

**Sealing of rock joints by induced  
calcite precipitation. A case study  
from Bergforsen hydro power plant**

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January 1991

SEALING OF ROCK JOINTS BY INDUCED CALCITE PRECIPITATION.  
A CASE STUDY FROM BERGEFORSSEN HYDRO POWER PLANT

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January 1991

This report concerns a study which was conducted for SKB. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

Information on SKB technical reports from 1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26), 1981 (TR 81-17), 1982 (TR 82-28), 1983 (TR 83-77), 1984 (TR 85-01), 1985 (TR 85-20), 1986 (TR 86-31), 1987 (TR 87-33), 1988 (TR 88-32) and 1989 (TR 89-40) is available through SKB.

SEALING OF ROCK JOINTS BY  
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Ludvika 91-01-04

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## ABSTRACT

The possibilities of sealing rock fractures by injecting water saturated with calcite solution, and hereby inducing a calcite precipitation inside the fracture, is investigated. The way of reaction and the amount of calcite precipitation will depend on the saturation of calcium carbonate in the water, the temperature, the pH and the CO<sub>2</sub>-pressure.

There is experience of lime-saturated water injection in the rock foundation below the dam at Bergforsens power plant (1955-1968). It was observed that the consumption of injected lime water decreased with time.

A possible reason to the decrease in lime water consumption is that calcite has precipitated such that the permeability of the rock in general is lowered. Another explanation to this could be that calcite precipitation is concentrated to the fractures surrounding the injection holes, thus preventing the lime water from penetrating further into the rock.

It is recommended that further studies of the fracture fillings in drill cores from Bergforsen is performed. The aim of such a study should be to determine the extent of induced calcite precipitation and to investigate its chemical and physical properties.

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## 1 BACKGROUND

In the research for safe solutions of the radioactive waste disposals one part is rock sealing research. The rock surrounding a future storage should be tight enough to ensure an extremely low ground water flow in the area. Therefore many possible sealing techniques are now under investigation (e.g. Pusch et.al. (1985), Stripa Project TR 87-12).

One possible way of sealing the fractured rock could be an artificial imitation of the natural calcite infillings. If this was possible it would have the advantage of being similar in composition to natural fracture filling materials which has shown to be stable and persistent for a long time.

This study concentrates on calcite, although other mineral could be possible as fracture filling material. Calcite seems to be the material which precipitation mechanisms is most known. The chemical conditions controlling calcite precipitation, cristallization and solubility is presented in this report.

A contributing factor to the initiation of this study was the fact that there exists some experience from injection of water saturated with lime in the rock foundation of a hydro power dam at Bergeforsen, Sweden. The experiences from Bergeforsen is summarized in this report.

## 2 EXPERIENCE FROM BERGEFORSEN HYDRO POWER STATION

### 2.1 General

The Bergeforsen Hydro Power Station started the production in 1955. The earthfill dam is 400 meters long and has a highest heighth of 35 m. The dam is founded partly on moraine partly on bedrock. The geological conditions in the area were discovered to be very complex. The bedrock is composed of a gneiss granite intruded by a series of dykes, so called Alnö dykes after the type locality the island of Alnö, not far from Bergeforsen (Eckermann, 1961).

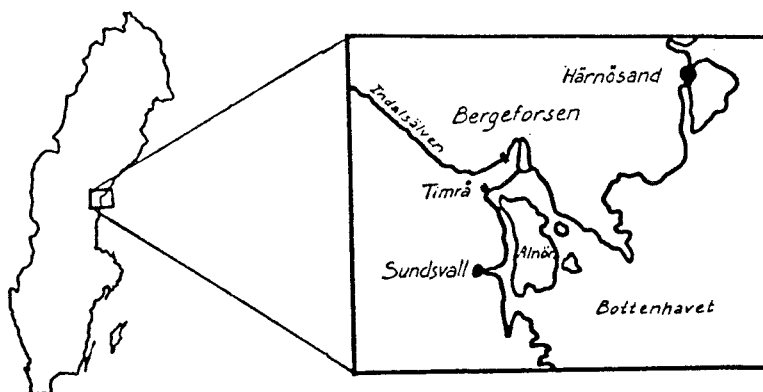


Figure 2.1 Location of the Bergeforsen hydro power station.

According to Eckermann (1961) and Barth (1962) the original intrusion of Alnö consisted of a carbonated alkali peridotite magma (kimberlitic) rich in fluorine but comparatively poor in water. Due to differentiation, reaction with the country rock and varying CO<sub>2</sub>-pressures the great variety of Alnö dykes including carbonatite, alnöite and kimberlit were formed.

The weathering process of the Alnö rocks is controlled mainly through a reaction between calcium carbonate, water and carbon dioxide. The disintegration of the dykes will increase with greater ground water flow and lower pH-value. Some of the dykes as well as the gneiss granite, which locally is altered due to the intrusions, contain montmorillonite which swells and decomposes when hydrated.

The unaltered, fresh rock in the foundation has a strength which is sufficient to sustain the loads from the dam structure. Since the joint system together with the dykes could result in high permeability of the rock mass there was a risk for fast destruction of the dykes. This would have led to low compressive strength and extensive compressibility.

To insure a good stability for the dam a number of measures were taken to tighten the rock mass in the foundation. These measures are described in figure 2.2 and consist of a combination of grouting, bitumen injection and injection of dry-slaked lime soluted in water. The design of the latter system will be described in more detail in the following section.

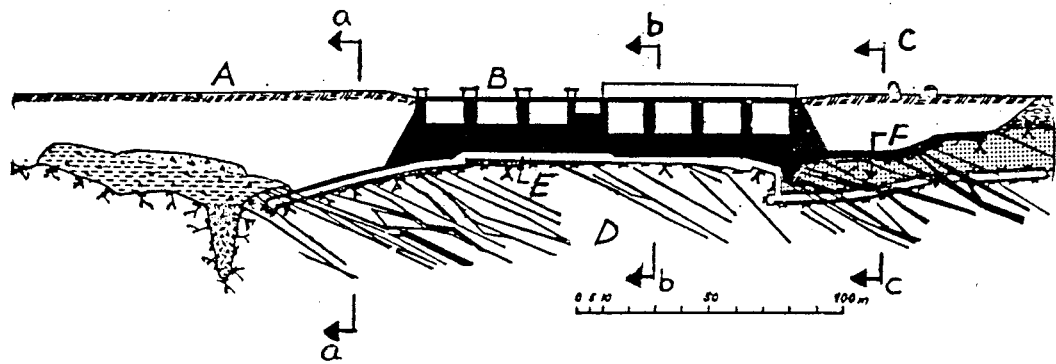
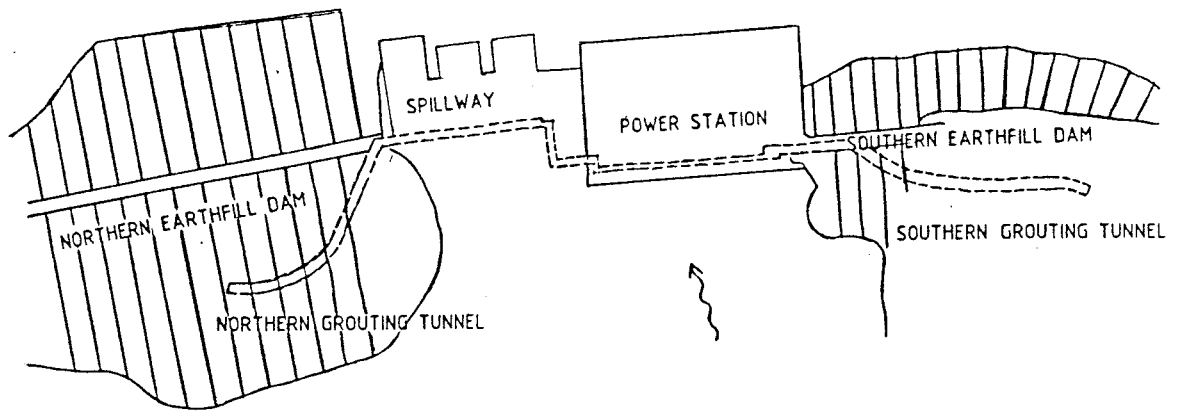
## 2.2 Injection of water saturated with CaO

A CaO saturated water was produced by mixing dry-slaked lime (Kronkalk Special) into water from the river. During the summer when the temperature in the river was too high, tap water was used in the production. The temperature was held below 8°C.

The production was controlled automatically via registration of the "lime water" reservoir level. About 4 kg of lime was added to 1 m<sup>3</sup> water. The saturation was held at a constant level of 98-100 % at 6-7°C, controlled once a day.

The lime water was filtered after mixing and was fully clear when injected to the rock. Without filtering the injection system would soon be clogged up. The total consumption to the system was measured once a day and for each group of holes once a month.

The water is pressed into the system by a constant water head from the reservoir in a building on the south side of the dam. The boreholes are  $\phi$  40 mm with c/c 1 m in a row between the two rows of cement grouting holes, see figure 2.2. Below the northern dam and below the intake section the holes are 20 m deep (sections a and b) while they are 6.5 m from the tunnel floor below the southern dam (section c). The total injection system has involved around 4500 borehole meters distributed on about 300 boreholes.



- |                            |                               |
|----------------------------|-------------------------------|
| A. Northern earthfill dam  | E. Grouting tunnel            |
| B. Intake and spillway     | F. Bitumen injection          |
| C. Southern earthfill dam  | G. Grouting curtain           |
| D. Bedrock with Alnö dykes | I. Lime water injection holes |

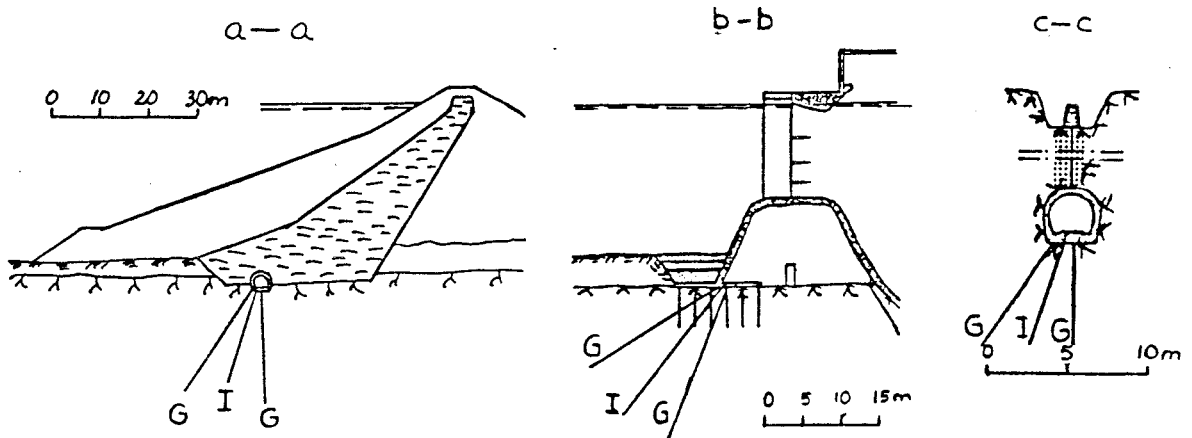


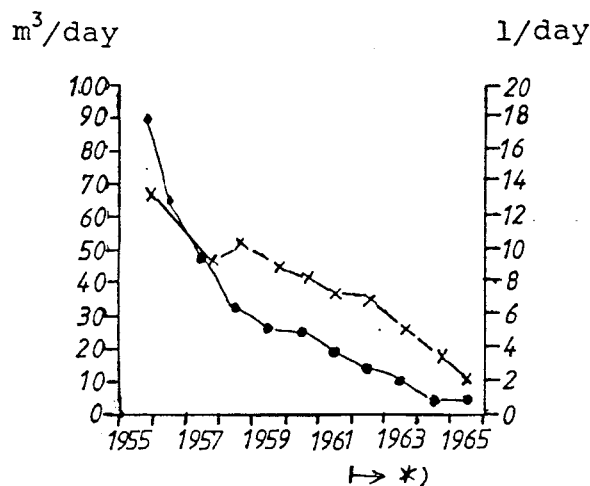
Figure 2.2 Sealing measures at the Bergforsen hydro power station (modified after Sällström, 1967).



## 2.3 Observations

### 2.3.1 Lime water consumption

The consumption of lime water in the injection system was continuously measured and it showed a clear decrease, see figure 2.3. Also the consumption in the bitumen injection system is given in this figure as a comparison.



- Lime water consumption, m<sup>3</sup>/day
- x Bitumen consumption, l/day

\*) Lime water shut off in some testsections

Figure 2.3 Consumption of lime water during 1955-1965 (after Sällström, 1967).

To investigate the possible clogging effects of the injection boreholes some of the holes were redrilled and brushed. The lime water consumption in these holes was not increased after the cleaning.

Since the clogging effect might not have been removed by cleaning measures another test was performed by drilling new boreholes between the old holes in the northern part of the dam. The lime water consumption in the new drilled holes were only slightly greater than in the old ones, thus indicating a decrease in rock permeability in general. Unfortunately this test was performed in a part of the injection system that had very small consumption of lime water compared to other parts.

### 2.3.2 Drill core investigations

About 300 diamond drill holes have been drilled in the bedrock below and around the hydro power station. Most of the drill cores are still available. The present control program for the dam includes a new drilling program every five years through the Alnö dykes to observe possible change in the rock condition.

Due to the observed possible tightening effects of the induced calcite infillings an investigation on this subject was performed 1959, after three years of lime water injection, by professor von Eckermann. He found that calcite was precipitated in the fractures but mainly in the very near surrounding of the injection holes. The longest distance from observed calcite to nearest injection hole was one meter and in those cases in crushed zones. He estimated the maximum volume of precipitated calcite to 1400 kg which corresponds to less than 1 % of the total injected amount of CaO.

In this drill core investigation von Eckermann (1959) concluded that sealing of the rock could had not with certainty been gained on a distance longer than 3 cm from the lime water injection hole, but occurred sporadically at distances up to one meter. The maximum depth of the observed calcite sealing was 6 m below borehole collar. In crush zones the calcite precipitation extended longer from the bore hole.

In the drill core investigations performed more recently the calcite precipitation has not been studied in particular since they have been focused on the condition of Alnö dykes. Therefore observations of calcite precipitations is rarely recorded or commented.

### 2.3.3 Water pressure measurements

The ground water pressure has been observed in a great number of observation holes. From the pressure distribution it can be seen that the grouting curtains has the intended tightening effect and influence the pressure distribution at least on the southern side. The water pressure difference between the upstream and the downstream side of the grout curtain is around 15 m in the central parts of the dam. The water pressure situation especially on the northern side is also influenced by the deep preglacial furrow.

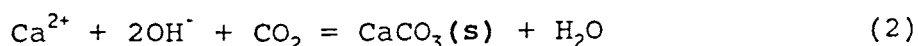
Water loss measurements has also been performed in the grout curtain showing that the permeability is low ( $Q < 0.03$  l/min m bar). Control measurements in the curtain from 1981 does not show any change in permeability.

### 2.4 Discussion of the experience from Bergforsen

The aim of the lime water injection at Bergforsen was no to get a precipitation in the first place but to protect the Alnö dykes from more aggressive water. The calcite precipitation was an effect discovered as the the injection went on. Therefore the injection system applied at Bergforsen can not be expected to be the optimum for getting calcite precipitation.

The injected lime water was composed of CaO, which was dissolved in water (reaction 1). Precipitation of calcite took place as the result of a reaction with CO<sub>2</sub> dissolved in the groundwater (reaction 2).





The low percentage of precipitated calcite with respect to the injected amount of lime can be explained partly by a relatively low  $\text{CO}_2$  content in the groundwater, partly by a nonsufficient oversaturation of lime. The  $\text{CO}_2$  giving the precipitations at Bergeforsen probably originates from the Alnö rocks holding  $\text{CO}_2$ -inclusions.

The decrease in lime water consumption may be explained either by calcite precipitation around the injection holes or by sealing of the whole rock mass between the holes. The observations from the new drilled injection holes indicate that some sealing of the whole fracture system has taken place along the sealing curtain. Also water loss measurements below the dam show that the permeability is low.

If there has been any other sealing effects than from calcite precipitation, for example from rock or grout material transported with the flowing water, this can not be discovered exclusively from lime water consumption observations.

### 3 CALCITE

Calcite is a common fracture filling mineral. It can be crystallized within a large temperature interval including temperatures prevailing at present in the bedrock.

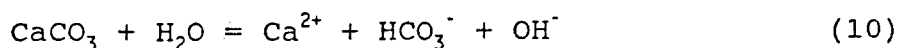
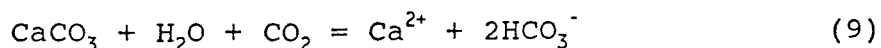
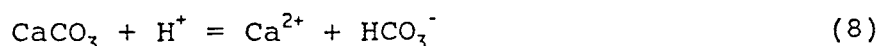
#### 3.1 Dissolution and precipitation

Dissolution and precipitation of calciumcarbonate (calcite) are to a high extent controlled by the carbondioxid system. In the system  $\text{CO}_2/\text{H}_2\text{O}/\text{CaCO}_3(\text{s})$ , the following reactions are of importance:



Dissolution and precipitation are also depending on variation in temperature. The solubility of calcite decreases with increasing temperature. This is illustrated in Figure 2.1, where the solubility product of calcite is plotted against temperature. An increase in temperature by  $80^\circ\text{C}$  means that the solubility decreases by 10% compared with the solubility at  $0^\circ\text{C}$ .

Dissolution of calcite follows in general three reaction paths:



The different reactions dominate under different chemical conditions. This is illustrated in Figure 3.2. The marked area shows the chemical conditions where two or more reactions co-operates.

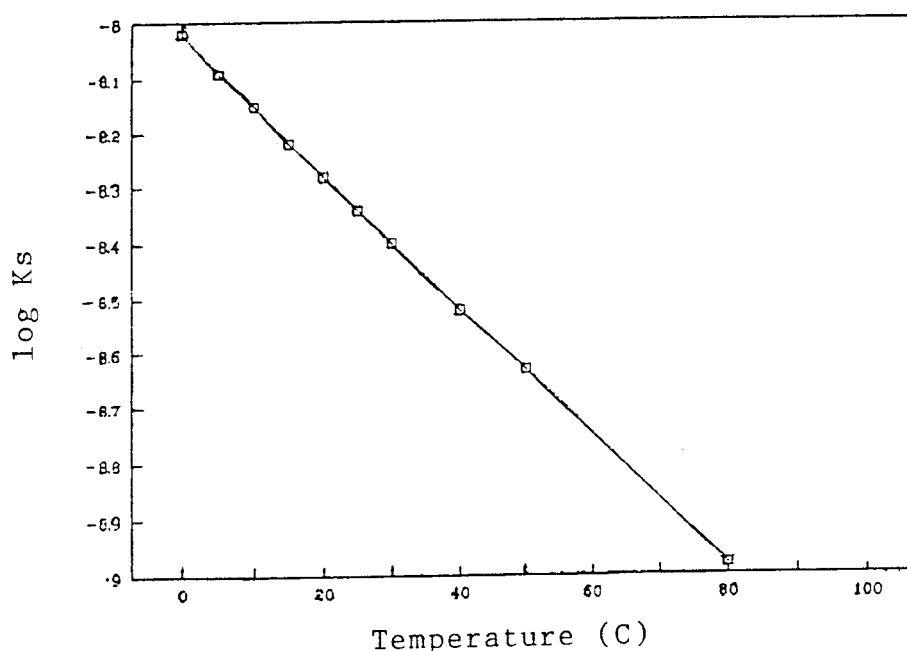


Figure 3.1 Temperature dependence of the solubility product.

Under conditions with low pH reaction (8) controls the calcite dissolution. The dissolution rate depends on the transport rate of dissolved  $\text{Ca}^{2+}$  and  $\text{HCO}_3^-$  ions. During dissolution in a stagnant aqueous system, a boundary layer of dissolved  $\text{Ca}^{2+}$  - and  $\text{HCO}_3^-$  ions will be established around the mineral surface. The boundary layer is also established in a stirred aqueous system, but to a lower extent. Mass transport through the boundary layer is due to diffusion. If the reaction rate is higher than the diffusion rate through the boundary layer, the dissolution process will be controlled by the diffusion. If the reaction rate is lower than the diffusion rate, the dissolution process is controlled by reaction kinetics.

Calcite dissolution is directly influenced by a high  $\text{CO}_2$  partial pressure, as indicated by reaction (9), and indirectly influenced through reaction (8) by an increase in  $\text{H}^+$  concentration, as the result of  $\text{CO}_2$  buffering (reaction 4 and 5). A high  $\text{CO}_2$  partial pressure increases dissolution in both situations. At higher pH-values ( $\text{pH} > 7$ ), reaction (9) or (10) are the dominating dissolution process dependent upon the  $\text{CO}_2$  partial pressure.

In general the dissolution of calcite is faster for fine grained than for coarsed grained varieties. The particle state at which calcites exists is therefore of relevance.

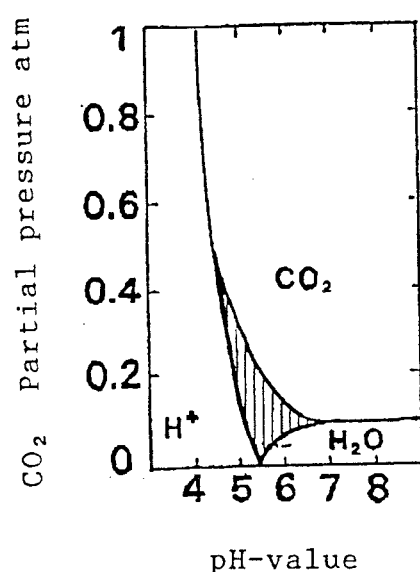
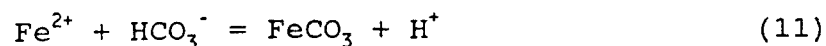


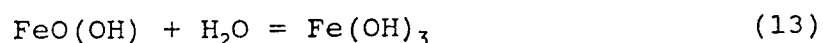
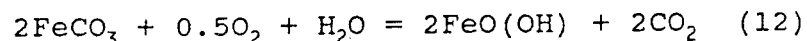
Figure 3.2 Dominating dissolution process under different Chemical conditions. (Sverdrup and Warfinge, 1987).

In the boundary layer around the calcite mineral, a high pH zone is developed, which produces a favourable chemical environment for the precipitation of metalhydroxides on the mineral surface. This occurs, to a large degree, in extremely low flow conditions, such as at the bottom of lakes. The precipitation of metalhydroxide decreases the dissolution rate. The precipitation of ironhydroxide is the most common, but aluminium and manganese can also develop strong precipitations.

The precipitation of iron onto calcite probably occurs through the following reactions:



Under the influence of oxygen,  $\text{FeCO}_3$  can be oxidized to



$\text{FeCO}_3$  has a solubility product which is about two orders of magnitude lower than the solubility product of calcite. In practice, this implies that  $\text{FeCO}_3$ -precipitation can develop at  $\text{Fe}^{2+}$ -concentrations around 10 mg/l (Swerdrup & Warfinge, 1987).

### 3.2 Crystal formation

Various processes are involved in the formation of a solid phase from an oversaturated solution. Usually three steps can be distinguished (Stumm & Morgan, 1981).

- 1) Interaction between ions or molecules leads to the formation of a cluster and formation of new centers (nucleation) from which spontaneous growth can occur.
- 2) Deposit of material on the new centers and formation of crystallites (crystal growth)
- 3) Large crystals may eventually be formed from fine crystallites.

The new centers will not be formed until a certain degree of supersaturation, a critical supersaturation, has been achieved. At a high degree of supersaturation, the nucleation rate is so high that the precipitate formed consist mostly of small crystallites, which can be in amorphous phase. The precipitate will in this case consist of an amorphous gel.

The predominant formation process in natural waters is heterogeneous nucleation. The formation process is initiated by heterogeneous solid substrates. For example, inorganic crystals, clays and sand can serve as heteronuclei. If the surface of the heteronucleus matches well with the crystal, the nucleation may take place at a lower concentration on a heterogeneous than in a homogeneous solution.

The crystal growth can be inhibited by trace concentrations of for example orthophosphates, polyphosphates,  $\text{Mg}^{2+}$  and organic material. Interference of Magnesium in the crystallization process of calcite, will also change the solubility. Mg-calcites are considerably more soluble than pure calcite.

## 4 DISCUSSION

Three problem areas can be defined for the practical applications of a fracture sealing technique based on induced calcite precipitation:

- 1) Dissolution of calcite to oversaturation
- 2) Control of calcite precipitation
- 3) Stability of calcite precipitation

### 4.1 Dissolution of calcite to oversaturation

To get a calcite precipitation, the injection solution must be oversaturated to a certain degree, the critical oversaturation. The oversaturation needed has in theoretical studies been estimated to be in the order of 10 - 100 times (Stumm and Morgan, 1981). Dandurand et al. (1982) found that a oversaturation in the order of 10 times was required to get a calcite precipitation (Tullborg & Larsson, 1982). Note that the water at Bergeforsen was saturated only up to 98 %.

### 4.2 Control of calcite precipitation

A controlled calcite precipitation implies that calcite solution is transported to a certain point, at which calcite precipitation and crystallisation occurs according to a controlled procedure. If the mechanisms for precipitation can not be controlled, a precipitation of the injected calcite solution may occur only locally near the injectionpoint.

One way of controlling the precipitation could be to keep the CO<sub>2</sub>-pressure high at the injection hole. The solution is thus kept oversaturated with calcite until the CO<sub>2</sub>-pressure has dropped at a certain distance away from the hole.

The stability of the precipitated calcite in the fracture should be guaranteed. Therefore according to the above discussion, it is important that the precipitated calcite crystallizes relatively quick. This leads to a larger grain size, which decreases the dissolution rate. Other important factors are the concentration of other ions in the groundwater, the temperature and the fracture geometry.

### 4.3 Stability of calcite precipitation

The stability of calcite precipitation is mainly dependent on the chemical composition of the surrounding groundwater. To get a stable calcite precipitation must the natural groundwater at the site in question be saturated with respect to calcite.

Analyses of deep groundwaters (Jacks, 1978 and Allard et. al. 1983) shows that the groundwater from these sites, were generally

saturated with respect to calcite.

Around a radioactive waste repository the temperature of the host rock will increase. This is a positive factor for the stability of calcite precipitation since, as mentioned earlier, the solubility of calcite decreases with increased temperature.

## 5 RECOMMENDATIONS

### 5.1 Investigation of drill cores from Bergeforsen

An investigation of the drill cores from Bergeforsen with respect to calcite precipitation should have a threefold aim:

- 1 To determine if calcite fracture fillings existed in the area before the lime water injection started.
- 2 To estimate the extent of calcite fracture filling existing in the area today and describe how it is distributed.
- 3 Define the chemical composition and texture of the precipitations found in the fractures. Show possible differences between natural and induced calcite.

These aims could be reached by the following activities:

- aim 1: Geological survey of 10 cores in the area drilled before the construction of the dam. The type of fracture fillings is observed and samples are taken if calcite or a similar infilling is found.
- aim 2: Geological survey of 10 - 20 drill-cores drilled around the lime water injection holes, at different distance, both upstreams and downstreams. The location and thickness of each observed precipitation is recorded.
- aim 3: Samples of calcite are taken from the drill cores. If it is possible samples should also be taken from precipitations in the injection system, which is now shut off, for example from the container. These samples should be analysed in microscope and with x-ray diffraction if required. The differences and similarities in structure between samples from natural fracture filling, "induced" calcite filling and calcite from the system, should hence be observed.



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#### ACKNOWLEDGEMENT

Critical review and fruitful comments on the manuscript by Jüri Martna, Carljohan Hardenby, Stig Sällström and Lars Lindström is acknowledged.

# List of SKB reports

## Annual Reports

1977-78

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### **The KBS Annual Report 1979**

KBS Technical Reports 79-01 – 79-27

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TR 86-31

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## Technical Reports

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#### **Description of geological data in SKB's database GEOTAB**

##### **Version 2**

Stefan Sehlstedt, Tomas Stark

SGAB, Luleå

January 1991

TR 91-02

#### **Description of geophysical data in SKB database GEOTAB**

##### **Version 2**

Stefan Sehlstedt

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TR 91-03

#### **1. The application of PIE techniques to the study of the corrosion of spent oxide fuel in deep-rock ground waters**

#### **2. Spent fuel degradation**

R S Forsyth

Studsvik Nuclear

January 1991

TR 91-04

**Plutonium solubilities**

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Spain

February 1991

TR 91-05

**Description of tracer data in the SKB  
database GEOTAB**

SGAB, Luleå

April, 1991

TR 91-06

**Description of background data in the SKB  
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**Version 2**

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March 1991

TR 91-07

**Plutonium solubilities**

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April 1991

TR 91-08

**Overview of geologic and geohydrologic  
conditions at the Finnsjön site and its  
surroundings**

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January 1991

TR 91-09

**Long term sampling and measuring  
program. Joint report for 1987, 1988 and  
1989. Within the project: Fallout studies in  
the Gideå and Finnsjö areas after the  
Chernobyl accident in 1986**

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December 1990