Investigations on the swelling behavior of pure anhydrites

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ABSTRACT: The swelling ability of anhydrite can be a major problem during tunnel construction. It's a fact that under humid atmospheric conditions every natural anhydrite ($CaSO_4$) sooner or later dissolves, or alters to gypsum ($CaSO_4*2H_2O$), which coincides with a calculated volume increase of 61 %. The swelling capability depends greatly on the type of anhydrite itself. In order to explain and verify the differences in the swelling behaviour of pure anhydrite rocks, different anhydrite samples were investigated. The results show that the "maturity" of the anhydrite rock provides the decisive difference. The maturity is herein reflected in a larger former rock cover which led to high temperature and stress conditions. That again produces a massive rock with large crystal grains and a relatively low specific surface area. The result is a material with a low reactive surface area and thus more or less inert ("sluggish") to swelling.

1 INTRODUCTION

1.1 *Problems with anhydrite swelling in tunneling*

Many tunnel projects of the past had to deal with the swelling capability of anhydrite. The prediction of the anhydrite swelling potential was thereby often difficult. In hindsight we now know that some anhydrites showed less swelling as expected (Tab. 1). Then again some anhydrites showed strong swelling pressure and swelling strain that resulted in floor heave and in damaging of the invert (Tab. 1). The anhydrite bearing rocks encountered in tunnel projects are usually investigated more or less intensive on anhydrite swelling. However to date comparisons between anhydrites from different formations have rarely been conducted. The question arose whether and why pure anhydrites differ in their swelling capacity and how these differences could be investigated. An additional aim of this study is to develop tools for a faster and more reliable prediction of the swelling behavior of pure anhydrites.

Table 1. Examples of anhydrite swelling in tunneling. (Erichsen & Kurz 1996*, Paul & Wichter 1996**, Laabmayer et al. 1996***, Spaun, pers. comm.****)

Tunnel	geol. formation	swelling strain [mm] / pressure [MPa]
Schanz*	Gypsum Keuper	1500 / 6-9 (calculated)
Wagenburg**	Gypsum Keuper	340 / 3.4 (measured)
Achberg***	Haselgebirge	no swelling measured so far
Tauern****	"Schieferhülle"	no swelling measured so far

1.2 *Principles of anhydrite swelling*

In contact with water every anhydrite dissolves or alters to gypsum. The basic principle of this chemical transition is shown in Figure 1. The 60.8 % volume increase from anhydrite to gypsum can be calculated from the solids. It is irreversible under atmospheric conditions.

	anhydrite 1 mole $CaSO_4$	+	water 2 mole H_2O	→	gypsum 1 mole $CaSO_4*2\ H_2O$
M [g/mol]	136,1		36,0		172,1
ρ [g/cm³]	2,95		1,00		2,32
V [cm³]	46,2		36,0		74,3
V [%]	100,0		77,9		160,8

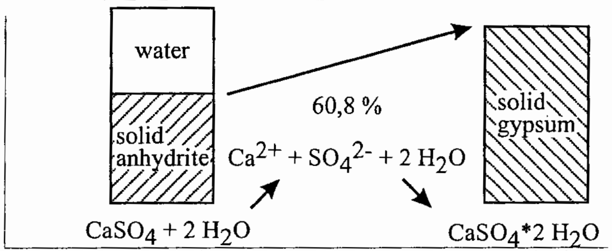

Figure 1. Chemical and physical basics of the anhydrite-gypsum conversion (M = molar mass, ρ = density, V = volume) (modified after Amstad & Kovari 2001).

Looking closer, this equation is quite simplified since the reaction involves a solution and a crystallization process. Therefore we have to deal with two processes. The first step is the solution of anhydrite;

the second step is the crystallization of gypsum (Fig. 1). Both steps are triggered through the different saturation concentrations of anhydrite and gypsum. Their value is dependent on temperature, pressure and the occurrence of foreign ions. Constant and ideal conditions apply only to a closed system in the lab where no calcium or sulfate ions are lost, the temperature is more or less constant and the perfect amount of water is always available. In contrast in nature there is always an open system with varying temperatures and ions.

2 TESTED MATERIAL

The investigations focused on anhydrite bearing formations in Germany and in the Alps. Because of the instability of anhydrite under humid conditions, it is not easy to get fresh samples from the earth's surface. It was possible to collect underground samples near Stuttgart, in the Inn and the Salzach valley. In the following the rocks are described in detail.

2.1 *Gypsum Keuper*

The lower parts of the middle Keuper (middle triassic) in south Germany and north Switzerland are called Gypsum Keuper. This formation is composed mostly of gypsum, anhydrite, clay, silt, marl and carbonate layers. The sedimentation of the 100 m to 150 m thick geological unit took place around 230 ma to 225 ma before today. The sediments were deposited in large alluvium planes with hypersaline lakes. Occasional marine ingressions flooded the area (Bachmann & Brunner 1998). The sample material was taken from an exploration drilling core near Stuttgart in Baden-Wuerttemberg. It is part of the 30 m to 60 m thick middle gypsum horizon. Since the samples were taken at a depth of around 50 m, the material is fresh and unaltered.

The rock consists of a dark gray and fine grained anhydrite which is interlayered with black claystone. The layers are between 0.2 cm and 3 cm thick. The aim of the study was to compare different pure anhydrite rocks. Therefore the anhydrite layers had to be excluded from the surrounding claystones by means of a hammer and a stone saw.

2.2 *Reichenhall Formation*

The Reichenhall Formation is part of the alpine triassic in the northern calcareous alps. It has been dated at 245 ma to 240 ma which is equivalent to the anisian stage. The marine deposits were formed on the continental shelf of the thetys ocean. The formation is very heterogeneous and consists mostly of dolomitic limestones, sandstones and rauhwacks. Nevertheless the warm climatic conditions lead to periodical evaporation of the seawater which resulted in the deposition of gypsum and anhydrite. The thickness of this geological unit varies between zero and a maximum of around 480 m in the eastern Karwendel mountains (Schenk 1967). The formation frequently shows tectonic disturbances.

The material from the triassic Reichenhall Formation was collected at a tunnel construction site in the Inn valley. The more or less pure anhydrite appears in massive breccias and laminated variations. It is dark gray and medium grained.

2.3 *Haselgebirge*

The alpine Haselgebirge formation is a saliniferous formation at the base of the northern calcareous alps. It was formed around 260 ma to 250 ma ago in the upper permian and lower skythian stage. The sediments were deposited under calm sabkha conditions on the shelf of the thetys ocean.

Today the alpine Haselgebirge appears as a big chaotic breccia. This deformation is due to the alpine orogenesis. The plastically deforming evaporates where transported to the north and act as sliding planes. In the course of this movement the original sedimentary structures were highly disturbed.

The sample material was collected at a quarry near Golling in Austria. The massive anhydrite is bluish-gray and coarse grained. A very good description of this anhydrite is given by Wiesheu (1997). He found that the Haselgebirge anhydrite is slightly metamorphic due to the high former stress and temperature conditions which occurred during the alpine orogeny.

2.4 *Different history of the samples*

As indicated earlier, the samples originate from different regions and geological units. Using published data it was possible to estimate the former rock cover of each formation. The differences between the anhydrite samples concerning the former rock cover is shown in Figure 2. The youngest anhydrite tested in this research is from the Gypsum Keuper and has undergone a maximum overburden of around 1000 m in its geological history (Geyer & Gwinner 1968). The Reichenhall Formation on the other hand had a former rock cover of approximately 5000 m (Gwinner 1971) and the rocks of the Haselgebirge had even been buried into depths of about 10,000 m (Wiesheu 1997) (Fig. 2).

The varying former rock cover lead to different temperature and stress conditions in the rock. Under normal conditions the temperature increases around 33 °C and the pressure around 25 MPa every 1000 m of rock cover. Ascending temperature and pressure conditions result in recrystallisation of the anhydrite which generally leads to coarser crystal grains.

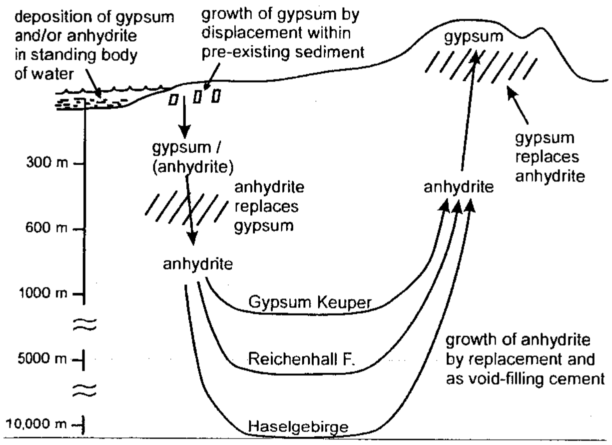

Figure 2. Schematic diagram illustrating the gypsum-anhydrite-gypsum cycle (according to Murray 1964). The varying maximum former rock covers of the investigated anhydrites can be seen.

3 MINERALOGICAL INVESTIGATIONS

3.1 *X-ray diffraction*

X-ray diffraction analysis was used to determine the qualitative mineralogical composition of the samples. In this method the diffraction of monochromatic X-rays on the surface of a crystal lattice produces varying reflection intensities (X-ray reflexes) at varying angles. These reflexes are measured and typical reflexes can be determined for each mineral, depending on the dimension of the spacing from the lattice planes (d-value). The identification of the mineral associations contained in the samples occurred by the means of the d-values and the characteristic lines of the diffraction (reflex) via an identification program.
All three anhydrite samples show similar peaks for anhydrite. They do not differ in their mineralogical character concerning X-ray diffraction

3.2 *Thin section*

The thin section analysis confirmed the macroscopic observations. The size of the anhydrite crystals increases with the former rock cover (Fig. 3). This simplified coherence is not always admissible but it seems valid in general, as it can also be seen at Reimann (1991) who gives a large overview of anhydrite deposits around the world. In Figure 3 a thin section of a relatively fine grained anhydrite from the Gypsum Keuper (GK) is shown. In contrast to that the grains get larger and larger in the samples of the Reichenhall Formation (RF) and the Haselgebirge (HG).

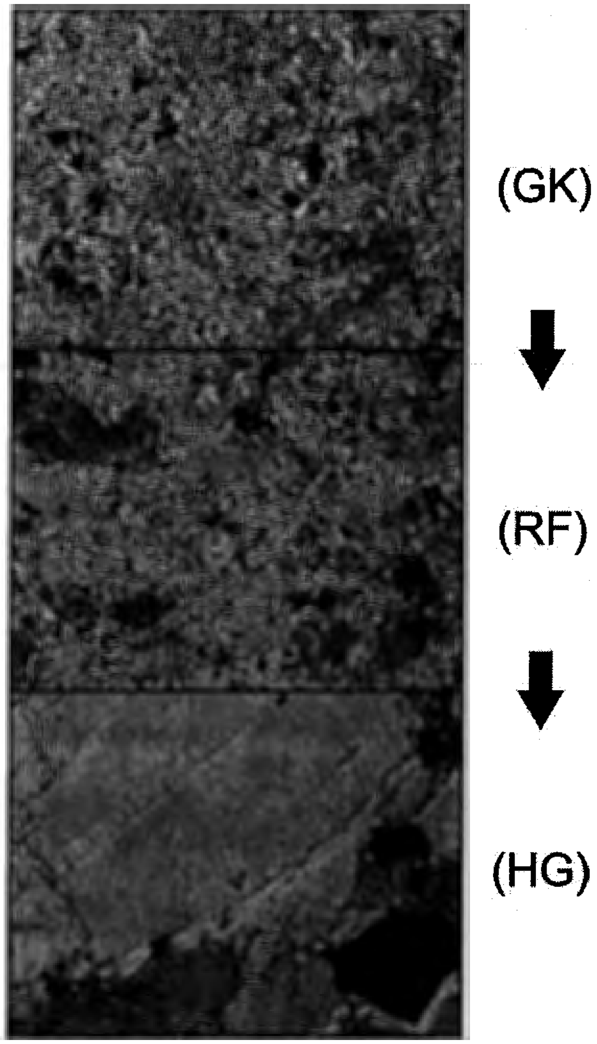

Figure 3. Thin section images of Gypsum Keuper (GK), Reichenhall Formation (RF) and Haselgebirge (HG) anhydrite samples, which show the typical increase in grain sizes from top to bottom. Crossed nicols, width of each picture circa 0.65 mm (modified after Rauh et al. 2006).

3.3 *Scanning electron microscope (SEM)*

The scanning electron microscope was used to examine the micro-structure of the samples. Special attention was paid to the texture and the surface formation. In general the Gypsum Keuper sample is more porous. These pores are crystal lattice failures and are possible water channels and weak points concerning the crystal stability. In contrast to that, the sample from the Haselgebirge appears compact and impervious. With the following SEM images (Fig. 4) it is possible to visualize the differences in the specific surface area. All three photos have the same enlargement of × 2000 and a picture width of around 56 µm each. From top to bottom the crystals shown increase in size and, even more important, they are better crystallized. Because of the good cleavage of the anhydrite the described body structure doesn't change even if the material is grinded.

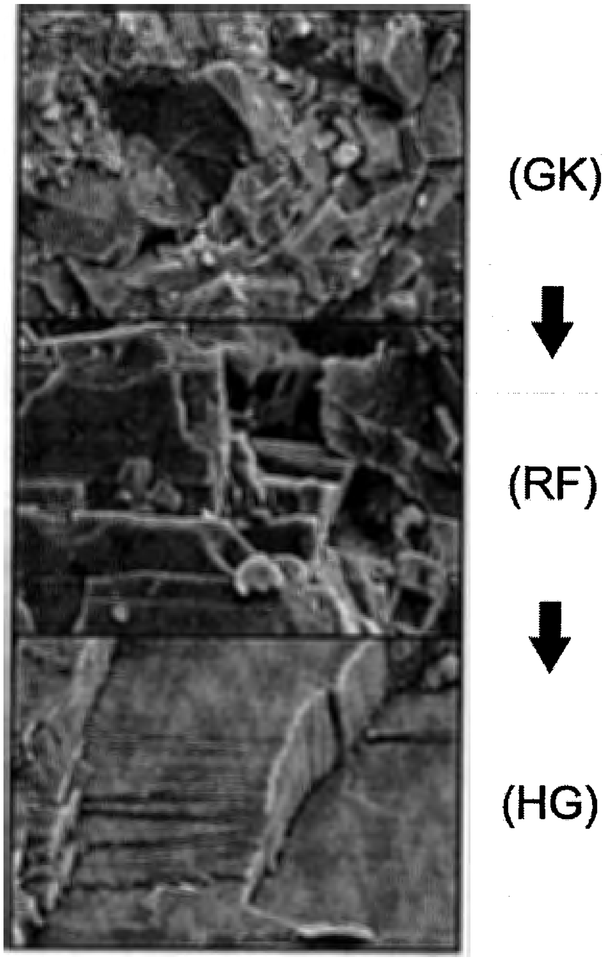

Figure 4. SEM images of Gypsum Keuper (GK), Reichenhall Formation (RF) and Haselgebirge (HG) anhydrite sample. Typical decreasing in surface area and -roughness from top to bottom. Each picture width approximately 56 µm (modified after Rauh et al. 2006).

3.4 *Specific surface area*

The air permeability method after Blaine was used to make a statement for the specific surface (quantity based surface) of the different anhydrite samples (DIN EN 196-6 1990).
The samples had to be all prepared in the same way before measuring the specific surface. In the test, air is sucked through the anhydrite sample, thereby recording the time that a certain amount of air needs to pass the sample. Using the measured time and some instrument constants the specific surface (Blaine-Value) can be calculated (Tab. 2).

Table 2. Results of the Blaine-Value test.

Sample	Blaine Value [-]	Surface area [-]
Gypsum Keuper	5680	large
Reichenhall Form.	3340	medium
Haselgebirge	2240	small

4 THE POWDER SWELLING TEST

4.1 *Principles*

The swelling capacity was determined with the powder swelling test (PST) according to Thuro (1993), where the swelling displacement of a powdered sample is measured. The separated anhydrite samples were dried, fractured and grinded to a homogeneous powder with a defined grain size between fine sand and clay. Then the material was inserted in a testing cell (Fig. 5). It is necessary to produce a constant density of around 1.5 g/cm^3. The height of the inserted material is around 2.0 cm. The axial surcharge was minimised and consisted of the upper porous plate and the cap (together 72 g which is equivalent to a pressure of 0.18 kN/m^2). Distilled water was added to start the swelling. A manual dial gauge measuring the vertical displacement is read off once a day.

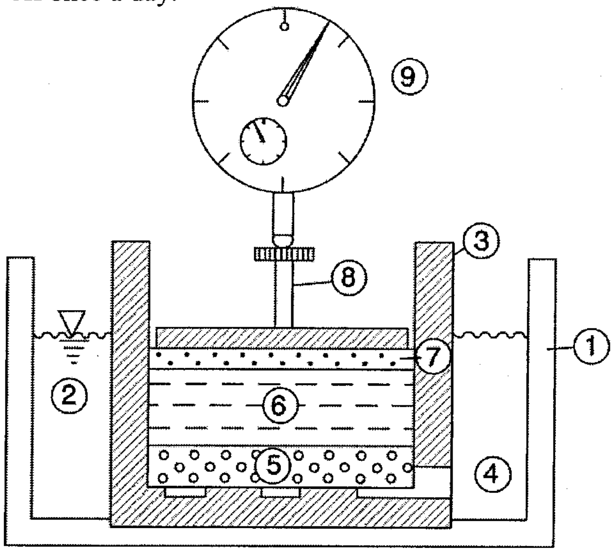

Figure 5. Drawing showing the test setup ("oedometercell") for the PST. 1 = water tray, 2 = water, 3 = swelling cell, 4 = drainage hole, 5 = lower porous plate, 6 = sample, 7 = upper porous plate, 8 = cap, 9 = dial gauge (after Thuro 1993).

The advantages of this method are obvious. It is possible to test almost every material and even only small layers of the sample. Another advantage is the ideal comparison of different samples because of the perfect identical preparation (Tab. 3).

Table 3. Comparison of different swelling tests concerning the most important swelling dependencies of anhydrite samples (Rauh & Thuro 2006).

Attribute of anhydrite	powder swelling test	other swelling tests
Content in sample	100 %	varying
Grain size	identical	varying
Distribution (in sample)	identical	varying
Water content	0 %	varying
Time for swelling	fast	slow

Additionally the test is less time consuming and first results are available after a short period of time, e. g. days or weeks (Tab. 3).

Nevertheless we should not forget to address the disadvantages. First of all it is not possible to gain any information about the swelling pressure of anhydrite directly from the results of the powder swelling test. Secondly we only determine the swelling strain of a disturbed sample.

It has to be pointed out that the second disadvantages is not essential, since in our opinion it is more or less impossible to collect an undisturbed and natural sample and fit it into the oedometer cell. There is always some tension release, additional cracking and change in water content.

4.2 *Different grain sizes*

The swelling capacity depends very much on the grain size of the anhydrite. This dependency can be visualized by calculating the surface areas of different grain sizes (Tab. 4). The calculation for each grain size is normalized to 1 g anhydrite with a specific density of 2.94 g/cm³. The surface area of 1 g anhydrite increases linear with decreasing grain size. To simplify the calculation, only cube and sphere shapes were used. This assumption is eligible, since anhydrite – also called "cube spar" – normally has cubic crystals (Fig. 8). Interestingly the surface areas of cubes and spheres are identical (Tab. 4).

Table 4. Correlation between surface area, grain size and subsequently grain shape. The calculated results are referred to 1 g anhydrite with a specific density of $\rho = 2.94$ g/cm³ (modified after Voll 1992).

Grain shape	sphere	cube
Diameter (d) or edge length (l) [mm]	surface area $(6 / (d * \rho))$ [cm²]	surface area $(6 / (l * \rho))$ [cm²]
0.063	323.939	323.939
0.2	102.041	102.041
0.63	32.394	32.394
2.0	10.204	10.204
4.0	5.102	5.102
6.3	3.239	3.239

The swelling behavior of anhydrite with different grain sizes was investigated with a modified powder swelling test. Therefore pure Haselgebirge anhydrite was crushed and sieved to obtain the grain sizes needed. 100 g of each grain size was mixed up with 50 g quartz powder. This procedure minimizes the caverns in the coarse samples. Then the material was placed in the swelling cell and the swelling process was started with distilled water. The swelling strain of the different grain sizes after 1.5 years can be seen in Figure 6. The swelling strain increases noticeable with decreasing grain size.

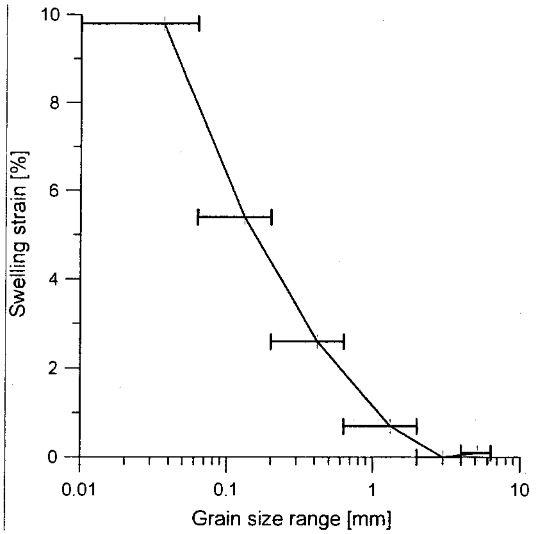

Figure 6. Swelling strain of pure Haselgebirge anhydrite that was crushed to different grain sizes. The swelling strain increases with decreasing grain size.

4.3 *Results of the swelling test*

The results of the powder swelling tests on different anhydrites are shown in Figure 7. The swelling displacement has finished or has gone below the accuracy of measurement after approximately one and a half years. The absolute identically prepared and assembled anhydrite samples have very different swelling capacities. The swelling strain decreases from the Gypsum Keuper and the Reichenhall Formation to the Haselgebirge. This corresponds very well with the former rock cover which increases vice versa.

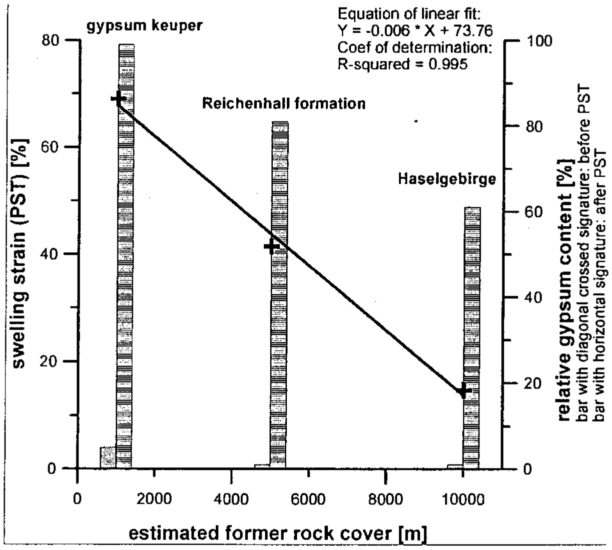

Figure 7. Diagram showing decreasing swelling strain in the PST against increasing former rock cover. The bars represent the gypsum content before and after the powder swelling tests.

5 GYPSIFICATON RATE AFTER PST

The anhydrite and gypsum content of the samples before and after the PST have been investigated. On the one hand the contents were determined by x-ray diffraction. The narrow tolerances could be achieved by using the Rietveld analysis program BGMN©. On the other hand the relative gypsum contents could be ascertained by a dehydration test after Henke & Hiller (1983). The results can be seen in Figure 7 and Table 5. Before the PST the samples consisted of anhydrite and more or less no gypsum. After the PST the anhydrite content in the samples varies from 1 % to around 40 %.

Table 5. Anhydrite content of the samples before and after the powder swelling test (analyzed with x-ray diffraction and dehydration tests according to Henke & Hiller 1983).

anhydrite content in volume-%	before PST absolute / relative	after PST abs. / rel.
Gypsum Keuper	81±3 / 95±1	1±1 / 1±1
Reichenhall Form.	81±2 / 99±1	16±2 / 19±1
Haselgebirge	96±2 / 99±1	40±2 / 39±1

These results are quite surprising since the anhydrite-gypsum conversion and therefore the swelling strain in the PST has stopped and/or undergone the accuracy of measurement. In other words there is still anhydrite left that does not change to gypsum.

The reason for this behavior can be found in the crystal grain sizes and therefore in the grade of transection of the different samples (Fig. 8). In the beginning the alteration process can only take place at the contact area between water and anhydrite or – in other words – at the outside of crystals or at grain boundaries.

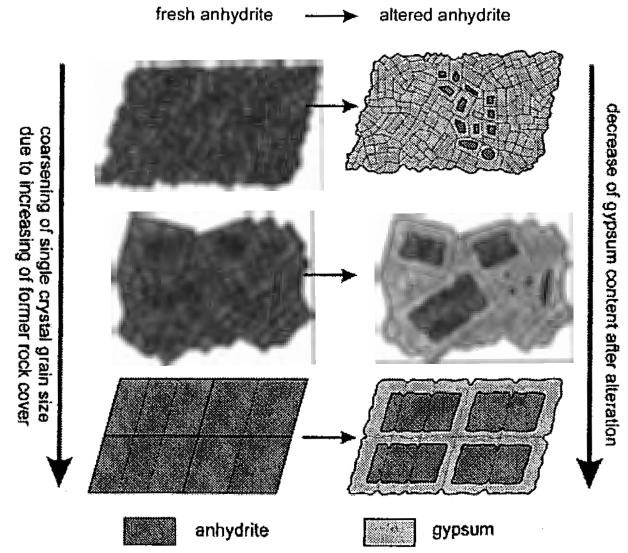

Figure 8. Alteration of anhydrite with different crystal grain sizes. The differences in the surface areas in the samples lead to varying gypsification rates.

Mature anhydrites with large crystals possess a small specific surface area (Tab. 2). Additionally the transection grade is low and the water can not easily enter into the sample. This leads to a small rim of gypsum around each large anhydrite crystal, which secludes the core from further contact to water and simultaneously prevents further alteration to gypsum (Fig. 8).

6 CONCLUSIONS

The results show that pure anhydrites differ in their swelling capacity even if they were grinded to the same grain size. The key to that behavior seems to be the discrepancy in the geological history of the materials. The decisive difference is the "maturity" of the anhydrite rock. Herein the maturity is reflected in a larger former rock cover which led to high temperature and stress conditions. That again produces a massive rock with large crystal grains and a relatively low specific surface area. The result is an anhydrite rock with a low reactive surface area and thus more or less inert (or "sluggish") to swelling.

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