 2018).

Conclusions

The conception, technical implementation, and realisation of the hydrochemical underground test was a success. Clearly, sulphate reduction was initiated and kept active for many months in the monitored portion of the Königstein mine. We consider that this process will help re-establish near natural geochemical conditions underground. The scope of remediation focuses on reducing the concentrations of sulphate, uranium, and the many other mobilized metals in the mine water outflow by retention in the mine under favourable geochemical conditions.

The test showed that it is possible to control such an in-situ reaction zone from above ground. It was also demonstrated that the concentrations of contaminants will not be quickly and easily forced below detection limits, although precipitation and absorption of the contaminants in newly formed minerals is expected. The results showed that there were large amounts of substances that had precipitated under anoxic conditions. Some portion of these phases will redissolve and eventually form other phases that will be more stable in the future prevailing reductive conditions. Meanwhile, they will be detectable in limited concentrations in the aquatic phase.

The results of the test have been an essential basis of the application documents for approval of the next step, flooding the Königstein mine up to 150 m a.s.l. Permission was granted in November 2022. The start of the first full-scale application of reductive substances for a time period of ≈3 years is planned for 2023.

Data availability The data was collected as part of the field test described in the paper on behalf of Wismut GmbH. The data are the property of Wismut GmbH. Inquiries in this regard can be directed to the company.

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