Due to acidic inflow from sulphur- and iron-rich groundwater into mine lakes in East Germany, the majority of the mine lakes and their run-off waters do not meet the national water quality requirements. Hence, water treatment is required either for the lake water bodies or the discharged waters. In-lake neutralization of the entire water body is the preferred option with regard to cost efficiency, water quality, and subsequent use. Over the last 15 years, LMBV and its partners has developed, tested and enhanced different in-lake neutralization techniques. Due to these efforts, the achieved state of technology will be presented. In East Germany, 17 acidic mine lakes have already been neutralized and need regular follow up treatments to stabilize the neutral conditions. At least five more lakes are to be neutralized in the next few years. This makes LMBV on of the most experienced organization in-lake technology for mine lakes.

Keywords: Lignite mining, contamination by sulphide minerals, active water treatment, in-lake neutralization of mine lakes

Introduction

Lignite mining in Germany has been carried out under soft rock conditions for about 150 years. In order to allow safe mining, the groundwater layers have been dewatered to a depth of about 50 – 80 m. To remove the overburden from lignite seam, the material has been displaced to outer damp sites. In the mined-out areas, large end lakes have been created with a volume of 20 – 450 mill. m³ (LMBV 2017a).

In the phase of mine operation and closure, sulphide minerals (e.g. pyrite and marcasite) in the overburden had been oxidised. The minerals formed water-soluble iron and sulphate ions, as well as small amounts of other heavy metal ions in some cases. When mining ceased, the groundwater layer outside the mining site was refilled and the sulphate and dissolved iron minerals were transported by the re-established groundwater flow. Due to acidic groundwater flow, the majority of the mine lakes and their run-off waters do not meet the national water quality requirements. Hence, water treatment is required either for the lake water bodies or the discharged waters. In-lake neutralization of the entire water body is the preferred option within regard to cost efficiency, water quality and subsequent use.

Water treatment principals

In order to comply with German water regulations §§ 27, 44 and 47 (WHG 2009) as well as Article 1 of the EU Water Framework Directive (WRRL 2000), the quality of surface and underground water may/must not be deteriorated by mining activities. To ensure the water from the mining lake as well as runoff waters do not adversely affect downstream biocenosis, a water treatment according pH values and iron components is necessary. Active water treatment is the most common form of water treatment in mining industry. Because the mine lake waters are slightly acidic (pH values from 3 to 7, iron levels from 1 to 150 mg/L), they require the addition of lime, limestone or soda to raise the pH value (LMBV 2017a). Once the pH value has been elevated, dissolved iron precipitates out of the solution and sinks to the bottom of the lake. As a result of the large amount of water being treated, techniques like ion exchangers, membrane filters or reverse osmosis are not suitable in this case.
In-lake neutralization involves the surface water body in the former opencast mining pit by mixing a neutralizing agent into the water. With this process, the total water body as well as sediment of the lake, erosion material and the water flowing into the groundwater aquifer must be neutralized. The main advantages compared to a run-off treatment are usually the following:

- the in-lake treatment is required just periodically, run-off treatments have to operate all the time,
- the degradation of ammonia by microbes in neutral water bodies,
- the iron sludge formed during neutralization settles on the lake bottom, and does not have to be disposed of elsewhere,
- the output of neutral lake water into the adjacent downstream groundwater aquifer and
- the higher usability of neutral waters (e.g. fishing, recreation, …)

**Choice of in-lake procedure**

Technically, not all treatment procedures are equally suitable for all types of lakes. Lake shore-based stationary water treatment plants would appear to be the preferable solution when continuous treatment cycles are required. They could be automated to a large extent so they can be both operated and monitored remotely. The lime should be introduced into the lake water body at an appropriate distance from the shore using pipes and subsurface turbulent jet technology. The required lime suspension is produced with lake water on the shore. Stationary plants of this kind are well-suited for relatively compact water bodies where good mixing as a result of the convective lake water currents can be expected (LMBV 2017b).

Some German mining lakes are heavily segmented and made up of several sub-basins. Here it is more likely that mobile plants (i.e. water treatment vessels) are capable of distributing the neutralization agent across the lake as required.

**Water treatment vessels**

The liming process of lakes has been long practiced in those regions in Scandinavia that have been affected by acid rain in soft water areas. One such Swedish water treatment vessel, the *Brahe type*, was used for the first time on an eastern German post-mining lake in 2008. The lime-water suspension was spread across the lake surface by two water guns. The vessels could easily be transported by road on a trailer from which they can be launched to a lake. One disadvantage was that the relatively coarse-grained pulverised limestone being used was much less effective than in the soft water lakes of Scandinavia. Another ecological drawback is the risk of the fine lime particles drifting into the reed beds along the lake shore as they are unable, or only slowly able, to penetrate the air-water boundary. Therefore this vessel was changed over from over water to under water distribution.

In this context, the *Barbara* (Fig. 1) is another water treatment vessel, based on under

*Figure 1 Water treatment vessel Barbara in operation, and (bottom right) being lowered – showing the delivery system between the vessel’s hulls (courtesy: LUG/LMBV)*
water technology. The vessel is a catamaran with two tube mixers mounted between the hulls. This allows additional mixing by the vessel’s two propellers at the ends of the twin hulls. The vessel has two lime bunkers, each with a capacity of 12 m³, allowing material to be bunkered according to the bulk density of the lime.

Positive aspects of this new technology are high effectiveness of neutralization agent and economic mixing technology. This is due to the high turbulence and the low concentration of the solid matter with a suspension of way less than 1 %. The main disadvantage of this vessel is the long loading time of the on-board silo. A further step of development of particular relevance to LMBV is the water treatment vessel Klara (Fig. 2). It has been designed and built specifically for use at the Lausitzer Seenland, which are former mining pits.

The Lausitzer Seenland consists of nine former mining pits, which are connected by navigable canals, in the federal states of Saxony and Brandenburg (Fig. 3) between Senftenberg and Spremberg. Although Lake Senftenberg is nevertheless connected to the chain by the Koschen Canal. The water surface area of the entire lake chain exceeds 55 km², and it has a volume of more than 800 million m³.

Figure 2 LMBV water treatment vessel Klara during trial operation (courtesy: LMBV)

Figure 3 Lausitzer Seenland, former mining pits and Lake Senftenberg – 9 interconnected post-mining lakes (courtesy: LMBV)
The Klara dimensions were defined so that it would be able to pass through all canals, bridges, and locks. Klara vessel consists of a push boat with two barges with silos (Fig. 4).

The engine, generator, and the bridge are on the push boat. There are two lime silos on each barge, and beneath each of the barges, two discharge units have been mounted between the catamaran twin hulls.

This discharge unit arrangement of the water treatment vessel Klara has the advantage that while the first barge is on the lake with the push boat discharging neutralization agent, the second barge can be loaded. It reduces the time between two treatment cycles to about 5 minutes for changing the barges. Klara can distribute caustic lime, pulverised limestone, or calcium hydroxide, and is able to deliver neutralization very efficiently. It reaches high levels of dilution in lake water by boosting the agent by the vessel propellers. The vessel is dimensioned for the discharge of approximately 40,000 t of neutralization agent per year. Klara is therefore able to carry out both the initial neutralization as well as the follow-up for the entire Lausitzer Seenland (s. Fig. 3).

**Choice of neutralizing agent**

With regard to the neutralizing agents most frequently tested so far, the following statements can be made (LMBV 2017b). These are based on experience gathered over the past few years, regarding their efficient and effective use:

**Caustic lime**, pulverised calcium oxide CaO:
- high neutralization equivalent (approximately 30 to 35 mol/kg),
- reasonable price,
- high reactivity ensures a high kinetic rate of reaction, but can also cause locally high pH values in lake water

**Calcium hydroxide**, main ingredient: ground Ca(OH)$_2$
- neutralization equivalent of approximately Neq = 25 to 29 mol/kg,
- relatively high specific price due to the additional process steps involved in its production,
- supplied as a prepared lime water suspension is an advantage compared to the use of caustic lime.

**Pulverised limestone**, consisting of ground CaCO$_3$ (limestone or chalk)
- relatively low neutralization equivalent of Neq ≤ 20 mol/kg,
- efficient use, it needs to be finely pulverised (e.g. d90 < 40 μm),
- low specific costs, approximately 0.3 Euro cent per mole of alkalinity,
- ability to reach higher alkalinity more efficiently than when caustic lime is used, developing hydrogen carbonate buffer up to $K_{S4.3} = 0.5 \text{ mmol/l}$,
- ecological advantages due to the fact that over neutralization to pH-values of more than 8 are not possible.

**Soda**, main component: ground Na$_2$CO$_3$
- easily soluble in all pH ranges, does not require any complicated technology,
- comparatively expensive, and
- ecologically not ideal as it increases salinity (i.e. elevated sodium level).
Although the specific prices per mole of alkalinity of caustic lime and of pulverised limestone are very similar, caustic lime is usually the more cost-effective option. Caustic lime would be preferred wherever neutralization does not require alkaline buffering of alkalinity > 0.25 mmol/l at 6 < pH < 7. The focus of initial neutralization will therefore be on the use of caustic lime, while follow-up neutralization of post-mining lakes will focus on pulverised limestone. In coming years, when the initial neutralization will have been completed, this proportion/ratio will shift further towards finely pulverized limestone. Under certain circumstances, the use of chalk products as a specific form of pulverised limestone may become increasingly relevant.

**Use of gaseous CO₂**

The use of CO₂ to develop a hydrogen carbonate buffer decreases the need for follow-up treatments significantly. Generally, it can be stated that hydrogen carbonate buffering is a relevant follow-up treatment as well from the economic as from the ecological point of view (LMBV 2017b). It avoids the rapid neutralization during the treatment phase, and slower re-acidification during intervals without treatment. The pH fluctuations from 5 < pH < 9 is assessed to be disadvantageous for bioocoenosis in the water. The use of CO₂ will remain of interest in the future, especially to prevent the pH value from sinking during the winter if the lakes freeze over. This had been the case at the Lake Drehna pilot project.

Here classic liming technology eight liming campaigns with caustic lime are necessary per year, and a pH-drop during the ice cover could not be avoided for sure. The use of CO₂ is limited to the fact, that over-dosage will harm biological species in the lake. Using CO₂ a three-month campaign is sufficient to keep the pH-value stable for more than a year.

**Summary**

A relevant example of a mobile plant used for neutralization is the LMBV water treatment vessel *Klara*. It was custom designed and built for the initial and follow-up neutralization of the Lausitzer Seenland, the mining lake. The *Klara* is the most powerful water treatment vessel built to date, which follows up the development of several in-lake treatment vessels. Other large treatment vessels such as the *Barbara* are suitable for post-mining lakes with relatively high demands for neutralization agent. They have proved their usefulness. However, hauling these large water treatment vessels from one lake to another by road is complicated. They also require suitable areas for launching the vessel and charging with neutralization agents. Small water treatment vessels such as the *Brahe* can easily be watered and hauled without requiring a heavy-load transport licence.

For initial neutralization, quicklime tends to be the most cost-efficient neutralization agent. For follow-up neutralization, finely ground limestone usually is the preferred agent due to health and ecological reasons. Under certain conditions the buffering of neutralized waters by the additional use of CO₂ gas can also be an appropriate method.

LMBV is able to carry out efficient and very effective in-lake neutralization measures using mobile water treatment vessels and stationary neutralization plants. By now, there are a number of well tested techniques available. The decision on technology and neutralization agent depends on both, technological and economic considerations. These decisions are usually made on a case-by-case basis.

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