In-Lake Water Treatment of Post-Mining Lakes – Reactivity of Lime Products

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Abstract

A new method has been developed which determines the alkalinity release of limestone powders in water under neutral conditions. This shows clear differences between the various limestone powder products. In addition, the products were also tested at different pH values. A significant decrease in alkalinity release was observed with increasing pH values, whereby product differences again came into play. The comparison with water treatment campaigns that have been carried out confirms in principle the correlations found in the laboratory. However, further influencing factors that led to the deviation from the norm can be identified in order to make future measures more efficient.

Keywords: Lime, neutralisation equivalent, post-mining lake, water treatment

Introduction

The remediation of the legacy of lignite mining became a state responsibility after the reunification of Germany. The LMBV restoration of the Lusatian water balance mainly consists of the refilling of ground water deficits, the creation of secure pit lakes, their connection to rivers and the enhancement of water quality.

Water treatment of post-mining lakes is usually carried out using in-lake technology. With this technology limestone powders are distributed by vessels into the entire lake water body thereby mixing it with the lake water to maximise lime dissolution kinetics. In this process, the entire lake water body is raised to a neutral pH value during initial treatment. Due to the continuous inflow of acidic groundwater into the post-mining lakes, repeated follow-up treatments are necessary to maintain neutral pH values. Calcium carbonate-containing neutralizing agents in the form of limestone powder are usually used for this purpose. The limestone powders available on the market are characterized by different chemical (e.g., purity) and physical (e.g., grain size) properties. When treating water in the neutral range, the various limestone powders therefore release varying amounts of alkalinity into the water body.

Until now, there has been no standard for determining the alkalinity release of carbonate lime products with respect to lake waters. On order from LMBV, a procedure for determining the reactivity of carbonate lime products was developed and a procedural specification was created.

Methods

Initial preliminary investigations to identify a practicable investigation concept have shown the titration to a stable pH value (pH-stat titration) method to be effective. This method experimentally measures the alkalinity of the lime product dissolved in solution as a neutralisation equivalent (Neq) based on acid consumption. The extensive series of experiments took into account factors that theoretically influence the dissolution kinetics of calcium carbonate particles, such as temperature, pH value, CO2 partial pressure, ionic strength, turbulence, particle diameter. The aim of developing the method was to ensure sufficient reproducibility of the measurement results.

As a result, a procedural rule was defined stipulating that a target pH value of 6.5 must be kept for titration over 90 minutes. The limestone powder sample must be homogenised and dried before testing. The suspension is made in deionised water, which is in CO2 partial pressure equilibrium with the laboratory air. This must be stored openly in the laboratory for at least 12 hours before the test. 0.1 molar hydrochloric acid is used for titration. To ensure a defined energy input via the magnetic stirrer for the preparation of the suspension, the speed must be adjusted so that the resulting vortex occupies 1/3 of the water height in the beaker. The consumption of hydrochloric acid is recorded after 90 minutes and the neutralisation equivalent (Neq) is calculated using the mass of the tested lime product. Eleven different lime products were examined as part of the method development. The results are visualised in Fig. 1. In addition, the alkalinity content according to the product data sheet is shown for comparison. Previous experience has shown that the alkalinity content can be almost completely released in an acidic water environment. However, to fulfill legal regulations the seawater treatment must ensure a lake water pH value of at least 6.0. This is why the developed procedure focusses on alkalinity release under neutral conditions.

The Neq90 values for the limestone products examined are compared in Table 1 with their geological origin, chemical composition and mean particle diameter.

The results clearly show the product-specific differences between limestone powders in terms of alkalinity release under neutral conditions. It can be seen that geologically young limestones, known as chalk, exhibit a substantial higher alkalinity release due to their large specific surface area. Older, more compact or metamorphically altered limestones, even when finely ground, show comparatively lower dissolution and alkalinity release in neutral water.

In addition, the reactivity of four selected limestone powders, specifically lime 2, 7, 10 and 11 was examined in a pH range of 5.5 to 7.5. As expected, there is a clear pH dependence, with alkalinity release increasing clearly as the pH value decreases for all products tested. At a pH value of 5.5, between approximately 65% and 90% of the maximum possible alkalinity release was measured, depending on the respective product. At a pH value of 7.5, only 13% to 14% of the product's alkalinity could be transferred to the water phase. The differences in the Neq value between the individual lime products decrease as the pH value increases (see Fig. 2).

In a further study, the effectiveness of a total of 86 water treatment campaigns using limestone powder was investigated at

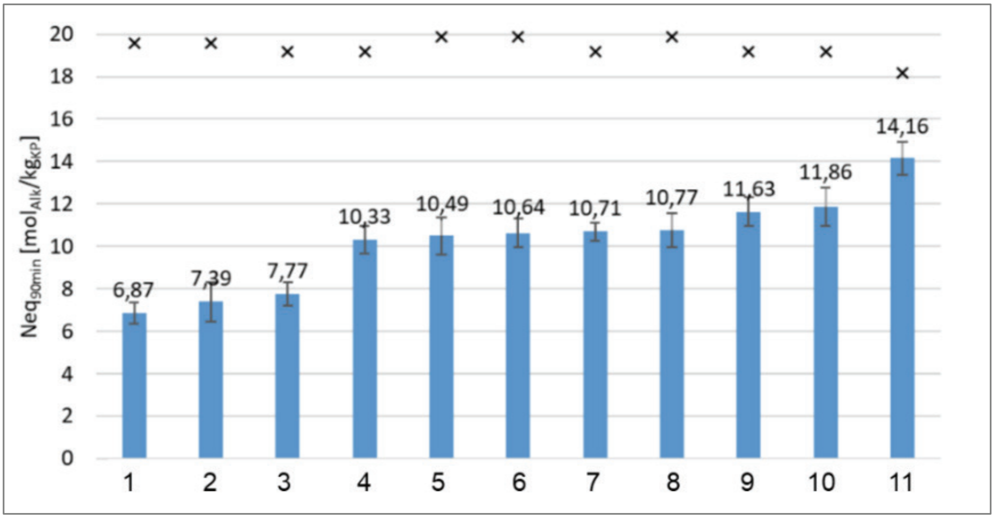

Figure 1 *Neq90 values [mol/kg] (columns) of the 11 carbonate lime products used for method development compared to the alkalinity content according to the product data sheet (crosses) (BTU, GFI, 2021).*

Table 1 Properties of the lime products examined.

lime	geological era	chemical composition [%]			Median particle diameter d50 [µm]	Neq90 [mol/kg]
		$CaCO_3$	$MgCO_3$	Fe_2O_3		
1	Devon	96,9	0,9	0,2	12,0	6,87
2	Devon	97,0	0,9	0,2	8,2	7,39
3	Early Palaeozoic	96,0	<1,0	0,3	3,0	7,77
4	Jura	>90,0	<1,0	<0,2	3,1	10,33
5	Jura	99,3	0,3	0,1	5,0	10,49
6	Jura	99,3	0,3	0,1	4,5	10,64
7	Jura	>95,0	<1,0	<0,2	3,1	10,71
8	Jura	99,3	0,3	0,1	3,5	10,77
9	Jura	>95,0	<1,0	<0,2	3,1	11,63
10	Jura	>95,0	<1,0	<0,2	2,9	11,86
11	Cretaceous	90,0	1,0	0,5	3,0	14,16

a total of ten post-mining lakes in Lusatia. According to [LUA, 2001], the overall efficiency η_{ges} of an in-lake process based on the mass application of the neutralising agent can be represented as the product of the material efficiency ŋ0 and the technological-chemical efficiency ŋ1 (IWB, 2025).

Equation 1: Overall efficiency of an in-lake process (LUA, 2001) .

$$\eta_{ges} = \eta_0 \cdot \eta_1$$

η_{ges} Overall efficiency [-]

η_0 material efficiency (active ingredient content) [-]

η_1 technological-chemical efficiency [-]

While the material efficiency η_0 is a largely stable product property of the neutralising agent (material purity), the technological-chemical efficiency η_1 is the ratio of the alkalinity that has become effective in the lake in relation to the alkalinity supplied with the material. This is influenced by various factors, such as the genesis and structure of

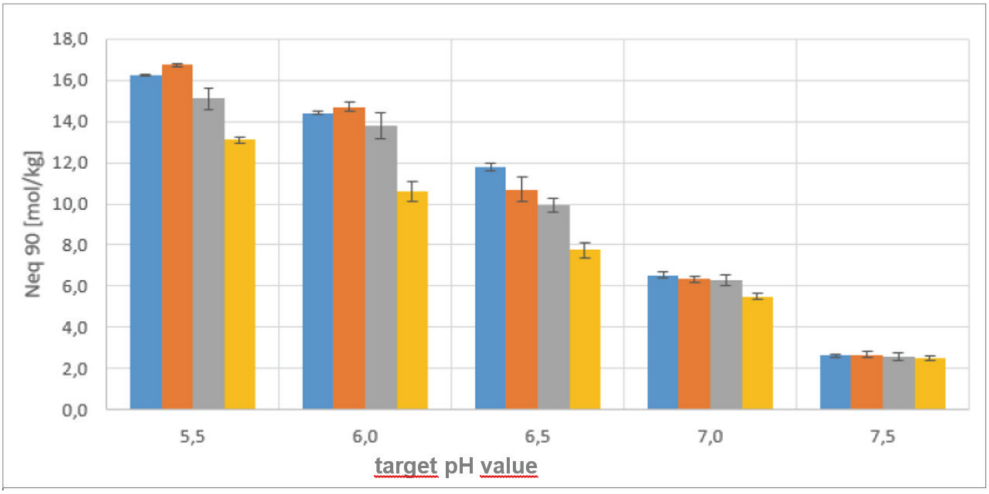

Figure 2 Neq90 values [mol/kg] of the four selected lime products at different final pH values in the pH-Stat test (GFI, 2025).

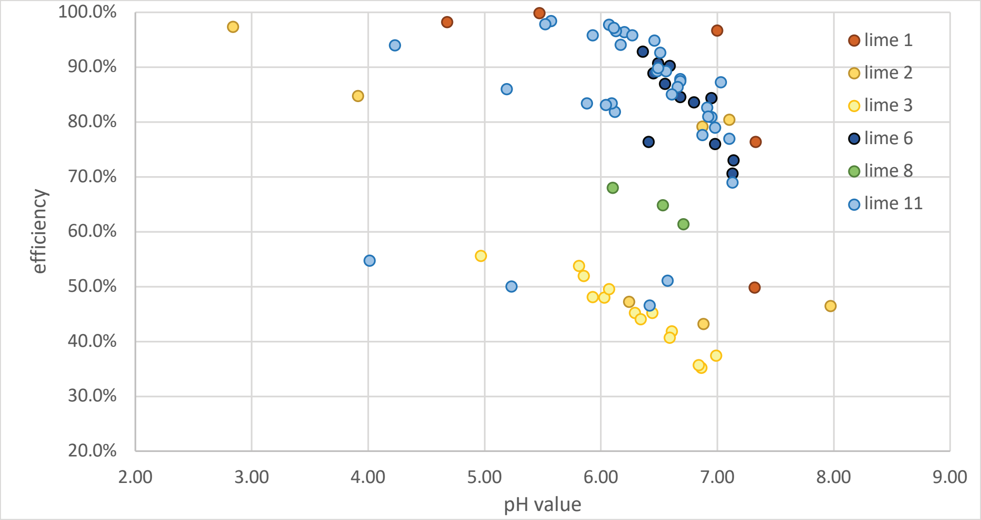

Figure 3 Technological-chemical efficiency of in-lake treatments as a function of the pH value of six different limestone powder products.

the limestone product (density, grain size distribution, specific surface area of the lime particles), the variable physical and hydrochemical conditions in the lake water (pH value and water temperature), the lake-specific boundary conditions (water depth, wind exposure of the post-mining lake) and technological factors such as application as a suspension or dry dosing, the distribution system and others (IWB, 2025).

The evaluation of the technological-chemical efficiency of in-lake measures at ten post-mining lakes and using six different lime products shows a clear correlation between the efficiency and the average pH value of the lake water during treatment. The correlations determined in the laboratory in Fig. 1 could in principle be found again in real-world applications (see Fig. 3). As a rule, the efficiency of lake treatment decreases as the pH value rises. This is particularly evident for lime products 3, 8 and 11 (Fig. 3) which are frequently used.

Furthermore, the evaluation shows that material with predominantly good technological-chemical efficiency levels, such as material no. 11, also appears sporadically in areas with efficiency levels <55%. These campaigns should be given special attention in the further evaluation. Here, the other influencing factors that led to the deviation from the norm must be determined. Possible causes are seen in the dosing rate, i.e. the amount of neutralising agent added per volume of water, the input energy, the water temperature and the hydrochemistry.

The in-lake treatment was carried out both by vessels and by stationary plants on the shore. The water treatment vessels either dose the lime products as a dry powder via a rotary valve or as a premixed suspension into the water. The propper distribution of the lime suspension in the lake is supported by the movement of the vessel. In the case of the shore-based systems, the lime suspension is fed through a pipeline and discharged into the water via nozzles on a discharge pontoon anchored in the post-mining lake. Mixing is primarily achieved by the impulse of the water flux. In both processes, currents within the lake contribute to further mixing.

The evaluation of the dosing technology used in 68 campaigns examined does not reveal any significant differences (see Fig. 4).

Results

The determination of the product-specific parameter neutralisation equivalent (Neq) for lime products in in-lake treatment and the model-based evaluation of the effectiveness

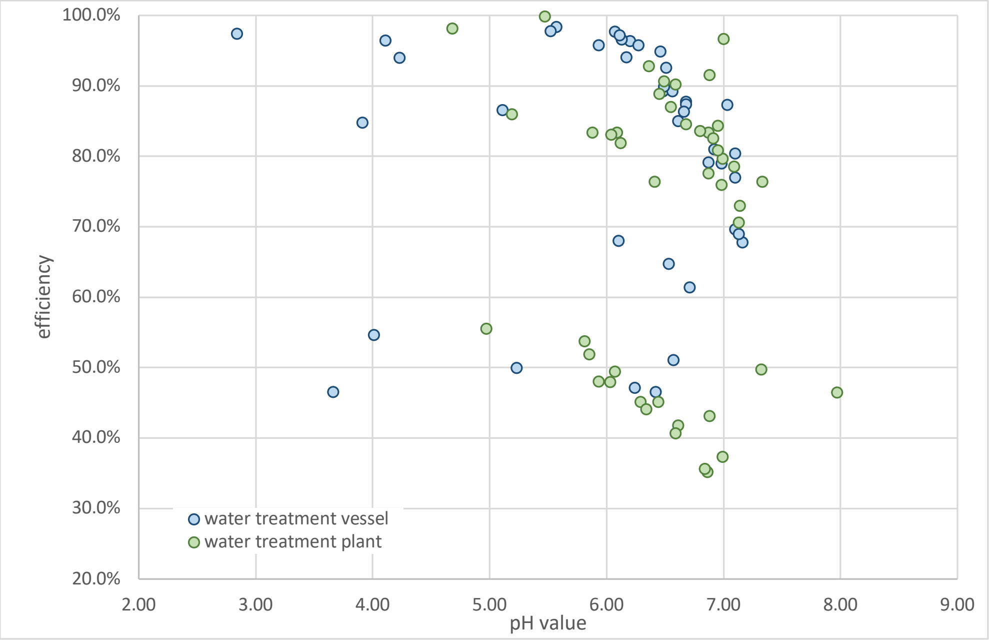

***Figure 4** Technological-chemical efficiency of in-lake treatments as a function of the pH value of two different application technologies.*

of in-lake measures in Lusatia have revealed both product-specific differences and the strong pH dependence of the dissolution of lime products. The dosing rate is also a key parameter influencing efficiency (IWB, 2025).

This suggests that the start and end of the treatment measure should be at the lower end of the pH range (pH 6.0 to 7.0) if possible. The technical prerequisite for such flexible treatment is a ready-to-use, stationary plant or vessel available in the water body. In addition, it is crucial to monitor the water quality at regular intervals and to adjust the treatment measure if too high pH values are encountered.

Conclusions and outlook

The results of this study are used for purchasing limestone powder for in-lake treatment. Suppliers are no longer asked to provide a quantity of lime product, but rather the required alkalinity. Water treatment measures for post-mining lakes with a pH value of more than 7 will also be avoided in the future. This will make in-lake treatments more cost-effective. In addition, quality monitoring and evaluation of measures is based on these more reliable laboratory data.

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