# Pit Lake Treatment Using Fly Ash Deposits and Carbon Dioxide

Christian Koch, Bastian Graupner, Florian Werner

Christian Koch, ckoch@dgfz.de, www.dgfz.de, DGFZ Dresdner Grundwasserforschungszentrum e.V., Meraner Straße 10, 01217 Dresden, Germany

#### Abstract

The research project CDEAL (carbon dioxide elimination using acid mine lakes and calcium oxide suspensions) combines the attempt to rehabilitate acid mine lakes with the attempt to reduce the environmental impact of atmospheric carbon dioxide emissions from coal-fired power plants. Due to the high overall acidity content and the high acidic inflow from mining dump groundwater (AMD), remediation of most mining lakes is financially and technically challenging. CDEAL's neutralisation strategy is the production of water with an alkalinity of 5 to 10 mmol/L using CO<sub>2</sub> and alkaline material. Less water needs to be pumped and better distribution of alkaline suspension within the lake is achieved, resulting in lower treatment costs. The CO<sub>2</sub> reduction can be achieved through the generation and accumulation of carbonate in the lakes using remaining products (CO<sub>2</sub> and fly ash). Neutral pH conditions are necessary for long-term stability of carbonates.

Key words: carbon dioxide, CO<sub>2</sub>, acidic lake treatment, pilot scale experiment, fly ash

## Introduction

Water treatment of pit lakes is an important research topic due to its environmental impacts and risks for connected natural lakes and rivers. The mining Lausitz region covers several pit lakes containing alkaline deposits from former remediation attempts or from its use as disposal site for fly ashes from local power plants. these alkaline deposits are investigated In the course of the R&D project CDEAL (carbon dioxide elimination using acid mine lakes and calcium oxide suspensions). The aim of the project is an assessment of both the alkaline potential of the sediments and the feasibility to use them in industrial scale processes for water remediation and  $CO_2$  storage purposes. In this respect the study team investigated the potential for a field scale application to improve water quality of acidic lakes and simultaneously mineralise carbon dioxide using alkaline deposits within the lake water body. CDEAL was conducted from the scale of laboratory over bench to the field-test scale. In the laboratory tests sediment from the Burghammer pit lake test site were investigated. Physical as well as chemical sediment properties were determined. As a result it can be stated that the sediments can be used to neutralise the acidic lake water and to mineralise additional carbon dioxide.

A two step strategy can be proposed. Firstly the lake is to be neutralised. Secondly the mineralization of carbon dioxide will be aimed at using similar technical layouts.

## **Research object – Lake Burghammer**

The post mining Lake Burghammer was chosen as research site for the project CDEAL. Between 1970 and 1990, approximately 26 million m<sup>3</sup> of suspended fly ash, which settled as a reactive fly ash body, were deposited in Lake Burghammer. The volume of the lake is currently about 31 million. cubic metres (Mm<sup>3</sup>).

#### Two step strategy for lake water neutralisation and CO<sub>2</sub> sequestration

Due to the high acidity of Lake Burghammer (total amount about 100 million moles) and the large yearly inflowing acidity mainly from dump groundwater (16 Mio. mol/a), carbon dioxide storage remains difficult. Therefore, a first step in the carbon dioxide storage attempt needs to be the neutralisation of lake water. The alkaline fly ash deposits can be used for this purpose. After the neutralisation the remaining fly ash deposits might be used for  $CO_2$  mineralization.

#### **Pilot scale test – technology**

The pilot scale experiment was conducted to show if the developed treatment scheme is feasible and to determine the alkalinity gained from the water treatment. The  $CO_2$  mineralization step was not part of the pilot scale experiment. It can be shown that pH elevation is partly based on dissolution of calcium hydroxide. This part might be used to mineralise  $CO_2$  in the second step. A suction excavator was used for sediment uptake and mixing with lake water. The tests were scheduled for a period of two weeks.

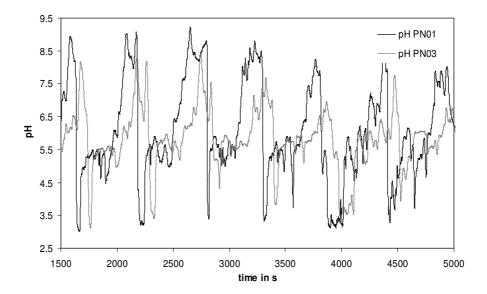
It was planned to excavate a trench to simplify sediment measurements after the tests. The discharge of the device was varied between 50 and 250 m<sup>3</sup>/h. Connected to the suction excavator a floating pipeline was installed. At the beginning of the 150 m long pipeline (reactor) carbon dioxide was dosed. The amount was varied between 5 and 50 kg/h. Overall 17 tests with varying parameters were conducted. A total volume of 75 m<sup>3</sup> or 140 t of fly ash sediment was relocated and mixed with 2500 m<sup>3</sup> lake water before being distributed within the lake water body. Carbon dioxide was stored at the lake bank. A floating gas hose of about 700 m length connected the  $CO_2$  cylinder bundles with the floating excavator pipeline.

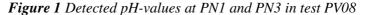
## **Process monitoring**

The process monitoring system consisted of three sampling points along the pipeline. The first one was installed before  $CO_2$  injection. The second and third sampling points in the middle and at the end of the pipeline respectively. Signals from pH and electrical conductivity probes at the last sample point (150 m away) were transmitted via a radio installation. Every test ran for 2 to 3 hours and was separated into (i) lake water pumping (ii) pumping of lake water and sediment suspension and (iii) suspension pumping with  $CO_2$ -addition. Samples were taken two times within one run at the sampling points. Analysis included major anions and cations as well as TIC and buffering capacity by titration. From several samples PACs were determined to out rule negative environmental impacts by fly ash usage.

#### Results

Figure 1 shows the recorded pH-values at PN1 and PN3 during test PV08. Depending on the test parameters PV08 serves as a good example for the course of each conducted run. The suspension was pumped at a rate of about 100 m<sup>3</sup>/h resulting in a residence time of 90 seconds within the pipeline.





Carbon dioxide dosing was set to 5 mmol/L and this caused a pH drop within the pipeline from PN1 to PN3. The suspension pH values vary during the run which can be explained by the technological approach. The sediment mining was done automatically to keep runs comparable. Due to the use of a small suction excavator the maximum achievable depth was 4 m from the water surface. Water depth of 2.8 meters limited therefore the maximum accessible sediment depth to about 1.2 m. As investigated during the conducted batch and bench scale experiments earlier in the course of the project, the first 0.5 to 0.8 m meters of the sediment is characterised by low reactivity due to high carbonate content. Carbonation of the reactive material is nearly finished in the first layer. Calcium hydroxide content of lower layers is higher and the reaction causes a pH increase from 2.9 (lake water) to values in the suspension between 7 and 9. Theoretically carbon dioxide mineralization is possible and was observed in the batch experiments. But, due to the focus of the pilot scale experiment on

water treatment, carbon dioxide was dosed to maximise the resulting buffer capacity of the suspension rather than for mineralization purposes. The buffering capacity of the treated water was increased significantly. A mean value of achievable buffering capacity is about 5 mmol/L. The maximum achieved during field tests was measured to be 9 mmol/L. This temporal effect is used when treated water is mixed with untreated water in the lake. The improved water treatment reduces the time of treatment due to the reduction of pumping effort which is directly related to  $CO_2$  emission.

# **General findings**

Due to the small amount of gypsum in fly ash sediments it was expected that sulphate might be a problem when using fly ash deposits in water treatment. It can be stated that sulphate concentrations increased slightly in the treated water. The already high sulphate as well as the elevated calcium concentrations limit the additional solution of sulphate mainly from gypsum. The maximum increase of sulphate concentration was measured to be about 11 %.

Parameter*	Lake water	treated water	comment
pН	2.9	5.3 - 9.2	strongly increased
Alkalinity	-3	0.7 - 8.6	strongly increased
TIC	0.08	0.8 - 17.5	strongly increased
Calcium	7.21	9.1 - 14.3	increased
Magnesium	2.27	2.3 - 4.1	increased
Sulphate	11.14	11.1 - 12.3	slightly increased
Manganese	0.1	0.01 - 0.09	strongly decreased
Iron (total)	0.33	0.00018 - 0.06	strongly decreased
Aluminium	0.17	0.001 - 0.02	strongly decreased
Zinc	0.005	0.00015 - 0.003	decreased
Copper	< 4 µg/L		no change
Lead	< 10 µg/L		no change
Cobalt	< 90 µg/L		no change
Cadmium	< 5 µg/L		no change
Chromium	< 6 µg/L		no change

Table 1 Comparison of quality for lake water and treated water

Note: units mg/L unless stated

Due to the pH increase metal hydroxides precipitated. Therefore, iron, aluminium and manganese concentrations were reduced in the treated water. Remaining concentrations were below 3 mg/L for iron and below 0.1 mg/L for aluminium below 0.5 mg/L for manganese. The same applies for heavy metal concentrations. If present in lake water these metals precipitated during the treatment (Cu < 4  $\mu$ g/L, Pb < 10  $\mu$ g/L, Co 90  $\mu$ g/L, Cd < 5  $\mu$ g/L, Cr 6  $\mu$ g/L). Before and after the pilot scale experiment the lake bottom was surveyed by an echo sounder which was synchronised with a GPS receiver. The expected trench from excavating is clearly visible. The maximum depth achieved is 1.0 to 1.2 m depending on the water depth during excavation.

# Full scale application based on field test results

The water treatment and therefore the first step was performed successfully. For a full scale application it needs to be assessed how much of the total fly ash body is necessary to neutralise the 30 million m<sup>3</sup> acidic water of Lake Burghammer. Treatment time is crucial due to the acidity intake by groundwater. For the calculation it is assumed that the treated water carries 5 mmol/L alkalinity. The treated water is released into the lake and mixed with untreated lake water. Using two midsize suction excavators or other excavation technologies (e.g. air lift pump) about 3600 m<sup>3</sup> water could be treated in one hour. If one further assumes that the operation is put into practice in a two shift system with 5 working days a week and 50 weeks a year every day 12 hours of operation are possible. One shift equals 8 working hours including two hours of maintenance, which is necessary due to the highly

aggressive waters. Using these assumptions it can be calculated that it would take less than 20 months to entirely neutralise the lake water. The dry content of the suspension is expected to be between 1 and 3 mass%. The relocated mass of the fly ash sediment body is therefore between 0.22 and 0.65 million tonnes which equates to 0.5 to 2 percent of the total available amount of reactive fly ash sediment.

#### Treatment costs and energy penalty

If one sums up the costs a total **2.15 million**  $\in$  would be necessary to treat the entire lake considering the site conditions. In this calculation it is assumed that 3 mmol/L CO<sub>2</sub> would be added to the suspension produced by the suction excavator. Over the treatment period a total of approximately 2850 t carbon dioxide would be used. To compare the treatment with other treatment strategies the specific costs per mole alkalinity are used. The proposed technology costs **1.5 cents per mole alkalinity** (or 8.6 cents per m<sup>3</sup> water). Treatment technologies within the region are estimated to be around **1.5 to 2.5 cent per mol alkalinity**. Costs could be reduced to **1.2 cents per mole alkalinity** (or 7 cents per m<sup>3</sup> water) if the emitted carbon dioxide (about 650 tonnes during operations) is used within the process and costs for carbon dioxide could be cut in half using synergy effects with the other research project related to carbon dioxide capture and storage.

The proposed technology is the most resource-conservative technology available for treating acidic lake water within the Lausitz mining area regarding carbon dioxide emission. Further, it is the cheapest treatment approach due to the use of two remaining products ( $CO_2$  and fly ash). Due to the small amount of fly ash sediment necessary to treat the entire lake the  $CO_2$  sequestration potential at Lake Burghammer is high. The amount of calcium hydroxide available is between 0.05 and 0.4 mol/kg. As mentioned before, the first layer of fly ash in the lake is characterised by low reactivity. A calcium hydroxide content of 0.05 mol/kg was calculated for this top layer. Lower layers show higher reactivity (0.2 mol Ca(OH)<sub>2</sub> per kg sediment).

The results confirm investigation of chemical sediment properties during the project. The total net  $CO_2$  sequestration potential (including emission during operation) is between 0.3 and 0.6 Mt  $CO_2$  for Lake Burghammer. A number of comparable lakes with fly ash deposits exist in the region. An alternative for the proposed treatment strategy is the widely known use of industrial produced chemicals like NaOH or Ca(OH)<sub>2</sub>. To put these into operation they firstly need to be produced and secondly transported to the remediation site. Both steps produce carbon dioxide.

#### Conclusion

It was shown in laboratory experiments that carbon dioxide can drastically enhance acidic lake water treatment and therefore reduce treatment costs. The bench scale experiment showed the general feasibility of the proposed technology. Reaction time plays an important role in the process scheme while turbulence and dry content of the suspension are less important but need to be considered. Based on the laboratory and bench experiments the pilot scale test was conducted in August 2007.

The results of the pilot scale experiment indicate that carbon dioxide and fly ash deposits could be used for acidic lake water remediation in an industrial scale. A low cost treatment scheme was successfully tested at Lake Burghammer, a post mining lake in Germany. Carbon dioxide and fly ash sediment were used in the pilot scale experiment. Generally it is possible to apply the developed and tested strategy at reduced costs to all treatment schemes where alkalinity is needed for remediation purposes.

#### Acknowledgement

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