

## In Situ Aquifer Treatment by Microbial Sulfate Reduction

Martin Gast<sup>1</sup>, Ralph Schöpke<sup>2</sup>, Manja Walko<sup>1</sup>, Friedrich Carl Benthaus<sup>3</sup>

<sup>1</sup>Research Institute for Post-Mining Landscapes (FIB e.V.), PF 1215, 03231 Finsterwalde, Germany, m.gast@fib-ev.de

<sup>2</sup>Technical University Cottbus (BTU), Chair in Water Technology & Engineering, PF 10 13 44, 03013 Cottbus, Germany, schoepke@tu-cottbus.de

<sup>3</sup>Lusatian and Middle German Mining Management (LMBV mbH), Knappenstraße 1, 01968 Senftenberg, Germany, fc.benthaus@lmbv.de

### Abstract

In order to reduce acidic impact to the numerously emerging mining lakes in the Lusatian Lignite District (NE Germany) a pilot system for in situ aquifer treatment was installed and operated. To promote the microbial driven sulfate reduction, biodegradable organic matter was infiltrated by injection lances into an aquifer of a dump. Thereby, a space in the aquifer was created with significant reduction of sulfate, iron and potential acidity. The infiltrated glycerine was completely metabolized. This produced a carbon dioxide input to the water and generated an additional buffering capacity.

**Key words:** groundwater, remediation, pyrite oxidation, glycerine, sulfide, precipitation, injection lance

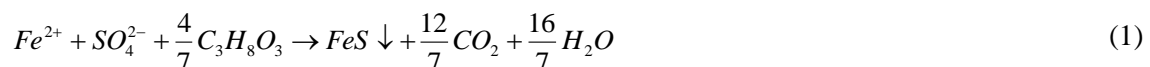
### Introduction

Actually, in Lusatia the largest man made lake district in Europe emerges as a result of lignite mining. The projected expanse of water will be about 140 km<sup>2</sup> in total (Luckner et al. 2003). Caused by pyrite oxidation, water quality is characterized by extreme acidity combined with strong salinity. To keep within legal standards and to utilize the water bodies, technical treatment is indispensable.

Based on previous studies, the enhancement of the microbial sulfate reduction is seen as the most promising way to improve groundwater quality in the upstream as well as in the downstream of the lakes (e.g. Schöpke 2010, Koch et al. 2006, Hoth et al. 2005, 2001). Therefore, the Research Institute for Post-Mining Landscapes (FIB e.V.) together with the Technical University of Cottbus (BTU) designed a test system for the in situ treatment of aquifers influenced by pyrite oxidation.

Main target of this system is to enhance biochemical reduction of sulfate in order to precipitate iron-sulphides in the aquifer. This should prevent the transport, respectively the input of sulfate and acid generating iron to lakes and other receiving waters.

By application of glycerine as a biodegradable organic matter, the striven remediation reaction can be described as:



For the conditions in the Lusatian Mining district the “Neutralisation Potential” (NP) can be used to quantify the acidification capacity of water affected by pyrite oxidation (Schöpke 2010):

$$NP = ANC_{4,3} - 2 \cdot c_{Fe2} - 3 \cdot c_{Al} - 2 \cdot c_{Mn} \quad (2)$$

The Parameter NP reflects the influence of the released CO<sub>2</sub> as well as the effects on the Fe concentrations due to sulfide precipitation. A decrease of the NP points out the aquifer treatment to be successful.

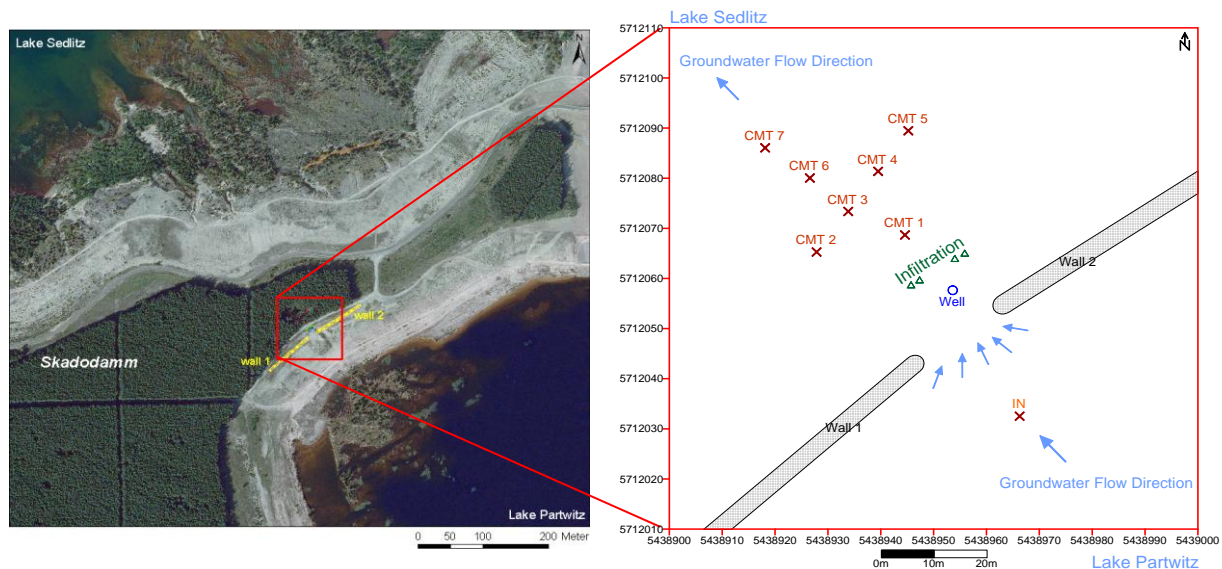
### Site Conditions

The pilot system is situated on the “Skadodamm”, a spoil heap respectively a dam between two mining lakes (Figure 1). Because of its higher charging level, the water of the Lake Partwitz passes through the dam into the Lake Sedlitz.

Hydraulic conductivities of about  $k_f \approx 1 \cdot 10^{-5}$  m/s (light loamy sand) and a thickness of 13 m are good conditions for in-situ treatment. Before infiltration, the sediments are free from lignite, iron sulfides as well as nitrogen and phosphorous.

The groundwater quality is determined at first from the infiltrated lake water. At second, the upper groundwater is influenced by the input of oxidized and acidic leachate from the overlying sediments. The main part of the water to be treated (intake) is characterized by pH values about 4.9 - 5.1,  $\text{Fe}^{2+}$  concentrations about 232 - 322 mg/L and  $\text{SO}_4^{2-}$  concentrations about 956 - 1152 mg/L.

As a result of a preliminary project, two walls built out of brown coal fly ash remain in the underground. With a length of 75 m each and a height of about 14 m, they represent a “funnel and gate” situation that was embedded in the pilot system and allowed the treatment of a concentrated groundwater stream.



**Figure 1:** Top view on the pilot system at the “Skadodamm”; CMT = multilevel groundwater monitoring systems .

### Design concept

In the centre between two underground walls there is a permeable gate of about 12 m width and 13 m height. Within this gate a well hoists a part of the through passing water to the surface (Figure 2). There, glycerine as well as nitrogen and phosphorous fertilizer are added by the use of an automatic dosage system. Under the requirement of minimizing oxidizing influences, this mixture is directly infiltrated back into the aquifer by four injection lances (DSI-Lances, Wils & Water GmbH, Germany). Infiltration takes place in depths of 15 m respectively 19 m under ground surface with rates in a range of 0.8 to 1.8  $\text{m}^3/\text{h}$ . This affords a water pressure of about 29 psi measured at ground surface. Inner diameter of the lances is 32 mm. The lances are charged singular with an automatically driven alternation every hour.

For the groundwater monitoring, seven CMT-Systems (Solinst Canada Ltd.) were installed (Figure 1). Every CMT-System allows the sampling of water in depths of 12, 16 and 20 m under ground surface (Figure 2). Sampling is done every 2 respectively 4 weeks.

Dosage of the glycerine is based on the above given remediation equation (1). At the beginning, the added on glycerine was calculated for a  $\Delta\text{NP} = 9 \text{ mmol/L}$ . This equates to a glycerine concentration of 2.6 mmol/L in the infiltration water. During the operation of the system, concentrations varied a few times and was risen up to 7.5 mmol/L. In relation to the C input, nitrogen was given in a ratio of  $\text{C/N} = 25$  and phosphorous in a ratio of  $\text{C/P} = 400$ . These ratios were derived from the estimated chemical structure of the microbial biomass.

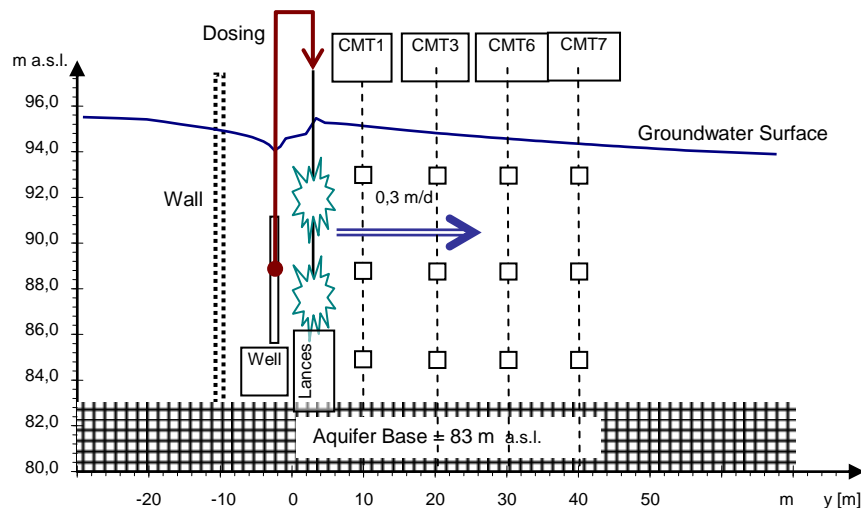


Figure 2 Longitudinal cut through the underground reactor along the flow path.

## Results

Within an operation period of 400 days, the formation of the groundwater in the downstream of the glycerine infiltration clearly reflects an enhanced sulfate reduction. Sulfate concentrations decreased in a ratio of  $> 0.73$  (Table 1). Simultaneously, iron was removed completely. Also, the increase of solute sulfide approves the process of sulfate reduction. The appearance of higher sulfide concentrations points to a lack of solute iron to be precipitated as  $\text{FeS}$ . To prevent problems associated with outgassing of  $\text{H}_2\text{S}$ , the system has to be run with reduced input of organic matter and with pronounced infiltration breaks. This contributes to a sufficient iron delivery for the striven sulfide precipitation.

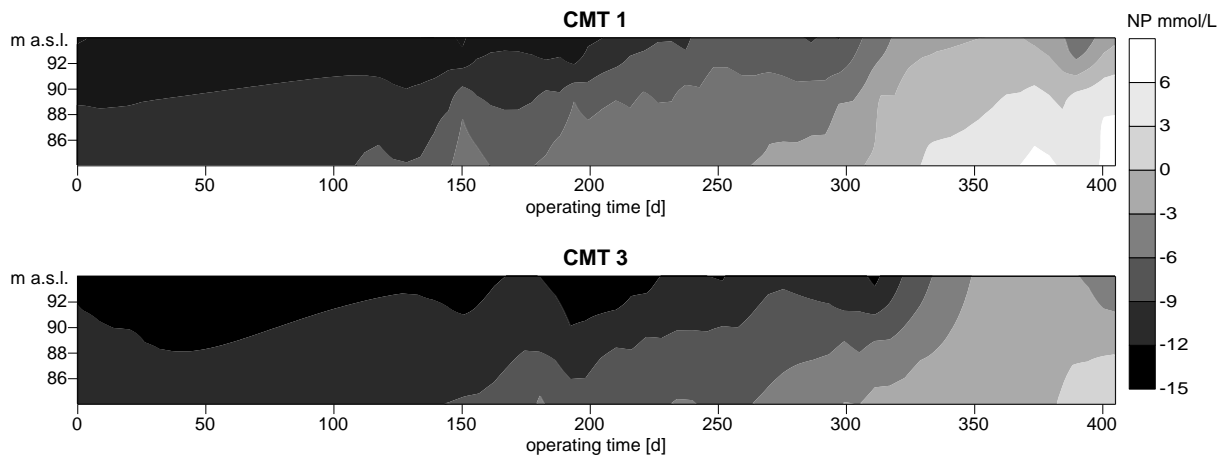
Table 1: Comparison of water quality in the untreated intake with the downstream at different monitoring points after 400 days operating time.

| Parameter             |        | Intake | CMT 1 | CMT 3 | CMT 6 | CMT 7 |
|-----------------------|--------|--------|-------|-------|-------|-------|
| Distance <sup>1</sup> | m      | - 15   | 9     | 19    | 29    | 39    |
| pH                    |        | 4.9    | 6.2   | 5.5   | 5.6   | 5.5   |
| NP                    | mmol/L | - 10   | + 7   | + 0.5 | + 0.1 | 0     |
| Fe                    | mg/L   | 290    | 1     | 50    | 42    | 41    |
| $\text{SO}_4$         | mg/L   | 1100   | < 100 | 150   | < 100 | < 100 |
| TIC                   | mg/L   | 46     | 190   | 134   | 135   | 119   |
| Sulfide-S             | mg/L   | < 0.1  | 86    | 20    | 10    | 8     |

<sup>1</sup> from infiltration

In consequence of the microbial decomposition of the organic matter, TIC concentrations rise four times higher compared to the untreated intake of the system. Especially the decrease of iron concentrations in connection with the TIC increase led to a distinct rise of the NP. The initial value of  $\text{NP} = - 10$  mmol/L improved to  $+ 7$  mmol/L. This means that the groundwater was changed from an acidic quality to a significant buffering quality. It can be shown that the development of the NP starts in the lower part of the aquifer (Figure 3). Probably, this part of the aquifer is more reductive in comparison to the upper aquifer.

During 400 d of operation the cumulative glycerine input is 1.8 t. The input of nitrogen and phosphorous is about 16 kg respectively 3 kg.



**Figure 3** Chronological development of the Neutralization Potential in the downstream of the infiltration shown at a flow path of 9 m (CMT 1) and 19 m (CMT 3).

### Conclusions

Infiltration of biodegradable organic matter induces sulfate reduction in an aquifer strongly affected by pyrite oxidation. The striven process of iron sulfide precipitation is limited by solute iron. This limitation requires a permanent monitoring and modulation of the input of organic matter. Otherwise, concentrations of solute sulfide will increase and may cause problems if outgassing of  $H_2S$  takes place in a notable volume. Besides of the control of the organic matter input, an operation mode with pronounced breaks helps to mix the iron rich groundwater with the treated groundwater. The decomposition of glycerine is associated with an increase of TIC that generates a considerable buffering capacity in the groundwater.

From a scientific point of view the operation of the pilot system was successful. To balance the costs and the benefits, more data is required; especially to test different infiltration modes. For this an extension over another year of operation und monitoring is projected.

### Acknowledgements

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