Advanced treatment of pit lakes using limestone and carbon dioxide

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Abstract

In Lusatia (Germany) a lot of pit lakes were flooded due to rising groundwater over the last few years. Many of the newly developed lakes are affected by an inflow of potentially acidic groundwater from the adjacent overburden dumps. Therefore, technical treatments are necessary for a sufficient water quality. Liming is the most widely used treatment. At present various liming procedures are used. However, the most of them have a relatively low buffering capacity i.e. less than 0.3 mol/m³. This leads to, repeated treatments at short intervals being necessary.

A significantly higher buffering capacity can be achieved by the use of limestone and carbon dioxide. In 2015 a pilot treatment was conducted within the pit lake “Scheibe”, one of the largest pit lakes in Lusatia with a volume of 110 Mm³. The input of limestone and carbon dioxide was carried out with two underwater nozzle pipelines. This technology has already been used for the primary neutralization of Lake Scheibe (Strzodka et al. 2013). In this paper we will present this new treatment concept in detail and the results of the pilot test.

Key words: Mine water, pit lakes, buffer capacity, treatment

Introduction

Usually, after open pit mining ceases, the residual hole is filled and a pit lake is created. In the Eastern German Lusatian lignite mining area, this will result in Europe’s largest artificial lake district. Many of these lakes comprise large water volumes and surface areas and are therefore amongst the largest lakes in Germany (Nixdorf et al. 2001). The inflow of potentially acid groundwater from the adjacent overburden dumps results in sulphate dominated acidic conditions.

Usually, lime products are used to neutralize acidified lakes. After pure lime treatment the waters show pH-neutral, weakly buffered waters. These environmental factors can only be maintained for a relatively short time. A continuous inward flow of acidic ground water leads to successive re-acidification and sometimes costly follow-up treatments.

A stronger pH buffering can also be reached with developing a hydrogen carbonate buffer (Alkalization) in the lake water body with the application of lime products and CO₂ (Koch 2009) thus reducing the periodic treatment cycles. The aim is to produce a reaction of the components incorporated into the lake water to produce hydrogen carbonate. The restriction of producing hydrogen carbonate solution in a technical reactor can be avoided by transferring the reaction to the water body itself (LfULG 2012). A considerable advantage is the utilizable reaction volume and the longer reaction times.

This paper presents our work with the aim to develop and test an inexpensive and efficient procedure of feed-in the alkalization media lime and CO₂.

Methods

To produce a hydrogen carbonate buffer a lime product and CO₂ are usually fed into the lake water body. The chemical reaction between the reactant and the lake water produces hydrogen carbonate
which is the aim of the Alkalinization process. The advantages of transferring the reaction to the water body itself lies in the utilisable reaction volume and much longer reaction periods for the hydrogen carbonate solution to reach a target concentration for the desired lake water quality.

The feed-in technology uses the underwater nozzle pipeline (UNP) with jets which had already been proven suitable to mix and produce a lime suspension (Strzodka et al. 2013). From the start of the pilot project of developing the procedure and concept of the system to neutralize the Lake Scheibe the existing UNP from the past treatment (as of now liming plant) was fully integrated in the planning phase.

To have an efficient method with a minimum of energy and thus inexpensive incorporation of CO$_2$ there are similar requirements as in introducing lime. An evenly mixed alkalinization medium in the lake water body (hypolimnion) is vital for the procedure. Bearing this in mind and using the free jet principle, another plant was erected on the existing site to feed in CO$_2$ (as of now CO$_2$-plant).

The technological approach was to apply the lime in suspension using the existing liming plant from the past liming treatment. The suspension will be fed over the UNP close to the lake surface (epilimnion) using the free jet principle. Within the same time frame and in the same area, conditioning of the deep waters (hypolimnion) with CO$_2$ will be carried out using the newly erected CO$_2$-plant. The CO$_2$ will be dissolved in water drawn from the lake into the plant on land and then fed via the UNP pipelines using the free jet principle once again towards the bottom of the lake.

By using the procedure in the natural thermal stratification period of summer the suspension spreads horizontally along this boundary layer (metllimnion) due to the density differences. In addition the boundary layer metalimnion has a function of reducing the premature CO$_2$ loss to the atmosphere. The lime particles dissolve successively on the way through the hypolimnion reacting with CO$_2$ and thereby have a buffering effect on the hypolimnion from the hydrogen carbonate. In the course of the water body’s full circulation there will be an even distribution throughout the entire lake.

**Area under Investigation**

The test of the UNP procedure for Alkalization was carried out in the Lake Scheibe near the town of Hoyerswerda. It has a volume of 110 Mm$^3$ and is rated as one of the largest pit lakes in the Lusatian lignite mining area. It has a length of 5.2 km and a maximum width of 1.7 km. As a result of the lignite mining technology used, the lake’s morphology is characterized by two distinct features: the eastern part of the lake consists of the former pit’s inner dump with a shallow water area with a depth of 2-6 m and the western part with a water depth of 35 m (fig. 1). Lake Scheibe is characterized by dimictic conditions with full circulation phases in spring and fall.

Determining aspects of the water composition of Lake Scheibe are the ground water inflows into the lake: from the south, from the mother rock a slightly acidic ground water with acidity of approximately 1.0 mmol/L and from the inner dump water an acidity of 6.7 mmol/L.

The initial determined state for Alkalization of the Lake Scheibe to be treated was 0.07 mmol/L buffering, and a pH around 5.0.
Pilot Project Implementation

To neutralize the water body a constant layering of pulverized limestone (KSM 60/3) in the epilimnion and \( \text{CO}_2 \) in the hypolimnion was planned. The change from using the quicklime (used in the primary neutralization) to KSM was necessary as for procedural purposes this lime product lies on the metalimnion with a more concentrated effect and so the time it is transported into the hypolimnion is retarded. During liming the feed quality requirement of the epilimnion must still be maintained. This is only possible with a carbonated lime product (may not exceed the pH-value of calcite saturation). Simultaneously the acceptable pH level in the epilimnion necessary for flora and fauna is retained.

After the full circulation in fall, a buffering of 1.0 mmol/L throughout the lake water should be attained. For KSM lime 83 g/m³ is the required dosage to reach a 65 % chemical efficiency. 9.075 t KSM is fed into the entire lake.

Firstly in buffering the hypolimnion the stoichiometric required amount of \( \text{CO}_2 \) to dissolve the pulverized limestone has to be present in the water. Secondly there must be enough driving force to be able to reach an adequate chemical efficiency of the reaction. As a result of pretesting it was established that a temporary lowering of the pH in the hypolimnion is necessary by feeding \( \text{CO}_2 \) to reach \( \text{pH} \leq 5.5 \). It was planned to feed in a total of 5,000 t \( \text{CO}_2 \) in within 10 weeks. The aim was to feed in 3.3 t \( \text{CO}_2 \)/h with 1,000 m³ pumped lake water per hour.

The process related components of the UNP procedure to alkalize the Lake Scheibe were kept very simple (fig. 2).

To produce and introduce the \( \text{CO}_2 \) solution, submersible pumps are used to draw lake water from below the metalimnion and feed it into the gaseous dissolving reactors on land. In this mixing installation, gaseous \( \text{CO}_2 \) from a tank and intermediate vaporizers is dissolved in untreated water. Pumps on land with an additional capacity ensure the required feed pressure. The \( \text{CO}_2 \)-rich water is then pumped through the pipes in the hypolimnion. As in the lime suspension feed process there are jets in pairs installed at the end of these pipes (positioned close the bottom of the lake - deep water area), for injecting the \( \text{CO}_2 \)-rich water according to the free jet principle. For the system at Lake Scheibe 10 pair nozzle configuration at intervals of 10 m was planned. To ensure a gaseous free exit from the jets the pipeline is kept under positive pressure.

With the proportion of the size of the water body to the entire configuration of the Alkalization – facility one point of feed only is needed (fig. 3).
The pilot project to alkalize the Lake Scheibe started 10.06.2015 (lime- and CO\textsubscript{2}-conditioning) After 14 weeks in operation the CO\textsubscript{2}-feed ended 28.09.2015. There was an interval with reduced CO\textsubscript{2}-feed as well as a break in operations (fig. 4).

The liming treatment ended 30.10.2015 after 17 weeks of operations. As a result of the reduced CO\textsubscript{2}-dosis and the break in operations in the CO\textsubscript{2}-plant the liming was temporarily stopped.
In monitoring the lime and CO$_2$-conditioning 14 water samples were taken and analyzed, at weekly intervals at 5 sampling points (fig. 1). Based on the changes of the water quality, the specifications for the further operation of the Alkalization plant were determined.

The monitoring of the hydraulic conditions of the lake provided the basis for validating a 3D lake model (MOHID-Water Modelling System). Both data were used to verify the design calculation algorithms previously used.

**Results**

The thermal stratification of the lake was fully developed when the treatment began at the beginning of June. The lake’s metalimnion was 10 m below the water surface. At the end of September 2015 the thermal stratification began to gradually breakup. The phase of full circulation only started in December.

The change of the average lake water’s condition over the time period is demonstrated in fig. 4. The measure for alkalinity is derived from the parameter neutralization potential NP (Evangelou 1995) as modified by Schöpke 2008.

During the course of feed-in of the materials carried out to October 2015, the buffering effect of the lake water increased to 0.87 mol/m$^3$. The periods of both interruptions of the liming plant and that of the CO$_2$-dosing can be seen in fig. 4. With the interruption of the feed-in of pulverized limestone the NP of the lake water remained constant.

Samplings of the after-run period showed that after the end of treatment the re-acidification due to a net-acidified groundwater was not immediately detectable. A secondary reaction progressed into December 2015, which was able to overcompensate for the acidity in the ground water inflow. Measurements in December indicate a lake water NP of 0.9 mol/m$^3$. In March 2016 there was an NP of 0.79 mol/m$^3$ (fig. 4). If the re-acidification trend of the values measured in December and March are followed to the end point of Alkalization (30.10.2015) it corresponds that the treatment had an effect of producing a final NP value = 0.95 mol/m$^3$. The buffering of the lake water was increased by 0.88 mol/m$^3$. The target value if 1.0 mol/m$^3$ was thus not quite reached.

![Figure 4 Temporal progress of the lake Scheibe treatment.](image-url)
As a result of the surface treatment with the pulverized limestone and the introduction of CO$_2$ close to the bottom of the lake a characteristic pH-profile developed which remained throughout the duration of the entire treatment. The Epilimnion remained pH-neutral. Towards the bottom of the lake the pH value was lowered to approx 5.5 due to the in-feed of CO$_2$. This had an effect of providing a driving-force or at least maintaining it, to dissolve the pulverized limestone. With the interruption in the CO$_2$-feed on 29.09.2015 there was a hypolimnion with a successively higher pH-value.

With the UNP equipment, CO$_2$ is fed into the lake with virtually no losses during the summer stagnation. At this stage there was no abnormal increase in CO$_2$ loss into the atmosphere. The CO$_2$-pipeline with mass dosage of up to 3.6 t/h is unique in the context of lake quality restoring.

In September 2015 a side reaction was detected with demanganesation within a period of three weeks only. However this reaction was restricted to the epilimnion. The reduction of the manganese content from 0.9 mg/l to 0.1 mg/l was presumably from a catalytic reaction of microbial origin. With the precipitation of the manganese oxide hydrate, the bottom of the lake turned temporarily black giving the water a black appearance.

A further microbiological metabolic process was detected, the decrease in ammoniacal nitrogen, from the end of September into November. In contrast to the demanganesation the metabolic materials did not remain exclusively in the epilimnion. Due to the pH and temperature gradients the reaction started up in the epilimnion, due to the favourable conditions. Although for nitrification the pH-environment in the hypolimnion was suboptimal (Rusten et al. 1995), comparable concentration differences to the epilimnion were reached. In total the lake’s average only changed from 1.1 to 0.7 mg NH$_4$-N/l. A key limitation of the metabolic conversion rate and the biomass growth is caused by the low concentrations of phosphorus in the iron-rich water or previously iron-rich water. (De Vet et al. 2012, Nordeidet et al.1994).

Since 2010 the development of the water quality of the lake has been simulated based on a balance model. This balance model continues to serve for forecasting future developments. Over the last few years there has been a steady increase of acidic water introduced via the groundwater. The annual inflow of acidic was previously 20 mio. mol and has now reached 30 mio. mol. The forecast is made on the assumption that the hydrogen carbonate buffering capacity is exhausted after 3 years. In comparison to simple liming the treatment intervals could be more than tripled.

After conditioning the lake water with lime and CO$_2$ buffering with hydrogen carbonate is 5.5 times more effective than plain lime, costs of alkalization 4.6 times. With this the economic viability of the pilot project was confirmed.

Conclusions
The UNP-procedure for introducing limestone and CO$_2$ was technically and economically proven as effective in the Alkalization of Lake Scheibe.

An Alkalization medium of pulverized limestone and CO$_2$ was evenly applied and mixed into the lake water in a targeted manner over a period of 100 days. The required mass flow rates achieved within a lake water restoration context are unique. With the introduction of CO$_2$ the driving force was produced to dissolve the KSM (pH-ratio in the hypolimnion of 5.5) was created and maintained during the entire treatment time. The CO$_2$-feed followed under stratification conditions during the summer stagnation with virtually no losses. With the completion of Alkalization the buffering was increased to an average of 0.9 mol/m$^3$. The target value of 1.0 mol/m$^3$ was partially reached. The forecast for re-acidification is based on the hydrogen carbonate buffering effect being exhausted after 3 years. This is equivalent to more than three times of a plain liming treatment. The field of application of the UNP procedure was successfully extended.

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