

Carbon dioxide elimination by using acid mine lakes and calcium oxide suspensions (CDEAL)

Merkel, B. (1), Werner, F. (2), Wolkersdorfer, C. (1)

- (1) TU Bergakademie Freiberg, Lehrstuhl für Hydrogeologie, 09596 Freiberg/Sachsen, merkel@geo.tu-freiberg.de
(2) Dresdner Grundwasserforschungszentrum e.V., Meraner Str. 10, 01217 Dresden, fwerner@dgfz.de

Introduction

CDEAL's concept is to combine CO₂ mineral trapping and treatment of acidic mine waters. In the Lausitz (Lusatia) mining district intensive mining during the last century has produced huge deposits of fly ash, carbon slurry, and iron sludge (caused by the treatment of acid mine drainage) in the abandoned and flooded mine pits. One of these former pits is the acidic Burghammer pit which contains large amounts of these substances deposited on its bottom. A treatment of lake Burghammer sediments and water with CO₂ appears thermodynamically feasible using carbonation. Kinetic aspects, the real world phase assemblage, and technical aspects of the dosage, mixing, precipitation, and settling of the carbonate solid will be investigated in the project. CDEAL will perform laboratory experiments in the first stage and in case of a positive evaluation a feasibility pilot scale plant will be constructed and a field scale CO₂-injection plant will be installed at the Burghammer open pit lake.

Overall Goal

Generally, the Kyoto-Protocol requires a reduction in greenhouse gases, especially carbon dioxide. As concerns exist over the effects that such an action might have on the economy of developed countries, a worldwide search for other approaches to reduce carbon emissions has started. Existing engineering alternatives can significantly reduce CO₂ levels, while still allowing to rely on inexpensive energy sources as the cost of renewable energy decreases and their economies grow. One of those alter-

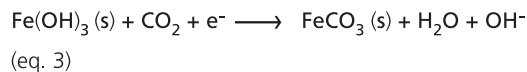
natives will be studied in the laboratory and then being developed to match field scale needs within the CDEAL consortium.

The overall goal of CDEAL is twofold: reduce the CO₂ emission into the atmosphere and to rehabilitate contaminated, acidic mine waters using carbonation. As the storage of carbon dioxide (CO₂) will be in the form of carbonate it will therefore be sustainable. Thus, CDEAL will positively contribute to the reduction of greenhouse gas emissions by CO₂-sequestration and to the goals described in articles 2 and 3 of the Kyoto-Protocol. Furthermore, as acidic mine waters belong to the largest waste streams in Europe, any significant reduction of such mine waters will result in a reduction of the total waste streams in Germany. CDEAL will only use pre-treated mine water and mine water with elevated CaO-contents due to fly ash deposition, where an excess of CaO exists and can be used to react with CO₂ to CaCO₃. In addition, iron-hydroxide sludge is available in great amounts in some parts of the open pit lakes and the waste rock piles, which can be used as reacting material as well.

Main goal of the proposed project is to investigate a sustainable low risk strategy to incorporate CO₂ into the subsurface and thus exclude it from the atmosphere. The precipitation of CO₂ and an alkaline earth metal (M) oxide or hydroxide can be written in the overall equations



Using iron hydroxides as cation source for a carbonate precipitation the following redox half reaction can be written



These storage mechanisms, the storage in the sediment and in the water phase may be applied in acidic mining lakes. The East German Lignite mining area is well suited for the demonstration of the proposed technology. CO₂ production and acidic lakes are locally concentrated. CaO is produced in combustion power plants. Over the past decades of intensive mining in the Lausitz area large amounts of fly ash and coal slurry from power plants and iron hydroxides from water treatment plants have been deposited in abandoned mines that now form lakes. Lake Burghammer contains sediment layers of these substances of several meter thickness on its bottom and was picked as field location in the proposed project. The Lausitz post mining lakes in general and lake Burghammer in particular yield a high potential for utilizing

equations 1 to 3. To utilize equation 3 it has to be investigated if lake internal or external electron donors may be used.

The proposed project comprises of two steps. In a first step the feasibility of the treatment will be investigated in the laboratory and bench scale identifying governing chemical equilibrium and rates. The second step will transform the findings to the pilot and field scale. The thermodynamic possibility for a precipitation of calcite (saturation) is given when the product of the calcium and the carbonate activities reaches the temperature dependant constant K_{sp}. The strong dependency of the appearance of the carbonate ion in solution from pH creates the need of near neutral to alkaline conditions for calcite precipitation. Given an acidic water with a typical high concentration of calcium (3 to 10 mmol/L) a base titration will lead to a saturation with respect to calcite if carbon dioxide is allowed to reach equilibrium concentrations with atmospheric conditions. Calcite precipitation will decrease the calcium concentration as long as saturation is maintained. DIC concentration increases as Ca con-



Figure 1: Aerial view of lake Burghammer (courtesy eta η Aktiengesellschaft engineering Cottbus).

centrations decreases. This shows the high potential of acidic lakes for the uptake of CO₂ into the water if the treatment is combined with a neutralisation of the water. The natural pH-buffer bicarbonate is introduced to the water and improves the chemical condition and the biological potential.

The amount of calcite that can be precipitated if only pH is raised is of course low and depends on the initial calcium concentration. Unlimited amounts of calcite can be produced if calcium and carbonate are delivered to the solution. Precipitated calcite is stored in the sediment. Lake sediments are natural sinks in the geosphere. The storage mechanism consequently is not based on technical elements but on the geochemical conditions. Calcite precipitation should not be used in lakes that have a long-time tendency of becoming acidic. Nevertheless the return to acidic conditions of a lake used for calcite storage will not result in instantaneous CO₂ release. Apart from the surface area that is too small for an effective dissolution (which has been experienced in many unsuccessful liming treatments) dissolution of calcite will be limited by calcite saturation. Iron precipitation occurring during neutralization can produce sediment layers that support a progressing exclusion of the carbonates from participating in lake internal processes as the sediment consolidates. Calcite precipitation is best applied in acidic lakes that will be neutral after an initial water treatment. Lake Burghammer is part of a storage system that will receive high fluxes of surface waters. A water treatment station at its inflow point can be used to maintain neutral conditions if necessary.

The supply of cations (Ca, Mg) is a limiting factor for the utilisation of the storage strategy. A positive effect concerning the overall carbon dioxide budget can be achieved using cations from the ashes of combustion power plants (eq.1). Whereas cations from lime ovens have previously released CO₂ this must be included in the budget.

Scientific and Technical Goals

CDEAL's scientific and technical goals comprise of several steps: in the first project phase the technology and scientific basis for the CO₂-injection into CaO-rich or Fe(OH)₃-rich substrate will be developed in the laboratory. In the second phase, this technology will be transported to the field scale, where all the necessary installations to inject CO₂ into the substrate of an open pit lake (exemplified by using the Burghammer pit lake) will be developed. In the same time, other potential sites with a CO₂-source and a CaO-rich mine water or substrate will be investigated and the potential for installing treatment schemes there will be evaluated.

During the first phase, the CDEAL consortium, namely TU Bergakademie Freiberg and DGFZ, will conduct laboratory, bench and numerical studies to develop the proper layout of the CO₂-injection into the lake's substrate. This step's goal will be to find parameters for the correct CO₂-flow or gas composition, contact time between CO₂ and CaO and the type of injection of the gas into the substrate. Furthermore, pressure and temperature dependencies as well as the correct physico-chemical characteristics of the installation will have to be tested. The final scientific goal in this first step will be to get a proper understanding of the reactions involved and the dependencies of the different parameters involved. Furthermore, a pilot scale plant shall prove that the method developed will be suitable for the field scale treatment.

From the results of the first step, the goals of the second step have to be defined. The second step's goal is to construct a field scale plant near the open pit lake. This field scale plant has to consist of a CO₂-receiver station and an injection system. Furthermore, the results of the CO₂-injection have to be monitored continuously throughout the duration of the whole project. At the final stage of the project, a technical installation at the Burghammer pit lake will demonstrate to treat the acidic mine water by using the excess CaO/Fe(OH)₃ in the lake's substrate and the CO₂.

The scientific goal of the project will be to show that a waste product (CaO , Fe(OH)_3) and a greenhouse gas (CO_2) can be used to treat acidic mine waters and therefore contribute to the environmental protection of Germany's mining landscapes. Furthermore, the method will help to decrease Germany's CO_2 budget and therefore positively contribute to the aims of the Kyoto-Protocol. Such a CO_2 -Sequestration system can also be implied in a ZEC (Zero-Emission Certificate) trading system (Kunsch et al. 2004).

The successful mediation of carbonate precipitation using native water from the post mining lake Burghammer and fly ashes and CO_2 from a nearby combustion power plant (or any other source) is dependent on a number of uncertainties that can be addressed as the main scientific and technical goals:

- *Influence of the specific mine lake water on the treatment reaction*

Water from mining lake Burghammer is characterized by high concentrations of calcium, magnesium and sulphate, a low pH, an electrical conductivity of 2300 $\mu\text{S}/\text{cm}$, total iron of around 0.2 mmol/L and an acidity of around 2-3 mmol/L. In comparison to formation waters from oil fields, the dissolved organic carbon is rather low (around 3 mg/L) resulting in a less pronounced effect of the organic acids on the acid-base system of the waters. The key parameters for the stipulation of any reaction are the activities of the involved species and the rates of the reaction. A sufficient knowledge of the occurring processes will lead to a numerical model that can simulate laboratory experiments. It is one major goal to set up such a model.

- *Influence of the ambient conditions on the treatment reaction*

Carbonate precipitation is strongly dependent on gas pressures and temperature. Depending on the water column that is to

be considered for the location of the reaction and the CO_2 fluxes that will be used, partial gas pressures and total pressure may vary. Seasonal temperature variations must be considered as well. The laboratory experiments must adapt to these conditions.

- *Influence of the technical layout of the bench and pilot scale experiments.*

Transport phenomena in the subsurface compartments will start to be of major importance entering bench and pilot scale experiments. The simulation model then needs to be multidimensional and be supplied with suitable transport parameters. The relevant compartments are the surface water (lake), the sediment and the groundwater.

Scientific and Technological Standard of Knowledge

To our knowledge, CO_2 and CaO have never been used to treat acidic mine water before, though CaO -rich fly ashes are commonly used to increase the pH of acidic mine water (e.g. Hellier 1998, Wisotzky 2001, Zoumis et al. 2000). An intensive literature review using the relevant literature databases (GEOREF, GEOBASE, FIZ-GEOL, FIZ-BERG, and ISI-Thomson) proved that there is no work using those two compounds actively for the treatment of acidic waters. Yet, there were several investigations studying CO_2 -sequestration by the use of CaO , MgO and CO_2 in different environments and CO_2 sorption on different materials, there under metal dotted CaO (e.g. Elfving et al. 1996; Anthony et al. 2000; Reddy and Smirniotis 2004).

The carbonate system in natural waters is a complex phenomenon that involves the transfer among three phases: solid, liquid and gas. $\text{CO}_2(\text{g})$ dissolves in water to form loosely hydrated $\text{CO}_2(\text{aq})$. Only a small part of it is forming carbonic acid H_2CO_3 . The dissolved species of inorganic carbon and water are produced by hydrolysis (exclusion of H^+ from hydration sheaths) of carbonic acid to form bicarbonate HCO_3^- and carbonate CO_3^{2-} . Calcite

$\text{CaCO}_3(s)$ is formed by calcium and carbonate. The number of unknowns rules the number of equations that have to be solved to describe the equilibrium of this system. As these reactions are simultaneously occurring with the speciation of the other dissolved species in a natural water, geochemical models are inevitable in predicting the effects of stipulated reactions. To the problem of »aquatic speciation« different theoretical concepts and computational strategies can be applied. Theoretical fundaments are described by Nordstrom and Munoz (1987), Stumm and Morgan (1996) or Bethke (1996) among others. Reardon (1994) gives an example of the a priori indeterminate effects of the increase in $p\text{CO}_2$ on the carbonate system:

Assume we have water at equilibrium with calcite at a specified $p\text{CO}_2$. What will happen to the pH, CO_3^{2-} , HCO_3^- , H_2CO_3^* and Ca^{2+} concentrations if the carbon dioxide pressure in the gas phase of the container is suddenly increased? Starting at the point in the reaction scheme where the perturbation occurs, if the $p\text{CO}_2$ is increased, then most definitely some H_2CO_3^* will form. In other words, the effect of increasing $p\text{CO}_2$ will be offset by the dissolution of some of this carbon dioxide into the water phase. This increase in H_2CO_3^* will result in some of it dissociating to form H^+ and HCO_3^- . Thus it can be concluded that the pH will decrease and HCO_3^- will increase as a result of an increase in $p\text{CO}_2$. Now what will happen to Ca^{2+} and CO_3^{2-} ? To determine this, the direction in which the following reaction will proceed must be determined:

$\text{HCO}_3^- \longrightarrow \text{CO}_3^{2-} + \text{H}^+$. It has been determined that HCO_3^- will increase and pH will decrease (i.e., H^+ increase), what will happen to the CO_3^{2-} concentration? At this point, it cannot be concluded which way the above reaction will proceed. If only the pH decreased as a result of the increase in $p\text{CO}_2$, then the reaction would proceed to the left and CO_3^{2-} would decrease; and if only HCO_3^- increased as a result of the increase in $p\text{CO}_2$ then the above reaction would proceed to the right and CO_3^{2-} would be increased.

For example, the dissolution of calcite can be expressed with $\text{CO}_2(g)$ by two overall reactions:



or



If Le Chatelier's principle is applied to the above two reactions to discern the effect of an increase in $p\text{CO}_2$ on the solubility of calcite, different conclusions would be drawn depending upon which reaction is selected. This merely reflects the indeterminacy of a change in $p\text{CO}_2$ on the solubility of calcite without knowing the chemical composition of the water.

This again underlines the need of geochemical modelling in describing carbonate species in a natural water. Knowing the composition of mining lake Burghammer and considering the fact that both CO_2 and CaO (H^+ decrease) are to be used, it can be concluded that the precipitation of calcite is the thermodynamically favoured reaction.

The following solution composition was analyzed in the epilimnion of lake Burghammer in 2004: pH 3.1, Na 3 mmol/L, Mg 2.5 mmol/L, K 0.5 mmol/L, Ca 9 mmol/L, C 0.2 mmol/L, S(6) 14 mmol/L, Fe 0.2 mmol/L, Cl 2.0 mmol/L.

A stepwise simulation of the equilibrium processes that might occur was performed using PHREEQC2.7. In the first step CaO and CO_2 was dissolved into the water and calcite was allowed to precipitate. CO_2 was introduced by assigning a partial pressure of 2 bar, assuming a treatment in 10 m depth and the use of a pure gas. In the second step this water was equilibrated with atmospheric CO_2 partial pressures to approximate the degassing of CO_2 from the treated water into the atmosphere (Tab. 1).

This rough calculation shows that calcite may precipitate in the same magnitude as CaO is dissolved in the water. The main influence of the partial pressure of CO_2 is the control of pH.

Tab 1: Results of PHREEQC modelling with CO₂ partial pressure of atmospheric partial pressure.

Step	CaO input	<i>p</i> CO ₂	CO ₂ flux mmol/L	Calcite		C in solution mmol/L	pH in solution
				precip. mmol/L	in solution mmol/L		
0	0	0.006	–	–	9	0.2	3.1
1	1000	2	1078	988	21	90	5.8
2	0	0.0004	-78	12	9.5	0.4	7.8

Tab 2: Results of PHREEQC modelling with CO₂ partial pressure at 0.1 bar.

Step	CaO input	<i>p</i> CO ₂	CO ₂ flux mmol/L	Calcite precip. mmol/L	Ca in solution mmol/L	C in solution mmol/L	pH in solution
0	0	0.006	–	–	9	0.2	3.1
1	1000	0.1	1006	997	12.5	10	6.5
2	0	0.0004	-6	3	9.5	0.4	7.8

Low pH favours CaO to dissolve, but hinders Calcite to precipitate and CO₂ to dissolve. A second calculation was performed with a pCO₂ of 0.1 bar (Tab. 2). Best results seem to be possible if CO₂ supply is controlled depending on the dissolution of CaO and the pH. The higher the pH will be in step 1 (the lower the partial pressures of CO₂ will be) the less CO₂ degasses unused into the atmosphere after the water equilibrates with atmospheric gases. In these calculation examples the unused fraction of CO₂ ranges from 0.6 to 7 %. Gypsum saturation was not exceeded in these examples.

The control of the mass fluxes during the reaction is a key task to optimize the efficiency of CO₂ turnover into calcite. Furthermore, biological activity is closely linked to the carbonate system. Primary production and respiration transfer carbon to and from its inorganic form into the organic form. Primary production in acidic lakes is very limited. Büttchner und Uhlmann (2004) calculated for the acidic lake 117 (Grünewalder Lauch) a carbon production rate of 78 g C m⁻² a⁻¹. Lessmann und Nixdorf (2002) have concluded from their investigation

of acidic mining lakes that inorganic carbon is a limiting factor for the primary production in these lakes. Biological activity in the lake will increase after neutralization. The uptake of CO₂ by algae will thus increase as well. Natural neutral lakes produce organic material in the order of 100 to 200 g C m⁻² a⁻¹. This carbon flux is some orders of magnitude smaller than the carbon fluxes that will be introduced by the proposed technique. At this rate no effect on the technical CO₂ flux and the calcite precipitation is expected to be visible. The effects of the CO₂ injection on biological primary production have to be monitored and quantified, yet no negative effect for the environment or the technical process is expected at this point.

Naturally occurring precipitation of calcite in lakes is a widely recognized process. It is often reported to take place as a consequence of increased primary production due to a high nutrient import and subsequent rise of pH (Klapper 1992). The precipitate can be described as a calcareous mud. During the process of precipitation very fine grains are suspended in the water creating a white bleary that is able to eliminate other dissolved constituents from

the water by co-precipitation and adsorption. The re-suspension of autochthonous calcite and the dumping of external calcite on the lake surface has been proposed to eliminate nutrients from eutrophicated lakes (Hupfer et al. 2000). These investigations show that the precipitation of calcite creates positive effects for the aquatic environment and that the precipitate is rather inert concerning the ability to re-dissolve into the water.

Description of planned work

Both problems, AMD forming and carbon dioxide emissions occur in areas where lignite mines and power plants are established. Thus, the major idea of this proposal is to solve the problems simultaneously. Since the deficit of mined coal forms open pits with lakes, that are often characterized by low pH and high sulphate concentration mainly stemming from the oxidized waste pile rocks, these water reservoirs are typical brown coal mining remains. If suspended calcium oxide (CaO), calcium-silicate, or iron hydroxide [$\text{Fe(OH)}_3(s)$] and CO_2 is added to a lake the CO_3 in the calcite (CaCO_3) or siderite (FeCO_3) will be formed from the gaseous carbon dioxide (CO_2). Thus, this is a true sustainable solution to decrease the amount of carbon dioxide emission. In particular, CaO , silicates and iron hydroxides are commonly available in waste rock piles and open pit lakes in huge amounts since ash and iron hydroxide sludge were often deposited. In combination with CO_2 from the flue gases, calcite and siderite can be precipitated in the lake resulting in a sustainable deposit of CO_2 in the subsurface and increasing pH and water quality of open pit lakes simultaneously.

Mining lake Burghammer is located 5 km east of the city of Hoyerswerda in Sachsen (Saxony). It is part of the river Spree catchment area. The mining lakes Dreiweibern, Lohsa II and Burghammer are planned to be operated as one storage pond system. The outflow out of this system will discharge from lake Burghammer into the river Kleine Spree which is a tributary to the river Spree. Today the lake is 59% filled at a water level of 104 mNN. Its final water

volume will be $36 \times 10^6 \text{ m}^3$. During the 1970ies fly ash from the Schwarze Pumpe combustion power plant has been dumped in the abandoned surface mine. During the years 2000 to 2002 a re-suspension of the dumped ashes was tested and demonstrated. An excavator on a floating platform was equipped with a suction tube and a rinsing device to suspend and dissolve sedimented ashes in the lake water. It was shown that this action was successful in neutralizing the acidic lake water. Furthermore, it was shown, that no buffer capacity could be built up in the water by this action. It was decided to postpone a further operation of the suction excavator until lake Burghammer will start to discharge water into public streams (river Kleine Spree) and to keep an instrument of controlling the effluent pH.

In the first phase of the project the feasibility of that concept will be investigated. The second phase of the proposed research project should pick up the threads of the above described re-suspension technology and combine it with the CO_2 carbonate storage technology.

The research work which will be assisted by LUG, a locally experienced consultant company, will be subdivided in two parts: part 1 will be a detailed feasibility study lasting one year. The goal of part 1 is to evaluate the general procedure investigating the thermodynamics and kinetics of the governing processes by means of laboratory experiments. If part 1 is completed successfully, part 2 will focus on the optimization of the procedure and the implementation of a pilot plant.

Thermodynamics and kinetic reaction rates of calcium oxide (CaO), calcium silicate, and iron hydroxide will be determined in laboratory experiments using natural acid lake water and synthetic flue gases with varying CO_2 concentrations between 5 and 95 vol. %. Grain size of CaO and age of iron hydroxide will be varying in the experiments to evaluate the range of reactivity.

To be more realistic, sediments from several lakes in the Lausitz lignite mining area will be

sampled from boat or by scuba diving. The amount of CaO, iron hydroxide and other components will be investigated by means of mineralogical and geochemical methods. The sediments sampled will then be used instead of pure CaO and iron hydroxide for further laboratory experiments with CO₂ injections. Besides the formation of calcite and siderite, the possible liberation of trace elements (metals, PAH) will be investigated as well as coatings on the CaO that may create certain passivity and decrease the theoretical reaction rates.

Laboratory experiments will be evaluated by means of geochemical modelling using the numerical code PHREEQC. Thermodynamic data and in particular kinetic reactions rates will be derived from the laboratory experiments in order to enable meaningful prognosis for modelling pilot scale and large scale treatment plants.

With respect to practical application, the amount of CO₂ in flue gases within the Lausitz will be determined. Furthermore, the amount of deposited ash and fly ash and their chemical composition with respect to CaO, Ca silicate, and iron hydroxide as well as trace elements and PAH will be evaluated from existing data. A comparison of distinct sites for the establishment of a pilot plant will be done and a preliminary study about engineering aspects of CO₂ elimination in the area of interest will be performed in order to evaluate the overall performance of the method and acceptance by shareholders. This includes the evaluation of different suspension and CO₂ injection techniques, however, only by means of theoretical considerations and calculations in conjunction with the partner DGFZ.

The second part deals with the optimization of the procedure and investigations in the lake in order to find suitable sites for a pilot plant. Those results will be an input for the necessary permits that will be compiled by DGFZ. Further laboratory and in lake experiments in the second and third year will investigate the CO₂ gas dissolution behaviour

with respect to the total pressure, which is a function of water depths and the partial pressure. According to average water depths in open pit lakes, the total pressure will be varied between 1 and 4 bar. Dissolution of carbon dioxide will vary not only with partial pressure and with total pressure but as well temperature and bubble size. Thus, by varying these parameters, optimal boundary conditions will be found. Because formation of carbonate is in particular a matter of pH, this is a master variable controlling the overall performance of the treatment together with the reaction rates. Reaction rates are depending on reactive surfaces. Thus, the surface of the carbon oxide and iron hydroxide available in the open pit lake might be a limiting factor. Investigations will thus focus on this item and simple methods and techniques will be developed to enlarge the surface of the buried carbon oxide and iron hydroxides.

Different redistribution and injections techniques will be tested and evaluated in close cooperation with the DGFZ and the industry partners and based on an already existing fly ash suspension facility. Design and construction of a pilot plant itself will be done within the second CDEAL project (by DGFZ). Laboratory and field experiments will be evaluated by means of geochemical modelling using the numerical code PHREEQC, PHAST (3d reactive transport code) and MODGLUE (a pit lake model). Thermodynamic data and in particular kinetic reaction rates will be evaluated in order to enable meaningful modelling of real world problems taking into account all relevant boundary conditions.

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