Report, Project KEM-17

# Over-pressured salt solution mining caverns and leakage mechanisms

**Phase 1: micro-scale processes**

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## Management Summary

The goals for this project for the Kennisprogramma Effecten Mijnbouw (KEM-17) are to improve knowledge of the processes that occur when brine pressure in the cavern (locally) exceeds the minimum stress in the cavern roof or wall, considering this at the scales of the rock grains, the salt caverns and the salt domes.

In the salt-solution mining engineering community, there is a long-standing controversy about the processes when brine pressure in a deep salt cavern reaches values close to the minimum principal stress. One opinion is that it is difficult to prevent hydraulic fracturing, localised brine flow and major loss of containment. The other opinion is that as long as the pressure build-up rate is moderate, leak-off by diffuse permeation is dominant, and hydraulic fracturing can be excluded.

This KEM-17 project presents a review of existing salt engineering, materials science and geoscience literature relevant to the prediction of the evolution of abandoned, overpressured, brine filled solution mining caverns. **Phase 1** of this project is reported here, focussing on the MICRO SCALE processes.

Based on a comprehensive digital database of reports, publications and available data on this topic we provide a critical review of the relevant experimental and microstructural methods and observations. We focus on macro- and micro-scale models for deformation and rheology, permeation and hydrofracturing, discussing grain boundary structure, stability of permeation, problems of extrapolating laboratory data to abandonment conditions. We identify areas where there is need of improvement for long-term abandonment prediction and recommend areas for further research and development which will improve practice of safe abandonment.

The main conclusions of the report are:

1. Prediction of cavern convergence after abandonment requires extrapolation of engineering constitutive laws to heterogeneous salt at strain rates much lower than in the laboratory. This extrapolation is not based on the available microphysical understanding of the deformation. Accuracy of predictions of convergence can be strongly improved by including this knowledge.
2. Prediction of brine migration after abandonment requires extrapolation of engineering constitutive laws to heterogeneous salt and rates much lower than in the laboratory. This

extrapolation is not based on microphysical understanding of permeation. Accuracy of predictions of brine flow from abandoned caverns can be strongly improved by including this knowledge.

3. Existing experiments and microstructural arguments show that kinetics of permeation in high pressure caverns vary strongly with microstructure and impurities. We propose that permeation after cavern abandonment will be strongly heterogeneous and localised, and not diffuse over large volumes as predicted by current models.
4. Natural examples of the migration of overpressured brine through salt over long timescales show evidence for both the processes of diffuse localized permeation and hydrofracturing.
5. A first look at microstructures of rock salt core from two different salt cavern fields in the Netherlands shows that these are heterogeneous and quite different. A full materials science-based analysis will much improve predictions of abandoned caverns in these cases.
6. We recommend incorporation of existing materials science knowledge in salt engineering and research to further quantify the microphysics of rock salt deformation and permeation.

# Introduction

## Scope of this study

The specific goals for this project for a Kennisprogramma Effecten Mijnbouw Project are to improve knowledge of the processes that occur when brine pressure in the cavern (locally) exceeds the minimum stress in the cavern roof or wall.

In the salt-solution mining engineering community, there is a long-standing controversy about what occurs when brine pressure in a cavern reaches the local minimum stress. One opinion is that it is difficult to prevent hydraulic fracturing of the roof and the subsequent major loss of containment. The other commonly held opinion is that as long as the pressure build-up rate is moderate, leak-off by permeation is dominant, and hydraulic fracturing can be excluded.

This problem is complex, nonlinear and has important implications. We address this problem at three different scales: micro- cavern- and salt dome scales, integrating our results. In this report which is the first of three, we report on the micro scale aspects.

A wide range of laboratory measurements have shown that the permeability of virgin rock salt can be exceedingly low ($10^{-21}$ $m^2$ and less). Microstructural studies corroborate this, showing that in deep, pure, recrystallized Zechstein rock salt the microporosity at grain boundaries is typically not connected.

Other studies, based on tests performed in boreholes and caverns, conclude that the as-observed permeability in many salt formations can be in the range $10^{-19}$ $m^2$ – $10^{-21}$ $m^2$, sometimes higher. This rock salt is inferred to contain permeable impurities, or dilatant microcracks which are not closed by the ambient stress.

Halite rocks are known as the best seal for fluids in the subsurface, based on three key properties. First, the near-isotropic stress state provides resistance to hydrofracturing. Second, in-situ permeability and rock-salt porosity often are already very small at a burial depth of 100 m. However, there are exceptions, depending on microstructure and impurities in the rock salt. Third, rock-salt has been shown to re-seal fractures by a range of mechanisms which are only partly understood.

Under suitable conditions, however, all rocks can lose their sealing capacity. For engineering (short-term) applications, the criteria for leakage through rock salt and magnesium salts have been studied experimentally. However, the conditions in an abandoned cavern roof under which (localised) permeation or hydraulic fracturing will occur are not sufficiently understood. This may not be surprising given the lack of microphysical understanding, the empirical nature of the results and the need to extrapolate.

In engineering studies, both diffuse permeation and hydraulic fracture are discussed. In initially impermeable rock salt, permeation is thought to be possible when fluid pressure is slightly higher than the local minimum principal stress in the salt; (it is not clear how this changes with deviatoric stress in the salt) while hydraulic fractures can form when fluid pressure becomes significantly (several MPa) higher. However, the exact values of the parameters controlling these processes are not well-understood, and their values even less so, giving rise to the above-mentioned controversy. In a bit more detail, the processes are as described below. The process is nonlinear and potentially unstable.

It has been shown in both laboratory experiments and in-situ tests that brine (and oil) can migrate through salt along diffusely dilatant salt-grain boundaries. Some current models assume that in an abandoned salt cavern's roof, this permeation process is slow, permeating a large volume of rock salt and creating a volume of dilated salt that both hosts significant brine and adjusts the permeability by nonlinear, effective-pressure-dependent dilatancy. However, considering the nonlinear nature of this process in a heterogeneous rock-salt mass, others argue that this process is unstable and will lead to preferential fingering — or even fracturing — and a fluid pathway with a much smaller volume. This is especially true, as brine density is smaller than rock density: when head losses are small, the gap between brine pressure and geostatic pressure in the dilated zone is larger when this zone moves upward, making the process potentially unstable.

Controlling parameters are thought to be the salt microfabric, the state of stress and the of fluid pressure. The infiltration along fronts that are irregular even in the laboratory scale has been studied in bulk experiments and in solution-mined cavities, and first-order trends in evolution of dilatant porosity and permeability of the salt were measured. Because of the long-time scales and low differential stresses over which predictions concerning this process have to be made, they are difficult to test. Several in-situ tests were performed (at Etrez (France) or Etzel (Germany, for example), but their results are not simple to interpret. Considering that the vast majority of models and criteria used in the engineering literature are based on bulk sample tests, it is difficult to extrapolate these results to real-life abandonment conditions. In our opinion, this is the main reason for the current controversy.

One obvious approach to this problem concerns both material science and microphysics, where the processes are studied at the scale of the grain boundaries and microcracks to develop mechanism-based constitutive relationships for the development of transport properties in rock salt at anisotropic stress and close-to-lithostatic fluid pressures, suitable for extrapolation to nature. **At this point, little of the**

**existing microphysical knowledge in this domain is used in the solution-mining community.** This is the central theme of Phase 1 (micro-scale) of this KEM project.

From our experience with different aspects of the above stated problem, it seems that there are very few studies in the literature of salt engineering of the microphysics of fluid permeation and hydrofracturing in salt. On the other hand, there are extensive microstructural and microphysical studies of the interactions of fluids and grain boundaries and of intergranular microcracks in salt in the literature of geosciences and material science.

Microstructural evidence suggests that, in nature (which is a good analogue for the long time scales expected in abandoned caverns), both diffuse dilatancy and Mode I fracturing can occur at very low effective stress conditions — i.e. at near-lithostatic fluid pressure and differential stress of a few MPa in the deep subsurface, accompanied by crack healing through precipitation from supersaturated solutions.

Experimental determination of the mechanical and transport properties of halite and extrapolation of these data to conditions of slow geological deformation have provided a solid basis for understanding rock salt rheology, and it is clear that brine plays a major role in this. On the basis of these experiments, it is possible to differentiate deformation mechanisms in halite based on their characteristic microstructure. Compared to other rock types, rock salt contains only small amounts of $H_2O$, but it was shown that even this small amount dramatically enhances fluid-assisted grain boundary processes such as grain-boundary dilatancy, grain-boundary migration and pressure solution. During grain-boundary migration, fluid inclusions interact with the moving grain boundary and that this process has a strong effect on rheological properties

A number of processes are known to create an increase of permeability in initially impermeable or poorly permeable material. The first is microcracking and associated dilatancy, which can take place at high differential stresses or high fluid pressures. Dilatancy in the presence of brine has been shown to be suppressed by pressure solution and recrystallization processes. However, the conditions relevant to this project, which are low differential stresses and high fluid pressures, are largely unexplored. These conditions are complex indeed: in the case of permeation, they include the establishment of a pore pressure in rock salt where Terzaghi's law is added to the time-dependent elastoplastic material law. (NB For low porosities it has been also considered to use Biot's effective stress). "Whether the notions of Darcy flow and Biot's coefficient (classical notions in Reservoir Engineering) apply is an open question'" Bérest P. (2013). The mechanical behaviour of salt and salt caverns. Key Note Lecture. Proc. Eurock 2013, Balkema, 17-30.

It has also has been shown that permeated rock salt can undergo complex dilation-compaction, recrystallization, crystal plasticity and pressure solution processes which change the microstructure; therefore, a number of potential feedback processes are present.

An additional process is the formation of topologically connected brine-filled pores and triple-junction tubes in halite at a pressure and temperature corresponding to depths > 3 km.

## Questions asked in this review

In this report, we will attempt to answer the following questions.

1. What are the micro-scale processes during (localised) permeation and hydraulic fracturing as a function of brine pressure and rate of pressure increase, stress tensor and salt material characteristics such as grain size, grain fabric, grain boundary structure, fraction and spatial distribution of second phase inclusions, solid solution impurities, and related mechanical properties?
2. At which combination of parameters is permeation a dominant mechanism that controls the development of preferential fingering. At which combination is hydraulic fracturing dominant? To which parameters is the transition most sensitive? What is the inherent parameter uncertainty? Is there a self-strengthening or self-weakening feedback mechanism?
3. To what extent can the flow of brine be accommodated by these processes?
4. What are appropriate models to simulate these processes?

## Aims and deliverables

1. Compile a comprehensive digital database of all relevant reports, publications and available data on this topic.
2. Provide a critical review of the available relevant experimental and microstructural methods and observations.
3. Provide a critical review of the available macro- and micro-scale models for permeation and hydrofracturing, discussing grain boundary structure, stability of the permeation, and the applicability to long-term abandonment prediction
4. Identify the lack of data/knowledge/models required for long-term abandonment prediction.
5. Synthesise data and models to arrive at a summary statement of the state of the art in our understanding of the microscale processes in (localised) permeation and hydraulic fracturing

6. Answer questions about the combination of parameters for which permeation is dominant: What controls the development of preferential fingering and at which combination is hydraulic fracturing dominant? To which parameters is the transition sensitive? What is the inherent parameter uncertainty? How big a brine flow can be accommodated by these processes? What are appropriate models to simulate these processes?
7. Study the microstructure in representative intervals of Zechstein rock-salt core relevant for solution mining in the Netherlands, demonstrating a currently available practical set of analytical techniques with automated image analysis to quantify the full set of key parameters required to provide a benchmarked description in future analyses.
8. Provide a basis for future micro-structural characterisation for salt engineering and also provide an impression of the variation in salt properties to be expected in typical Dutch solution mining.
9. Formulate practical recommendations for determining when and which leakage mechanism (permeation/hydraulic fracturing) is dominant at the micro scale and in what cases it cannot be determined.
10. Formulate recommendations for further study and development.

## Outline of this report

Micro - scale understanding is relevant for two aspects of over-pressured caverns and leakage mechanisms: rheology of the rock salt and permeation:

Part (1)

Deformation of salt around a cavern, which leads to changes in (i) in-situ stress, the amount and morphology of porosity, (ii) permeability, around a cavern

Here we consider rheology and micro-scale deformation mechanisms, with special attention on grain boundary structure

Part (2)

Permeation of the salt roof by fluid at pressure slightly higher than the minimum stress in the salt

Here we consider the micro-scale mechanisms of permeation: the creation and sealing of connected porosity

Part (3)

Microstructural study of rock salt core samples from the Netherlands taken from solution mining wells

Illustrate the available modern methods available. Detailed results of this are presented as Appendices A and B

These results will be integrated with the cavern scale and salt dome scale work packages of this KEM project (Phase 2 and 3).

### Literature database

We have compiled a Zotero database of a wide range of relevant literature. Present content about 1500 publications shared with team. Fully searchable, includes annotations, the original papers and reports as pdf. Sources from Salt Engineering, Materials Science, Geoscience and Numerical modelling. **Fig. 1** shows a screenshot of the database which is one of the deliverables.

## Part 1: Rheology and Microphysical deformation mechanisms of salt rocks - a materials science- based view

Rock salt is a very weak rock and deforms readily under small deviatoric stresses in sedimentary basins. Exactly how weak salt is more controversial. Rheology of rock salt at low deviatoric stress can vary by orders of magnitude by the operation of different microphysical mechanisms and a strong role of grain boundary water.

The constitutive laws governing the deformation of evaporites are related to microfabric, impurities, and fluid distribution. Calculation of the processes around salt caverns requires extrapolation of constitutive laws to conditions outside those of laboratory experiments. This has to be done based on understanding of the microphysical processes operating, however this is rarely done in salt engineering.

Study of microphysical deformation mechanisms integrated with mechanical and fluid transport properties is a well-established method for this. It is well documented in the geo-scientific literature on deformation and fluid flow in salt. It allows to distinguish dislocation versus pressure solution creep, connected versus non-connected porosity etc. Fluid-assisted grain boundary processes of dissolution-precipitation and recrystallization are important in deformation of rock salt. This has important consequences for mechanical and transport properties, but has not been routinely implemented in salt engineering models. Recently, there are projects (**Fig. 2, Fig. 3**) starting in the salt engineering community to incorporate a change in stress exponent into constitutive laws, albeit without including microstructural study. (Bérest et al., 2019) have summarized the state of engineering knowledge as follows:

"... the stress levels in the large rock mass surrounding the cavity are well below the stress levels used in laboratory investigations. This means creep behavior for stresses levels $\sigma_v < 8$ MPa is not well based on observation in lab but based just on extrapolation" … "Langer (1984) stated that reliably extrapolating the creep equations at low deformation rates can only be carried out on the basis of deformation mechanisms."….. "However, Spiers et al. (1990) and Urai and Spiers (2007) observed that pressure-solution creep, which is an important deformation mechanism of most rocks in the Earth's crust, is especially rapid for rock salt in the low-stress domain. Theoretical findings and experimental evidence strongly suggest that the relationship between deviatoric stress and strain rate is linear for this mechanism. According to the literature (Peach et al. 2001; Ter Heege et al. 2005a, b; Urai and Spiers 2007; Urai et al. 2008), modestly accelerated dislocation creep behaviour in the transitional region between dislocation-dominated flow and pressure-solution creep is caused by rapid, dynamic recrystallization that involves fluid-assisted grain-boundary migration. In this context, water content in the sample and grain size play a significant role during a creep test."

Similar conclusions are drawn in a recent paper on cavern closure (Cornet et al., 2018) (bold by JLU).

"Based on numerical and analytical modeling, the closure rates of a circular hole in rock salt have been evaluated under a variety of in-situ conditions. The steady state behavior of rock salt has been treated using the incompressible **Carreau viscosity model, which combines pressure solution at low rates of deformation and dislocation creep at high rates of deformation.....** The influence of grain size dominates over all other parameters such as temperature or dislocation creep parameters. **The difference in closure rates between coarse- and fine-grained salts can reach several orders of magnitude** and significant hole closure can be attained in less than one day in very fine-grained salts. Given that grain size may vary considerably within the same salt body, closure rates may also be very different and it is important to characterize the range of grain sizes at the location of the hole.........In the power-law regime, hole closure is almost n times more sensitive to shear than pressure loads, demonstrating the **necessity of considering far field shear stresses for hole closure in applications going through moving salt bodies (effect of far field anisotropic stress)**..... The relative contribution of dislocation and pressure solution creep depends on the load, temperature and especially grain size. In general, **pressure solution is dominant in cold fine-grained salts under low loads while dislocation creep is dominant in warm coarse-grained salts under high loads.** Dislocation and diffusion creep dominated zones may co- exist around a single hole due to complex stress perturbation in the near field..........**A thorough understanding of salt grain size and water availability is needed to properly assess the relative contribution of dislocation and pressure solution creep in hole closure.** The dominant deformation mechanism in a particular salt is usually determined by grain size alone, but this is not enough: the type of salt and temperature also matter"

Bérest (2013) argued: "One can expect that brine influence be even more significant. Spiers et al. (1990) and Cosenza et al. (2002) proved that, when saturated brine is added at the periphery of a salt sample submitted to a moderate mechanical loading (say, 10 MPa), creep rate immediately accelerates. However, when mines are flooded with brine, it seems, in most cases, that the brine-filled mine is more stable than the dry mine had been. [See Van Sambeek & Thoms (2000), who observed that subsidence rate above the Jefferson Island Mine decreased by a factor of 10 after the mine was flooded, or Feuga (2003), who performed a sonar survey in an old flooded mine at Dieuze to find that actual mine geometry was exactly the same as what was expected from a map drawn by the miners 130 years before.] These results suggest that creep acceleration only exists in a relatively thin damaged zone at pillar walls and that the favourable effect of the increase in mine internal pressure (from atmospheric to halmostatic) is much more significant from the perspective of mine stability"

This points to the complicated feedbacks between, stress, water content, and evolving microstructure in deformation of rock salt.

Deformation of Sodium Chloride has been studied for over 100 years. The book by Schmid E., Boas W. (1935) *Plastizität und Festigkeit von Ionenkristallen* gives a summary of the early studies, mostly on single crystals. In this review we do not aim to provide a full comprehensive discussion of all literature, because these have been provided by recent reviews: (Fredrich et al., 2007)(Urai et al., 2008)(Urai et al., 2008)(Hunsche et al., 2003)(Hampel, 2016). Rather, we present and discuss the literature which illustrates the key points relevant for the questions asked in this study.

Early studies of steady state flow of polycrystalline Halite (natural or synthetic rock salt) **(Fig. 4)** recognized that confining pressure, temperature, strain rate and composition of solid solution impurities **(Fig. 5)** were important parameters controlling the flow stress. The classic studies by Carter, Hansen and coworkers used triaxial experiments **(Fig. 6)** together with materials science methods **(Fig. 7)** of investigating microstructure to understand the rheology of their samples and to extrapolate their results to much lower stresses and strain rates, at relatively low temperatures. At this time, no techniques were available to study the full microstructure, and microscopy was done on cleavage chips etched to reveal dislocation and subgrain boundaries. As expected, dislocation density was shown to increase with deviatoric stress, **(Fig. 8)** while subgrain size decreases with deviatoric stress **(Fig. 9),** providing a laboratory-calibrated method to measure the deviatoric stress which was acting in the rock salt in the past. It was shown that deviatoric stresses in rock salt varied between 0.2 and 2 MPa, indicating that the virgin in-situ stress in rock salt is not completely isotropic as commonly assumed in engineering studies. These techniques have become much easier with the invention of several microstructure decoration techniques **(Fig. 10)** and there is now an extensive database of subgrain size measurements in rock salt from a range of settings. An important contribution to our understanding of the mechanism of dislocation creep by Muhammad and coworkers **(Fig. 11)** used the pressure-sensitivity of flow stress to show that dislocation creep in nature is controlled by dislocation climb.

The creep of rock salt is extensively studied in a large number of triaxial experiments, some of which have combined this with microstructural study. **(Fig. 12)** shows a collection of the power law creep parameters of a number of these studies. while **(Fig. 13, 14)** shows plots of a large dataset of experiments, which however require extrapolation of the trends found to the low deviatoric stress and strain rate relevant for operation and abandonment of caverns. These studies were not combined with microstructural study, and (as will be shown below) microstructural changes related to grain boundary water and the onset of pressure solution, which will cause dramatic changes in strain rate at low deviatoric stress, not investigated.

In several studies which are illustrated in **(Fig. 15, 16, 17, 18)** it was shown that rock salt samples from different stratigraphic units of the Zechstein creep at different rates (by several orders of magnitude) under otherwise the same conditions, and that this will have strong effects on how caverns converge. This is an important finding and consistent with what is expected based on microstructure, but is not a classification which can be extrapolated to long-term, low deviatoric stress deformation without showing that the microphysical deformation mechanisms remain the same.

At low confining pressures and high deviatoric stresses, flow of rocksalt is accompanied by dilatant grain boundary microcracking and rapid permeability increase. The mechanical conditions under which this occurs have been determined in triaxial experiments, providing an important concept in the integrity of engineered structures in rock salt. **(Fig. 19)** shows that under a given confining pressure there is a critical deviatoric stress (differential stress) under which the deformation of the sample is dilatant (with the creation of microcrack porosity). How this boundary translates to conditions at the walls of a salt cavern where the deforming rock salt is in contact with brine under pressure is not clear, and the effects of small amount of water which are always present in natural rock salt, especially at low deviatoric stresses of a few MPa have not been investigated, so it is difficult to know how these concepts translate to rock salt around abandoned caverns. While the onset of microcracking has a minor direct effect on creep behavior, it is important to note that it can strongly influence the effects of water on creep. In salt containing small quantities of water, microcracking disrupts grain boundary films and inhibits both grain boundary migration and pressure solution, particularly if the water can escape from the sample. On the other hand, under conditions where microcracking allows free brine or water vapor access to the interior of a creeping salt sample, then both recrystallization and solution-precipitation creep effects can be strongly enhanced.

In 1984 the discovery of methods to easily image the full microstructure of rock salt allowed to investigate changed in natural rock salt deformed in the laboratory. A series of triaxial deformation experiments **(Fig. 20)** on Asse domal salt with very low water content (present as small unconnected brine inclusions at grain boundaries) has shown, that in the non-dilatant field, and after rapid deformation to about 10% strain, at lowering the deviatoric stress to values below 10 MPa these samples deformed many orders of magnitude faster than predicted for dislocation creep. Samples initially deformed in the dilatant field did not show this effect. This dramatic weakening was explained by the formation of thin brine films along the grain boundaries which dramatically increased their mobility, leading to recrystallization **(Fig. 21)** and grain refinement by grain boundary migration and the activation of pressure solution creep. The same effect was seen in wet, synthetic, fine grained rock salt samples which deform much faster than predicted by dislocation creep, and this deformation is well explained by pressure solution. This effect is expected to be prominent in the rock salt around caverns and play an important role during abandonment by making the rock salt around the cavern creep much faster than predicted based on extrapolation of the results discussed earlier in this report.

In the geoscience literature on rock salt rheology these concepts have been widely published and used to describe observations in naturally and experimentally deformed rock salt and other evaporite minerals **(Fig. 22),** showing that dislocation creep, dynamic recrystallization and pressure solution creep are common processes operating in deformed rock salt in the laboratory and in nature. The microphysically validated constitutive equations describing this deformation form a material-science based framework which can be extrapolated to low deviatoric stress- low strain rate deformation around abandoned caverns, by calibration against naturally deformed microstructures.

The grain boundary fluid films which dramatically increase the grain boundary diffusivity and mobility have been shown to be very thin, of the order of a few nm **(Fig. 23).** These fluid-filled grain boundaries migrate rapidly through deformed rock salt by solution-precipitation processes, driven by contrasts in dislocation density and by grain boundary curvature. Their migration can be modelled quantitatively by assuming that diffusion across the migrating grain boundary is the rate limiting step in the process **(Fig. 24).** If the rock salt contains small but significant amounts of water in the form of saturated brine inclusions which transform into grain boundary films, as is generally the case for both natural and synthetic samples, fluid assisted grain boundary migration is an efficient process of reducing dislocation density and hence removing the stored energy of dislocations, even at room temperature. This recrystallization process is driven by chemical potential differences across grain boundaries related to the dislocation density differences between old deformed grains and newly growing grains. In strongly deformed, wet halite, the migration process is very rapid, reaching rates up to 10 nm/s at room temperature.

An additional important process is when below some critical difference in driving force for cross-boundary solution-precipitation transfer, surface energy driving forces cause necking of grain boundary fluid films to form isolated fluid inclusions, thus rendering the boundaries immobile and stopping pressure solution **(Fig. 25, 26, 27, 29).** This process is reversible, restoring the fluid films at sufficiently high deviatoric stress. The grain boundary fluid films in their different structural states were imaged by cryogenic BIB-SEM techniques and by FTIR **(Fig. 25, 26, 28, 30).**

We infer that these processes were commonly active in the samples reported in (Figs. 13-19) but not detected due to the absence of microstructural information; they play an increasingly important role at low stress-low strain rate conditions of cavern operation and abandonment.

Another independent source of data on slow natural deformation of rock salt has recently been provided by measuring the surface displacement field of salt in nature, in areas of active salt tectonics, in salt mining districts, on sediment rafts above mobile salt, on emerging salt diapirs, and in areas where removal of ice sheets has led to a change of overburden load. These data can be inverted using non-linear finite element techniques, to obtain constitutive equations for salt flow during slow natural deformation, for example in Mount Sedom (Dead Sea) (Weinberger et al., 2006). These results are in very good agreement with models based on simultaneous operation of dislocation creep and pressure solution. For example, the resulting Mount Sedom rock salt viscosity is determined to be $2.5\times10^{18}$ Pas, and the associated strain rate is between 5 and $6\times10^{-13}$ $s^{-1}$, in very good agreement with the rheology outlined in **(Fig. 22).** Steep uplift gradients observed by InSAR along the western margin of the diapir are higher than predicted by modeling of Newtonian viscous flow. This could imply that flow of power law viscous fluid may be more suitable than that of Newtonian viscous fluid for the Mt. Sedom rock salt at high strain rates.

In laboratory triaxial deformation experiments integrated with microstructural study it was shown by extensive studies that the effect of small amounts of water in rock salt (> 10 ppm, which is always present in nature, so that rock salt in-situ is always "wet") have a clear difference with artificially dried samples which creep about three orders of magnitude slower than wet samples under the same conditions. There is extensive dynamic recrystallisation in the naturally wet samples as shown by detailed microstructural analysis and the samples deform by equal contributions of dislocation and pressure solution creep. This is the basis of the well-known microphysical field boundary model. The dynamically recrystallized grainsize is a function of deviatoric stress: the lower the deviatoric stress, the higher the dynamically recrystallized grainsize **(Fig. 31, 32).** In the volumes of rock salt close to cavern walls where deviatoric stress and strain rate are relatively high, this is expected to be a common process.

As already indicated in the beginning of this report, the salt engineering community is starting to adopt the change in constitutive behavior at low deviatoric stress, albeit without integration with microstructural study of deformation mechanisms **(Fig. 31, 32, 37, 38, 39).**

Solution - precipitation creep is an important de-formation mechanism in most rocks in the Earth's crust, but is especially rapid in halite. In brief, the differences in chemical potential between points in the solid at grain boundaries under high stress and those under lower stress provide the driving force for dissolution, transport by diffusion in the intergranular fluid, and precipitation. Additional driving force (chemical potential drop) both along and across grain boundaries can be provided by internal plastic deformation of the grains, giving rise to combined grain boundary migration and solution-precipitation creep. While dislocation creep processes take place in the crystal lattice of the halite grains, solution-precipitation creep, or "pressure solution", is a process, which occurs in the grain boundaries. Here, in the presence of a small amount of saturated grain boundary brine, grains dissolve at highly stressed boundaries, and after diffusion of the material through the grain boundary fluid, the material crystallizes at interfaces under low normal stress. This process is accompanied by intergranular sliding and rotation (grain rearrangement), and can lead to compaction of porous salt or to deviatoric strain of non-porous aggregates **(Fig. 35, 36).**

Because of the strong grain size dependence and limited duration of laboratory tests, solution-precipitation creep is rarely seen in experiments on natural rocksalt (grain size several mm), and it is therefore not usually included in engineering descriptions of salt rheology. Nonetheless, predictions made using the constitutive equations summarized in **(Fig. 22)**, provided the rock salt contains sufficient water (>10-20 ppm, as most natural salts do), pressure solution creep should become important at strain rates below those reached in experiments.

We note here that despite the large amount of data now available on solution-precipitation creep in salt, details of the microphysics of the process are incompletely understood and more work is needed to completely understand this process.

The geoscience community has also developed in-situ methods to measure the rheology of large rock salt masses in the subsurface at very low deviatoric stress and strain rates **(Fig. 40, 41)**. This is based on deviatoric stress around large anhydrite inclusions in rock salt which are common in the Zechstein, caused by the density difference. These deviatoric stresses are very small for inclusions of a few 100 m in size, providing a way to interrogate the in-situ deformation of rock salt at very low deviatoric stress by measuring the gravitational sinking rate. This was investigated in two series of numerical models with the two end-member rheology (dislocation creep vs pressure solution) and has shown, that in the case of the Tertiary evolution of the Zechstein, pressure solution cannot have been active because the stringers would all have sunk to the bottom of the Zechstein by now. An explanation for this may be that below some critical difference in driving force for cross-boundary solution-precipitation transfer, surface energy driving forces cause necking or healing of grain boundary fluid films to form isolated fluid inclusion), thus rendering the boundaries immobile and switching off pressure solution. At present, the long-term rheology of Zechstein rock salt seems to be dominated by dislocation creep. This compares well with the microstructural observation that despite the high rate of fluid-assisted grain boundary migration observed in experiments, most naturally deformed rock salt is not completely recrystallized and preserves subgrains **(Fig. 29)**.

These complex changes in rheology are illustrated in (Fig. 42), providing a basis for a microphysically and geologically constrained constitutive law which can be incorporated in numerical models to predict the deformations around salt caverns more accurately.

State of the art materials science models are increasingly based on numerical, grain-scale models of the microstructure, to compute aggregate behavior and upscale these to bulk properties. **(Fig. 43 and 44)** are examples of this for rock salt. These studies are in their early stages, and we recommend development of these models as part of further studies of rock salt deformation and permeation around salt caverns.

Results of these studies will result in a microstructure-based constitutive equation for rock salt, which couple microscale and macroscale properties of rock salt and can be used to determine the constitutive law (also at low deviatoric stress) based on microstructural study of core material. At present, mechanical properties of salt for cavern design are exclusively determined by laboratory measurement of stress-strain relations. The measurements are time consuming and therefore usually incomplete for prediction of long-term behavior. Over the past decades there were attempts to predict this using microstructure, which would allow property determination in the whole core. For dislocation creep, important parameters which affect creep properties are: solid solution impurities, second phase impurities, grain size, grain shape, subgrain size and density, dislocation density, crystallographic fabric and grain boundary mobility (the presence or absence of continuous fluid films on the grain boundaries), pore pressure (if present). For pressure solution creep, important parameters which affect creep properties are grain boundary mobility (the presence or absence of continuous brine films on the grain boundaries) and grain size, and to lesser extent solid solution impurities and pore pressure and porosity. To couple microscale and macroscale properties of rock salt, once the complete set of material variables is defined, these are measured for a sufficient number of samples, for which also the bulk properties are measured. Then based on microphysical models and upscaling using micromechanical models, first order correlations are explored. Finally, the dataset is analysed using modern machine learning techniques with sufficient training data **(Fig. 46 and 47)**, to predict the bulk properties from microstructure.

## Conclusions: Rheology and Deformation mechanisms

The materials science of the rheology and microstructure evolution of rock salt is reasonably understood but this knowledge is not integrated between engineering and materials science domains. Especially deformation at low deviatoric stress and the effect of grain boundary water needs more study.

The constitutive laws governing the deformation of rock salt are related to microfabric, fluid distribution, impurities, strain rate, temperature. Study of deformation mechanisms tells us about mechanical properties and fluid transport: Power law versus Newtonian creep, grain boundary structure and mobility, connected versus non-connected porosity, permeability versus effective stress. Rock salt contains only small amounts of $H_2O$, and in the Zechstein (at virgin state after a long geological quiet period) it is generally present in unconnected grain boundary pores, so that at present pressure solution is not generally active. However, when the effective stress is changed significantly (values of this are not well known) even this small amount dramatically enhances fluid-assisted grain boundary processes such as grain-boundary dilatancy, grain-boundary migration and pressure solution and has a strong effect on rheological and transport properties. Current engineering practice does not use this information.

Reliable modelling of the deformation and fluid transport in rocksalt under the very low strain rates characterizing long term engineering conditions requires **extrapolation of experimentally derived constitutive equations** to deviatoric stresses and strain rates lower than those attainable in the laboratory. This extrapolation must be **based on a modern materials science approach (but is currently not)**, with quantitative understanding of all the microphysical deformation mechanisms operating in the experimentally deformed samples, integrated with studies of natural laboratories where deformation took place under much lower strain rates. The engineering creep laws generally used in the salt mining industry are based on dislocation creep processes quantified in laboratory experiments of necessarily limited duration. However, a large body of evidence clearly demonstrates that grain boundary dissolution-precipitation processes, such as solution-precipitation creep and dynamic recrystallization, commonly play a significant role in natural rock salt and are expected to be common around solution mined caverns too. Currently the salt engineering community is starting to investigate this effect. Therefore, **current predictions of convergence rate are quite uncertain, and this uncertainty can be strongly reduced by using a modern materials science approach to determine rock salt rheology under all relevant conditions.**

**In-situ (virgin) deviatoric stress in rock salt** as measured by subgrain size piezometry can be as high as several MPa, in areas of active salt movement and in the order of kPa around Anhydrite bodies enclosed in the salt. When tectonics or glacial loading are over, this value relaxes and the in-situ virgin stress tensor becomes much closer to isotropic. This is the case in the vast majority of the Zechstein.

We reviewed the microphysics of **grain boundary water-related dynamic recrystallization and solution-precipitation processes** in rock salt, together with the constitutive relations associated with these processes, and we discuss the contribution of these mechanisms around solution mined salt caverns, during their operation and during abandonment. In many current engineering studies, the effects of water-activated grain boundary processes are neglected, and this omission leads to errors in prediction of displacement rates, especially over long term. **Geomechanical modelling of the deformations around salt caverns can be significantly improved by using constitutive equations based on microphysical models for dislocation creep in combination with solution-precipitation creep, fluid-assisted recrystallization and surface energy driven grain boundary healing, site-specific for the rock salt around the cavern**. Recent development of methods to study microstructures in rock salt include (i) transmitted light microscopy of Gamma- decorated thin sections, (ii) subgrain size paleo-piezometry of polished and chemically etched samples using reflected light microscopy, (iii) micro-CT analysis of grain boundary fluid inclusions, (iv) analysis of grain boundary structure and microchemistry by cryogenic BIB-SEM and (v) X-ray or EBSD orientation imaging.

**This toolbox is readily available for engineering studies** and the world's top laboratory to carry out these studies is in the Netherlands. There is a growing understanding of on deformation mechanisms, constitutive laws and fluid flow in naturally deforming rocksalt (at stresses and strain rates relevant to caverns) by inverting data from a wide range of geological settings. Integrating this knowledge base provides an improved basis for making better predictions of the evolution of abandoned salt caverns. There is need for additional materials research, especially of the kinetics of transformation of grain boundary fluids, and the role of material variables like composition and impurities.

Deformation **at the walls of brine filled caverns** is usually modelled assuming that the salt is initially impermeable, and if it deforms in the non-dilatant field it will remain impermeable, so that the brine only acts as a confining pressure. This can however better be compared with "unjacketed" triaxial tests, and it is likely that deformation leads to change in grain boundary structure, where a small amount of brine penetrates the grain boundaries, establishing a pore pressure which slowly reduces the effective stress at the cavern walls, leading to slow dilatancy and permeation, even under conditions which would traditionally be evaluated as non-dilatant. Calculated deviatoric stresses around producing brine filled caverns can be of the order of 10 MPa, leading to considerable strains, **dynamic recrystallisation and change in grain size and grain boundary structure - during abandonment and relaxation of deviatoric stress this will have a much weaker rheology than predicted based on dislocation creep alone**. More work is needed to implement this knowledge to predict these changes at the cavern walls.

We note here that these conclusions are formulated for brine-filled caverns, and may be different for caverns filled with compressed air, oil, natural gas or hydrogen.

## Recommendations, Rheology and Deformation mechanisms

Based on this literature review it is clear that the constitutive equations used to model the mechanical response of solution mining caverns can be much improved, especially for the conditions which cannot be accessed by laboratory experiments.

We recommend the use of constitutive laws which are supported by microphysical materials science evidence, to arrive at much improved and less uncertain predictions of convergence, incorporating state variables for changing of grainsize, grain boundary structure, and the corresponding microphysical deformation mechanisms.

**Such a project would integrate laboratory measurements of stress-strain response of rock salt samples with microscopic and microanalytical characterization of the undeformed materials (parameters to be measure are summarized in Fig. 47). This will provide quantitative understanding of the microphysical processes operating in the samples, provide a basis for the right conditions of the deformation experiments and allow extrapolation of the measured constitutive equations to low strain rate and time scales which can not be accessed in the laboratory. Examples of such projects are provided by many of the papers cited in this review. An emerging approach which is well suited for this problem is microdynamic simulation, where the microphysical processes identified are implemented numerical models of the microstructural evolution, and allow upscaling to bulk constitutive equations. Site-specific analyses can be done**

**using the core materials taken in the well from which the cavern was developed. Validation of the predictions for very low stress can be done by comparison with natural, actively deforming salt structures.**

**One key microphysical process which is incompletely understood is the conditions and kinetics of grain boundary structure transitions in rock salt; in other words, the stresses strain rates and strains required to activate grain boundary fluid films, and the stresses, strain rates and strains required to start disruption and healing of these films. This is required to model cyclic rheology around salt caverns and is not completely understood – more basic research work is recommended here.**

We recommend more materials science-based investigations of the changes in the properties of the rock salt at the cavern wall, during operation and during abandonment.

Materials science studies of rheology, AND microstructure of salt cores from cavern intervals to provide much improved constitutive equations for calculating deformation during solution mining and during abandonment, with site-specific predictions of rheology to improve predictions for cavern abandonment.

Development of a knowledge base of microstructure AND mechanical properties for a series of rock salt samples, to allow better extrapolation and the development of microstructure-based rock salt rheology predictions.

## Part 2: Microscale permeation mechanisms of salt rocks

### Problem statement

Rock salt is known as the best subsurface seal for fluids, based on three key properties. First, the near-isotropic stress state provides resistance to hydrofracturing. Second, in-situ permeability and rock-salt can be very low, when the salt is pure, porosity low and grain boundary fluid films are healed **(Fig. 49)**. Third, plastic deformation of halite in nature is usually ductile and non-dilatant.

After shut-in of a solution mined cavern in rock salt that is impermeable, convergence and temperature rise may lead to a state where at the top of the cavern pressure rises to values above the minimum principal stress in the surrounding salt and consequent flow of the cavern fluid into the surrounding salt **(Fig. 48)**. Here, the vertical dimension of the abandoned cavern plays an important role. If this dimension is sufficiently small, the abandoned cavern may be stable over long periods, as shown by "fluid pockets" in salt:  porous regions in rock salt filled with brine at lithostatic pressure which can be stable over geologic time and cause drilling problems when encountered in hydrocarbon wells (e.g. TNO report TNO2018 R10975). If the excess pressure at the top of the cavern is sufficiently large, it has been shown in both laboratory experiments and in-situ tests that brine (and oil) can migrate through salt along diffusely dilatant salt-grain boundaries. Some current models assume that in an abandoned salt cavern's roof, this permeation process is slow, permeating a large volume of rock salt and creating a volume of dilated salt that both hosts significant brine and changes its permeability by nonlinear, effective pressure-dependent dilatancy. Alternatively, the permeation can be localized, or if the excess pressure can rise several MPa above the minimum principal stress in the salt, hydraulic fractures can form.

Controlling parameters are thought to be the salt microfabric, the state of stress and the amount of fluid pressure. The infiltration along fronts that are irregular even in the laboratory scale has been studied in bulk experiments and in solution-mined cavities, and first-order trends in evolution of dilatant porosity and permeability of the salt were measured. Considering the nonlinear nature of this effective pressure-dependent dilatancy in a heterogeneous rock-salt mass, we propose that this process is unstable and will lead to preferential fingering — or even fracturing — and a fluid pathway with a much smaller volume. This is especially true, (as brine density is smaller than rock density): when head losses are small, the gap between brine pressure and geostatic pressure in the dilated zone is larger when this zone moves upward, making the process potentially unstable.

Under suitable conditions, all rocks can lose their subsurface sealing capacity. For engineering (short-term) applications, the criteria for leakage through halite and magnesium salts have been studied experimentally. However, the conditions in an abandoned cavern roof under which (localized) permeation or hydraulic fracturing will occur at low overpressures and over long periods, are not sufficiently understood. This is not surprising, given the lack of microphysical understanding, the empirical nature of the results and the need to extrapolate.

When a rocksalt seal is breached in nature, microstructural evidence suggests that, in nature, both diffuse dilatancy and hydro fracturing can occur at very low effective stress conditions — i.e. at near-lithostatic fluid pressure and differential stress of a few MPa in the deep subsurface, accompanied by crack healing through precipitation from supersaturated solutions.

Also, in engineering studies, both end-members permeation and hydraulic fracture are known. At first approximation, permeation is thought to become possible when fluid pressure is slightly higher than the local minimum principal stress in the salt; and hydraulic fracture when fluid pressure becomes significantly higher than the local minimum principal stress in the salt (several MPa).

Although the microphysical parameters controlling these processes are not well-understood, but measurements point to differences between samples and heterogeneous permeation parameters, giving rise to the controversy studied in this project.

Note that in the rock salt surrounding a cavern, when brine pressure is rapidly increased, the state of stresses is heterogeneous, non-isotropic and evolves over time. For instance, in the cavern-scale report of this KEM project it is stated:

“These relations above between the shut-in pressure and the least compressive stress is accepted by most authors when rock behavior can be reasonably considered as elastic (i.e., reversible): when fluid pressure in the well is equal to the overburden pressure, the state of stresses in the rock mass equals what it was (“virgin state of stress”) before the well was drilled. This is incorrect in the case of rock salt"

In-situ permeability of undisturbed halite can be very low at $10^{-21}$ $m^2$, but higher permeabilities also have been reported. This allows rock salt to seal fluids and fluid pressure cells over geologic time. The main controlling microphysical parameter here is the structure of grain boundary porosity which can be unconnected by fluid assisted healing processes, or connected by dilatancy or the presence of second phases **(Fig. 51)**. Grain size will play an important role in both cases.

Existing data suggest that permeation will start when excess pressure rises above zero (although the effect of deviatoric stress on this has not been explored well), the transition to hydrofracturing is not well known. Therefore, it is not possible at this point to predict accurately at which excess pressure hydrofracturing will dominate above slow permeation.

Fluids can move through rock salt over geologic time, in fractures, in dilatant grain boundary networks and in Grain Boundary triple junction channels. Opening and sealing of transport paths is an important process in this migration.

Infiltration will start (i) if the salt is permeable, for example through impurities, (ii) if fluid pressure raises to equal the minimum stress even in practically impermeable salt, (iii) if P-T conditions allow grain boundary triple junction channels and pore pressure is close to the minimum stress or (iv) at low effective stress and significant differential stress by dilatant fracturing.

The experiments on which these models are based provide very little microstructural information at present, so extrapolation to longer timescales is difficult because the microphysics is not well known. Even in samples selected to be homogeneous, permeation is heterogeneous at the scale of the samples. Re-sealing processes are not well understood

In pure, very low permeability rock salt, infiltration rate increases rapidly with the difference between fluid pressure and minimum principle stress: the available results suggest that if this difference is more than about 2 MPa, the flow is so rapid that it can be considered a hydraulic fracture. In abandoned caverns, this can lead to localized and rapid leakage of brine from the cavern, and hydrofracturing in the overburden if the high-pressure fluid permeating the salt reaches the overlying sediments.

In current numerical models of infiltration around caverns, it seems that the surrounding salt is assumed to be homogeneous. We propose that if realistic heterogeneous properties are included, infiltration will be much more localized, tending towards preferential fingering. One key parameter here is inferred to be the first order fabric of the salt: the layering. In bedded salt, this is sub horizontal, in domal salt this is sub vertical, suggesting major differences between permeation processes in these two settings.

Accurate prediction of the rate and distribution of percolation of fluids into the salt roof of solution mining caverns is a hard problem, requiring understanding of several coupled, nonlinear processes.

Although the main macroscale processes are recognized, further progress will require microphysical understanding of processes and parameters.

## Experimental study of permeation at high fluid pressure

It is reasonably well established that in very low permeability salt, if the fluid pressure becomes approximately equal to the minimum principal stress in the salt, the fluid starts to permeate along dilatant grain boundaries **(Fig. 52)**. These experiments use cylindrical or spherical samples **(Fig. 55)** with brine in a central cavity at controlled pressure. If the fluid pressure exceeds the minimum principal stress in the salt by more than 2 MPa, the risk of rapid permeation and hydrofracturing becomes high. However, it is not easy to use this criterion in practice because the minimum principal stress in salt around a cavern is not accurately known and also changes during solution mining and abandonment.

Although it is reasonably well established that the rate of infiltration increases nonlinearly with excess pressure **(Fig. 53, 56)**, it is also clear different samples of salt (with different microstructure, fabric or heterogeneities) have strongly different permeation kinetics **(Fig. 54)**. Heterogeneities like large Halite crystals, strong crystallographic fabric or anhydrite layers, (these have a different structure in bedded salt and in domal salt), have a major effect on permeation. Especially horizontally continuous, significantly thick impermeable anhydrite layers can form barriers, but may also allow fluid pressure to rise to more than 2 MPa above the minimum stress, leading to hydrofracturing when finally breached.

In the Clausthal experiments, the fluid contained fluorescent marker, so that the permeated volumes could be visualized post-mortem. These results suggest that permeation was predominantly along grain boundaries, but we note here that there is no microscopic evidence presented to demonstrate that this is indeed the case.

Noting the almost total absence of microphysical studies of the permeated samples, we infer that extrapolation of these results to low excess pressures and long time scales is not possible. For example, over long time scales grain boundary healing processes may or may not become more important than in short-term experiments. **Perhaps the most important practical conclusion one can draw from compilations of permeation experiments is that the rate of this can vary by orders of magnitude between different salts. This implies that in nature, permeation will be strongly heterogeneous, much more than suggested by current numerical models.**

The permeation experiments are usually presented separately from triaxial experiments at significant deviatoric stress **(Fig. 60, 61)** where grain boundary microcracking leads to dilatancy and increase of permeability. Here we note that the part of this dilatancy transition at low deviatoric stress, low confining pressure is probably the part relevant to permeation around abandoned caverns, but this part of the curve is not well studied, and its connection to the experiments discussed above **(Fig. 52)** is not clear although they must belong to a continuous trend. In addition, it is not clear how this transition would be if brine was used as a pore fluid instead of gas, and what the microphysical grain boundary dissolution-precipitation processes are. A first approximation of this could be made by combination of these results with the pressure solution models discussed above **(Fig. 35),** and with models of crack healing by dissolution-precipitation processes **(Fig. 64).** Note however that these experiments did not study grain boundaries but cracks in single crystals which heal much faster than grain boundaries. The kinetics of grain boundary healing are not well known and need further research to quantify and predict over long time scales. Finally, we mention briefly recent papers **(Fig. 65)** who proposed that thermodynamically controlled re-equilibration of grain boundary triple junction fluid connectivity could provide an additional process of permeation, perhaps assisted by deviatoric stress. This model was critically discussed by Brückner et al., (2016) albeit without microstructural investigations, which bring their conclusions in question.

## Natural examples of permeation

In the South Oman Salt Basin (SOSB), diapirs of Infra-Cambrian Ara Salt enclose isolated, often overpressured, carbonate reservoirs. Hydrocarbon-impregnated black halite provides direct evidence that this salt repeatedly has lost and then restored its sealing capacity **(Fig. 57, 58)**. The black staining is caused by intragranular microcracks and grain boundaries filled with solid bitumen that was formed by alteration of oil. The same samples show evidence for crystal plastic deformation and dynamic recrystallization, and subgrain-size piezometry indicates a maximum past differential stress of less than 2 MPa. Under such low shear stress, laboratory-calibrated dilatancy criteria indicate that oil can enter the halite only at near-zero effective stresses — i.e. at fluid pressures very close to lithostatic. In our model, the oil pressure in the carbonate reservoirs increased until it was equal to the fluid pressure in the low, but interconnected, porosity of the Ara Salt plus the capillary entry pressure. When this condition is met, oil starts flowing into the halite, which dilates and increases its permeability by many orders of magnitude. Sealing capacity is lost, and fluid flow will continue until the fluid pressure drops below the minimum principal stress, at which point the rock salt will reseal to maintain the fluid pressure at lithostatic values.

This study is a good example of natural localized permeation in rock salt, followed by resealing of the grain boundaries, showing that slow deformations in a salt system can lead to natural permeations.

In a study by Schléder et al. (2008), naturally deformed rock-salt core samples from the Fulda basin are analysed. The halite is folded into tight, isoclinal folds and is cut by an undeformed, 1-cm thick, coarse-grained halite vein **(Fig. 59)**. The microstructures were investigated in etched, gamma-irradiated thin sections from both the wall rock and the vein. Deformation microstructures are in good agreement with the solution-precipitation creep process. Strength variations in anhydrite-rich and-poor layers account for the strong folding in the halite beds. The vein is sealed completely and composed mainly of euhedral to subhedral halite grains, which often overgrow the wall-rock grains. Those microstructures, together with the presence of occasional fluid inclusion bands suggest that the crystals grow into a solution-filled open space. Based on considerations on the maximum value of in-situ differential stress and dilatancy criteria, as well as on the amount of fluid and the volume change, it is proposed that the crack was generated by natural hydrofracturing of the rock salt by the near-lithostatically pressured brine. In-situ paleo-differential stress values are not available for the samples because of the absence of subgrains. This suggests that the differential stress was below 0.3 MPa; otherwise, the fine-grained wet halite described (grain size D = 0.5-1 mm) would have developed subgrains. Model calculations suggest that this fine-grained salt is very weak as compared to domal salt, and it deforms relatively fast (e.g., for T = 353 K and D = 0.5 mm) even at low differential stresses. This study is a good example of natural hydrofracturing in rock salt, followed by resealing of the fracture, showing that slow deformations can lead to natural hydrofracturing in a salt system if the fluid pressure is sufficiently high.

## Numerical modelling of permeation

The numerical modelling of fluid permeation into an impermeable polycrystal is a hard problem. **Fig. 62 and 63** show examples of numerical simulations of the process. These simulations tend to produce rather large volumes of permeated salt, into which the brine is driven by the cavern convergence.

In the publications on these simulations the full details of the discretization and material property distribution are usually not given. In the author's opinion, these results may be the consequence of assuming a model with homogeneous material properties, and the discretization scheme.

Based on the discussion above, we propose that if realistic heterogeneities in the rock salt surrounding the cavern would be included in these models, the results would show much more heterogeneous, preferential permeation, with much less brine stored in the salt **(Fig. 62, right picture).** There are several feedback systems to consider here, and one of these is that an instability in the propagation front will be amplified by the density difference between fluid and rock salt, so that the excess pressure becomes higher in the part of the fluid which propagates faster. It is not clear how much faster the brine would reach the top of the salt in this case, and at what rate the brine would flow into the overlying sediments, possibly resulting in hydraulic fracturing. Clearly more work is needed here, and this is discussed in the cavern scale part of the report.

Discrete element modelling of the permeation process has seen first attempts (**Fig. 66)** in the salt engineering community. While it is an interesting approach and may in the future be used in multiscale nested modelling, at present the scaling of the models is many orders of magnitude different from the ratio of grain size (including heterogeneities) to the size of the salt dome. What these models do show, is that once you introduce even small heterogeneities in the model, permeation rapidly becomes localized.

## Conclusions, Microscale Permeation Mechanisms of salt rocks

Rock salt is known as the best subsurface seal for fluids, based on three key properties. First, the near-isotropic stress state provides resistance to hydrofracturing. Second, in-situ permeability in pure, equilibrated rock salt in nature is very low, as porosity is unconnected are very small. Third, plastic deformation of halite in nature is ductile and therefore non-dilatant. Fourth, fractures can be healed by ductile flow and solution-precipitation processes.

Under suitable conditions, however, all rock salt can lose their subsurface sealing capacity. For engineering (short-term) applications, it has been established that if the fluid pressure is several MPa higher than the minimum principal stress in the salt, rapid propagation of fluids through hydraulic fractures will initiate. The exact pressure for this depends on the rock salt, on material parameters like grainsize and anhydrite layers.

In-situ permeability of undisturbed halite can be very low at $10^{-21}$ $m^2$, but higher permeabilities also have been reported. This allows rock salt to seal fluids and fluid pressure cells over geologic time. The main controlling microphysical parameter here is the structure of grain boundary porosity which can be unconnected by fluid assisted healing processes, or connected by dilatancy or the presence of second phases. Grain size will play an important role in both cases.

In pure, very low permeability rock salt, infiltration rate increases rapidly with the difference between fluid pressure and minimum principle stress: the available results suggest that if this difference is more than about 2 MPa, the flow is so rapid that it can be considered a hydraulic fracture.

It has been shown in both laboratory experiments and in-situ tests that brine (and oil) can migrate through salt along diffusely dilatant salt-grain boundaries if the fluid pressure is higher than the minimum principal stress but not high enough to cause hydraulic fractures. It is not well known what the effect of a deviatoric stress in the rock salt is on the conditions of permeation. Some models argue that in an abandoned salt cavern's roof, this permeation process is slow, permeating a large volume of rock salt and creating a volume of dilated salt that both hosts significant brine and changes its permeability by nonlinear effective pressure-dependent dilatancy. However, considering the nonlinear nature of this effective pressure-dependent dilatancy in a heterogeneous rock-salt mass, others argue that this process is unstable and will lead to preferential fingering — or even fracturing — and a fluid pathway with a much smaller volume. This is especially true, as brine density is smaller than rock density: when head losses are small, the gap between brine pressure and geostatic pressure in the dilated zone is larger when this zone moves upward, making the process potentially unstable. In our opinion all available evidence points to this second process, when the fluid overpressure is small. In abandoned caverns, this can lead to localized and rapid leakage of brine from the cavern. Large scale diffuse dilatancy and homogeneous permeation is unlikely to occur after abandonment.

Controlling parameters of the process are salt microfabric, the state of stress and the amount of fluid pressure. The infiltration along fronts that are irregular even in the laboratory scale has been studied in bulk experiments and in solution-mined cavities, and first-order trends in evolution of dilatant porosity and permeability of the salt were measured. However, **the microphysics of this process is not well understood and more materials-science based work is needed to establish the likely long-term processes. It has been shown that dilatant (permeated) rock salt can undergo complex dilation-**

**compaction, recrystallization, crystal plasticity and pressure solution; therefore, a number of potential feedback processes are present.**

The conditions in an abandoned cavern roof under which (localised) permeation or hydraulic fracturing will occur are not sufficiently understood given the lack of microphysical understanding, the empirical nature of the results and the need to extrapolate.

Measurements point to differences in the rate of infiltration and permeation between homogeneous samples and predict spatially heterogeneous permeation. In heterogeneous samples, permeation is much more heterogeneous, controlled by the distribution of heterogeneities.

A First look at rock salt core from around Dutch solution mined caverns clearly shows that these heterogeneities are present and important (see part 3 of this report). In bedded salt, this anisotropy is subhorizontal, in domal salt this is subvertical, suggesting major differences between permeation processes in these two settings.

At present there is no microstructural information to quantify the grain boundary dilation and healing processes, although the toolbox to study this is readily available.

We also note that in an abandoned cavern, when brine pressure is increased, the state of stresses in the surrounding salt is quite different from lithostatic. This adds an additional uncertainty to predictions of the allowable brine pressure.

A number of processes are known to create an increase of permeability in initially impermeable or poorly permeable material. The first is microcracking and associated dilatancy, which can take place at high differential stresses or high fluid pressures. Dilatancy in the presence of brine has been shown to be suppressed by pressure solution and recrystallization processes. **However, the conditions relevant to abandonment, with low differential stresses and high fluid pressures, are largely unexplored. These conditions are complex indeed: in the case of permeation, they include the establishment of a pore pressure in rock salt where Terzaghi's law is added to the time-dependent elastoplastic material law.**

In numerical models of infiltration around caverns, it seems that the surrounding salt is assumed to be homogeneous. We propose that if realistic heterogeneous properties are included, infiltration will be much more localized, tending towards preferential fingering. The evolution of an abandoned cavern which leaks by preferential fingering has not been modelled and is not well understood.

Accurate prediction of the rate and distribution of percolation of fluids into the salt roof of solution mining caverns is a hard problem, requiring understanding of several coupled, nonlinear processes.

Although the main macroscale processes are recognised, further progress will require microphysical understanding of processes and parameters.

In bedded salt, horizontally continuous, impermeable anhydrite layers can form barriers which may allow fluid pressure to rise to more than 2 MPa above the minimum stress. Once these layers fail, the brine will rapidly penetrate the overlying rock salt.

Therefore, permeation of the salt after abandonment is much more heterogeneous than suggested by current models.

At present, there is very little information on the relevant microphysical processes, as the existing permeation studies did not carry out microstructural study of the deformed samples. Salt microstructure is variable and heterogeneous. Including microstructure in abandonment will help to better predict cavern evolution after abandonment.

The available evidence strongly suggests that infiltration will be localized but, further progress will require microphysical understanding of processes and parameters.

## Recommendation, microscale permeation.

It is clear from this review that the deficiency in microphysical understanding of the permeation process is substantially larger than that for deformation. We recommend

1. materials science-based research to provide a microphysical understanding of the permeation and healing processes to allow extrapolation to low excess pressure and long time scales, starting with state of the art microstructural study of samples tested in the permeation experiments, and designing suitable experiments to explore processes at lower excess pressure, low deviatoric stress and long time scales. This process may well be studied efficiently in transmitted light microscopy, or live micro-CT, to collect dynamic data in-situ.
2. **Such a project would integrate laboratory measurements of stress-strain and permeation response of rock salt samples with microscopic and microanalytical characterization of the undeformed and permeated materials. This will provide quantitative understanding of the microphysical processes operating in the samples, provide a basis for the right conditions of the permeation experiments and allow extrapolation of the measured**

**constitutive equations to low deviatoric stress and slow permeation time scales which can not be accessed in the laboratory. Examples of such projects are provided by many of the papers cited in this review. Site-specific analyses can be done using the core materials taken in the well from which the cavern was developed. Validation of the predictions for can be done by comparison with natural, permeation of salt structures.**

3. Use this understanding of microphysical processes of permeation to upscale these to constitutive models for long time scales and incorporate heterogeneous material, to be implemented in dome and cavern-scale computations. Here, a link to geoscientific magma migration models is suggested, as this process is in many respects similar.
4. Develop a standardized, automated microstructure characterization protocol to integrate macro and microscale analysis or rock salt samples.

# Part 3: Microstructural study of salt core from the Netherlands

This part of the report presents the results of a microstructural study of representative rock-salt core from the Netherlands salt-cavern drillings, (**Fig. 67)** undertaken to demonstrate the currently available practical set of analytical techniques (optical microscopy, electron microscopy, microchemical analysis, etc.) with automated image analysis to quantify the set of key parameters required to provide a benchmarked description in future analyses. unfortunately, the scope of this project did not allow full analysis in combination with deformation experiments: these are recommended in a follow-up study.

## Description of microstructures

We sampled drill cores from the Barradeel (BAS) and the Zuidwending (ZW, WSN) area, after consultation with NURYON and FRISIA. After viewing selected core boxes in the Zeist core store, we selected representative 20 cm samples which were slabbed and photographed by MaP, followed by selection of representative locations for thin sections.

Both sets of samples (layered and domal) have clearly recognizable bedding, different lithologies, anhydrite layers and highly heterogeneous grainsize in rock salt. The core samples consist of three main phases: 1. Anhydrite, associated with Mg/K-Sulfates, 2. Halite megacrystals, and 3. fine- to coarse grained recrystallized Halite. A summary is provided in (**Fig. 68).**

In BAS, (**Fig. 69-73 )** the layering is dominantly subhorizontal, with local folding setting the layers in vertical position. Anhydrite layers are 1 mm to 10 cm thick, and are present in every core investigated. In BAS, grain size in Halite is highly variable. Fine grained layers are typically 1-5 cm thick, alternating with 1- 2 cm thick layers which are essentially single crystals (called megacrystals here, in the German literature these are called Kristallbrocken). In BAS, as expected, there is only minor tectonic deformation, and most of the layers are intact.

In the domal samples (ZW, WSN), **(Fig. 74-80)** the layering is strongly tectonically disrupted but still recognizable. Anhydrite layers are 1 mm to 5 mm thick, and are present in every core investigated. They are boudinaged, folded, and often associated with Halite megacrystals which are also deformed, boudinaged and surrounded by recrystallized halite with finer grainsize, together with the original fine-grained material. In the domal samples (ZW, WSN), the Anhydrite layers are folded, and often associated with Halite megacrystals which are also deformed and boudinaged. Layering is strongly tectonically disrupted but still recognisable. Anhydrite layers are 1 mm to 5 mm thick, and are present in every core investigated. They are surrounded by recrystallized Halite with fine- to coarse grain size. Halite megacrystals in all samples were partly recrystallized and replaced by clear, fine-to coarse grained polycrystals, indicating grain boundary migration recrystallization. Grain boundaries of the recrystallized grains were straight to interlobate, often forming 120° triple junctions, pointing to static annealing of the microstructure. Next to the interbedded Anhydrite layers, all samples contained Anhydrite fragments and single crystals within the recrystallized areas. Particle size and distribution is thought to affect the recrystallized grain size.

Presence of Halite megacrystals together with fine- to coarse-grained recrystallized areas result in an overall bimodal grain size distribution of the Halite fraction in all (BAS, ZW, WSN) core. Halite megacrystals in all samples contained numerous inclusion (fluid inclusions and Anhydrite crystals). Most of the grain boundaries contain a large number of isolated fluid inclusions, usually arranged in arrays of similar sizes and shapes. Fluid inclusions in the BAS samples were typically irregular and patchy, while there were usually well equilibrated (often nearly spherical) and aligned in regular arrays or trails in the ZW and WSN samples.

High-resolution BIB-SEM images showed micro porosity within the Anhydrite layers, especially adjacent to the Mg/K-Sulfates.

## Conclusions, microstructural study

The three fabric elements identified in the core have strong and different effects on deformation rates and pathways for permeating fluids at pressure close to the minimum principle stress.

For the case of the bedded salts (BAS):

1. the very large cm scale contrast in grainsize in Halite indicates that there will be a large cm-scale variation in the contribution of pressure solution to the total strain. Together with the stronger, cm-scale distributed anhydrite layers this will cause a very large transverse anisotropy in effective viscosity which will have to be considered when modelling the squeeze mining process and convergence after abandonment.
2. Especially close to the cavern, the large strains are predicted to lead to strong recrystallization of the halite megacrystals and grain size reduction around these. After abandonment, this material will be much weaker than previously thought.
3. the Halite megacrystals are essentially impermeable and the observations in the cores suggest that they will remain intact during permeation. This means that the vertical resistance to fluid flow of the package of fine grained rock salt, megacrystals and fine anhydrite layers may be quite high, allowing pressures in the cavern to rise to values which can lead to hydrofracturing. This fabric may be disrupted locally by the deformation during cavern operation, causing highly localized permeation.

For the case of the domal salts (ZW, WSN):

1. the very large cm scale contrast in grainsize in Halite indicates that there will be a large cm-scale variation in the contribution of pressure solution to the total strain. Together with the stronger, cm-scale boudinaged and folded anhydrite layers this will cause a very large transverse anisotropy in effective viscosity which will have to be considered when modelling the squeeze mining process and convergence after abandonment.
2. the Halite megacrystals are essentially impermeable and the observations in the cores suggest that they will remain intact during permeation.
3. the Anhydrite layers are microporous and may be somewhat permeable (this needs further study)
4. the recrystallized finer grained halite will concentrate the strain and form the primary pathway for permeation. patterns and rate of permeation therefore will be controlled by the distribution

of recrystallized halite. Larger continuous Anhydrite layers and Halite megacrystals will channel permeation.

5. Therefore, based on the observations to date, we infer that permeation by fluids at pressure close to the minimum principle stress is going to be strongly heterogeneous and channelled along some of the sub-vertical layers.

### Recommendations, microstructural study

Considering the finding that the cores show many structures which point to strongly heterogeneous deformation and permeation, We recommend materials science investigations of deformation and permeation of the relevant core material, and modelling the evolution of these caverns during operation and abandonment based on these results.

## References cited in the text.

(somewhat unusually, the papers referred to in the figures are cited in the figure captions). The list of over 2000 items from the database is made available to SodM.


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# Figures

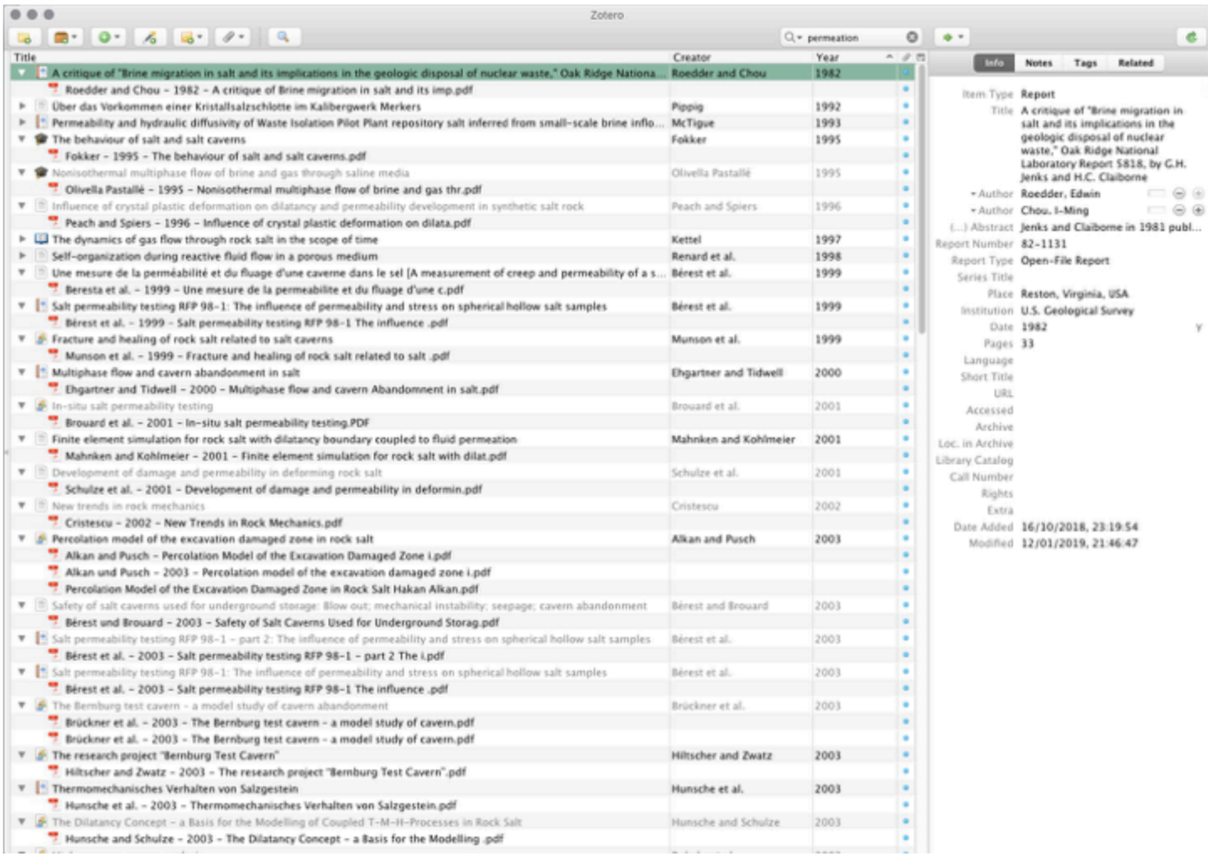



*Figure 1. Screenshot of the Zotero database used in this study.*

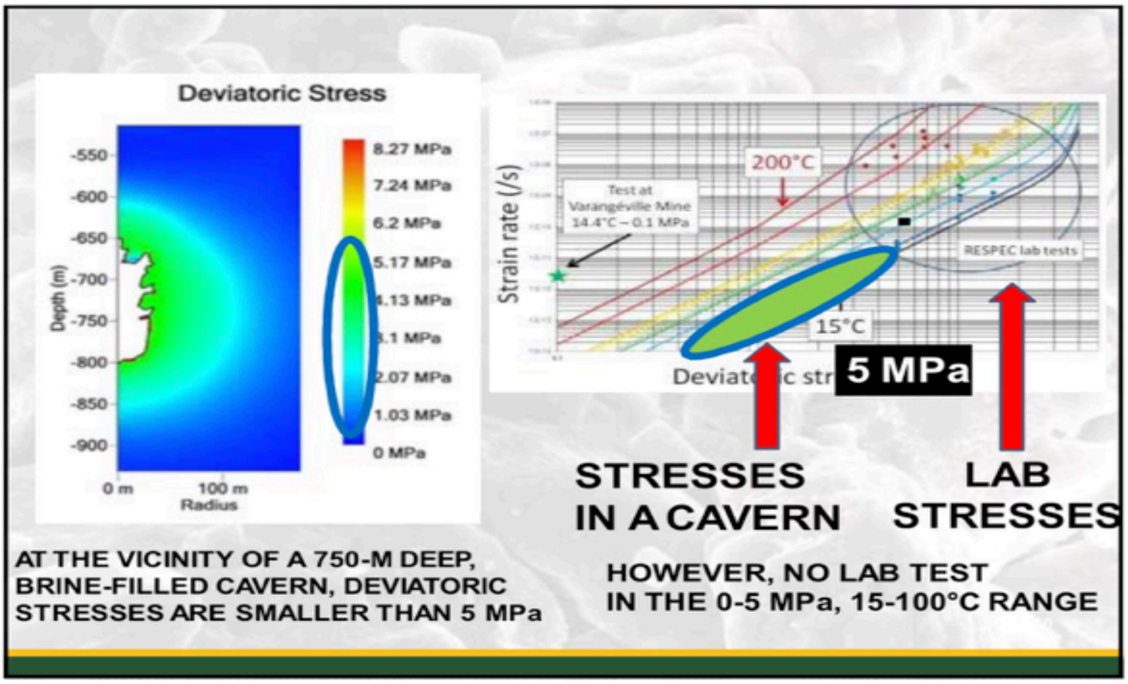



*Figure 2 Deviatoric stresses in the vicinity of brine-filled caverns are generally lower than the stresses applied in laboratory experiments, and in the far field much lower. This means that experimental data must be extrapolated to compute deformation and fluid permeation around caverns. From: Pierre Bérest, Benoit Brouard, Dieter Bruckner, Kerry DeVries, Hakim Gharbi, Grégoire Hévin, Gerd Hofer, Christopher Spiers, Stefan Stimmisher, Janos L. Urai: Very Slow Creep Tests on Salt Samples. 8th US/German Workshop on Salt Repository Research, Design, and Operation - Middelburg, The Netherlands September 6-8, 2017.*

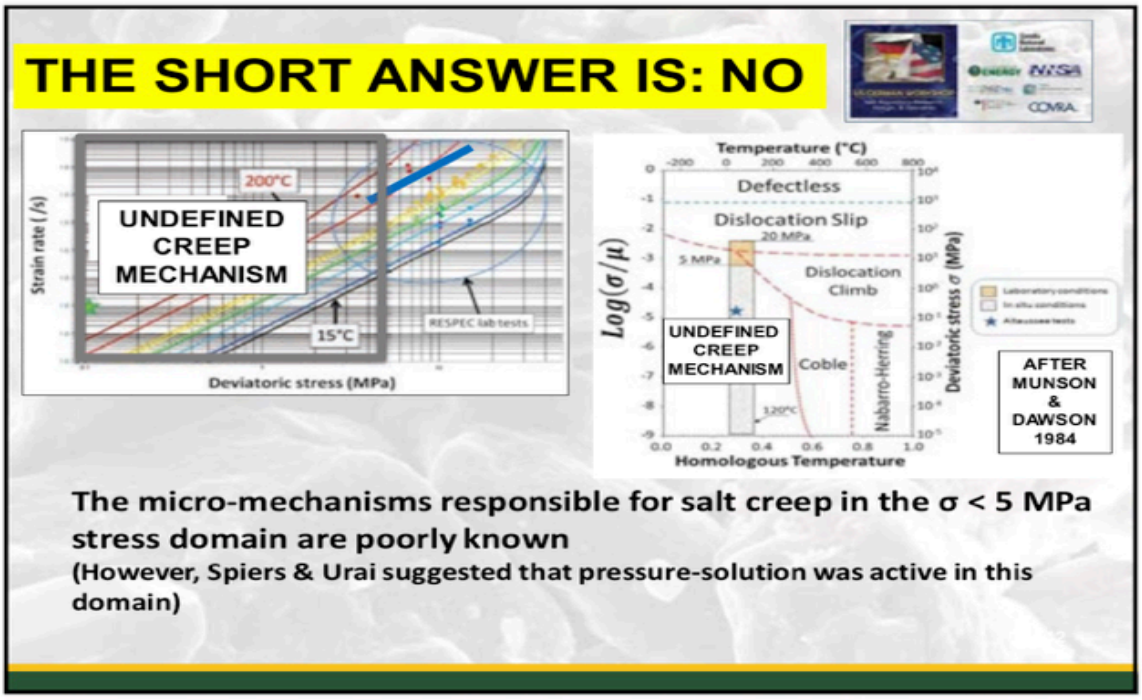



*Figure 3. The microphysical basis for this has been well known in the materials science/ geoscience literature for 30 years, however this knowledge has not been implemented in the salt engineering practice. A good illustration of this is the above presentation. From: P. Bérest, B. Brouard, D. Bruckner, K.y DeVries, H. Gharbi, G. Hévin, G. Hofer, C. Spiers, S. Stimmisher, J.s L. Urai. Very Slow Creep Tests on Salt Samples. 8th US/German Workshop on Salt Repository Research, Design, and Operation - Middelburg, The Netherlands September 6-8, 2017*

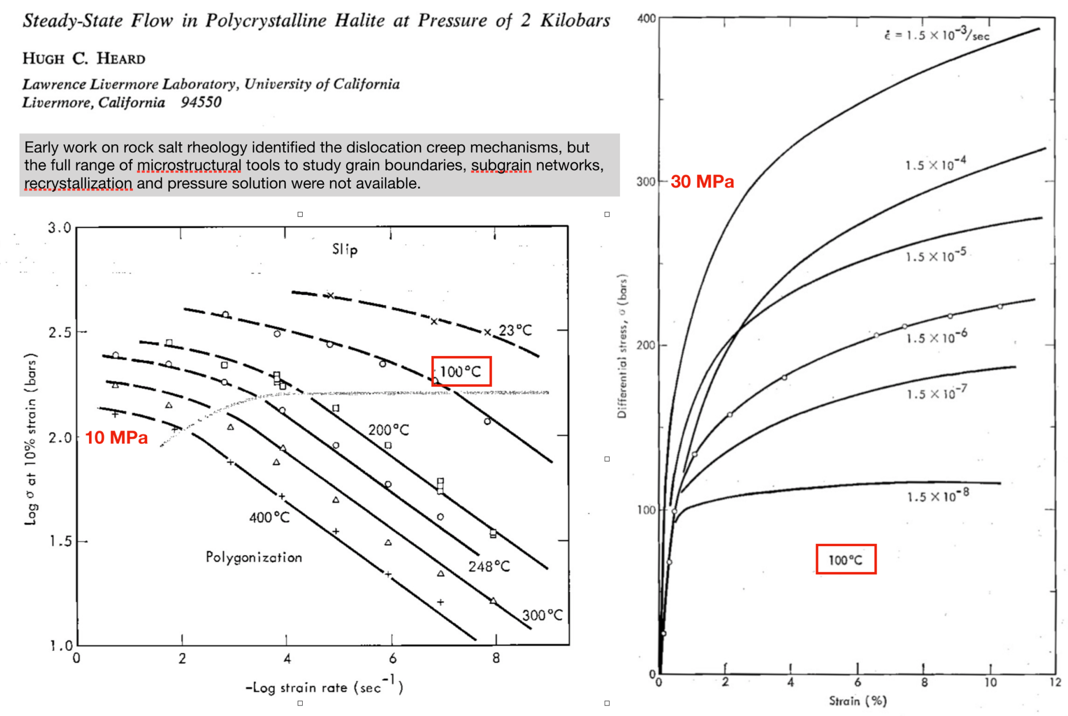



*Figure 4. Early studies of the deformation of polycrystalline Halite combined stress-strain measurements with microstructural observation along the lines of good materials science practice. Summary of the stress-strain data of triaxial tests on synthetic polycrystalline Halite, with a grainsize of 2-3 mm and a water content of around 30 ppm. After Heard, H.C., 1972. Steady-State Flow in Polycrystalline Halite at Pressure of 2 Kilobars. Flow and Fracture of Rocks, AGU Geophysical Monograph Series. 191–209.*

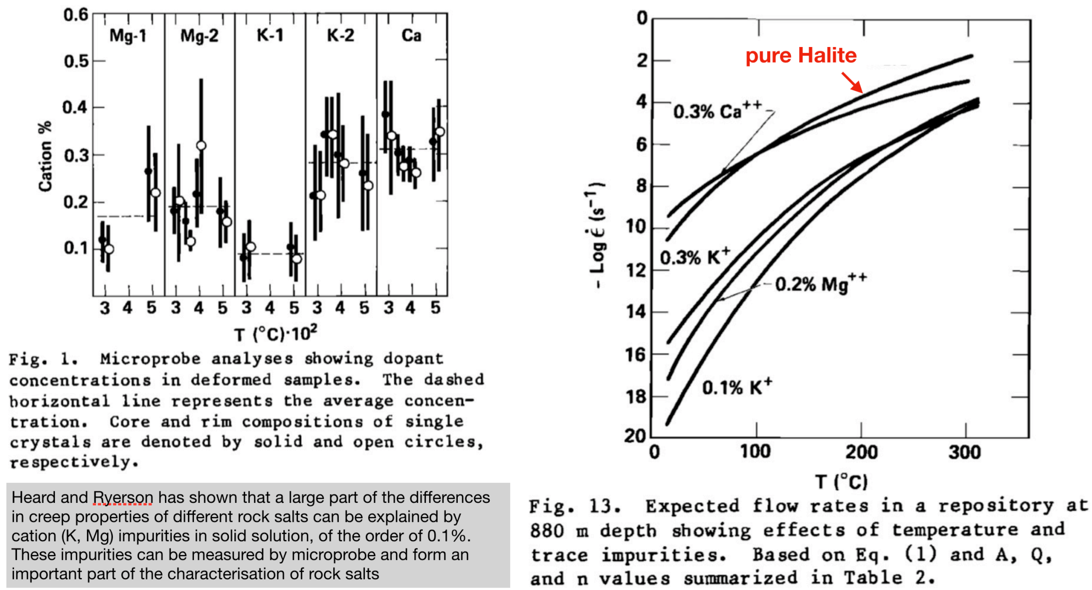



*Figure 5 Effect of cation impurities in solid solution on the steady-state flow stress in polycrystalline Halite. These results show how impurities in solid solution (there are not measured in current salt engineering) have an important effect on dislocation creep. After: Heard, H.C., Ryerson, F.J., 1986. Effect of cation impurities on steady-state flow of salt. In: Hobbs, B.E., Heard, H.C. (Eds.), Mineral and Rock Deformation: Laboratory Studies, Geophysical Monograph Series. American Geophysical Union, Washington, D. C., 99–115.*

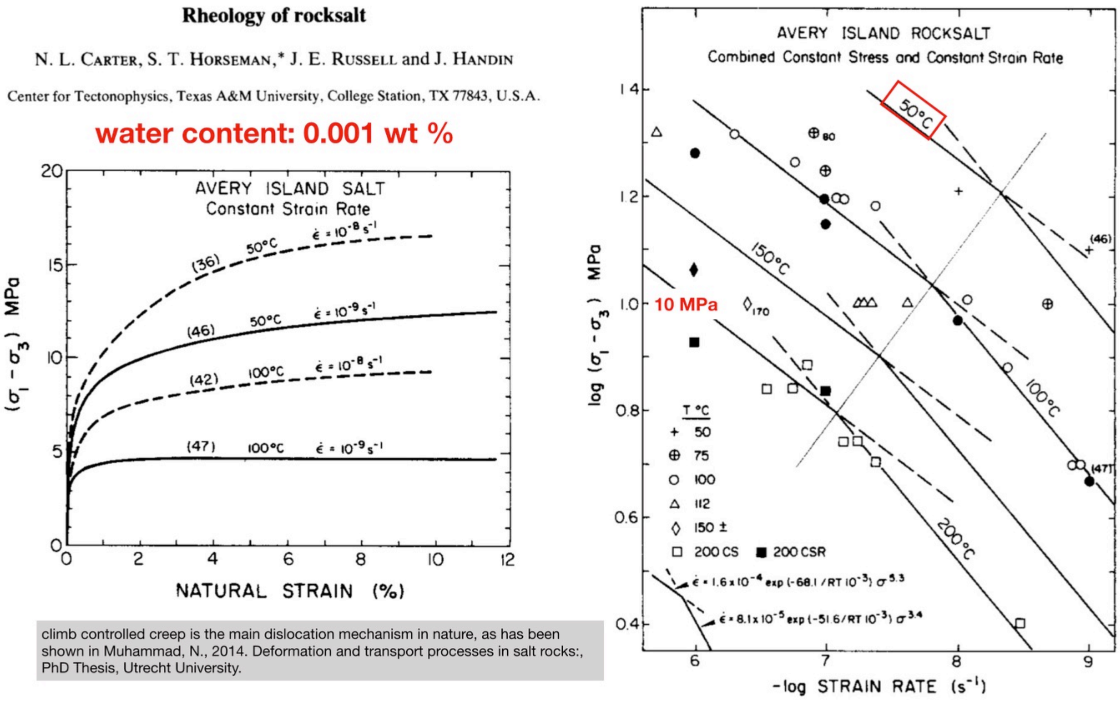



*Figure 6. Summary of the rheology of Avery Island rock salt. After: Carter, N.L., Hansen, F.D., 1983. Creep of rocksalt. Tectonophysics 92, 275–333 and Carter, N.L., Horseman, S.T., Russell, J.E., Handin, J., 1993. Rheology of rocksalt. Journal of Structural Geology 15, 1257–1271*

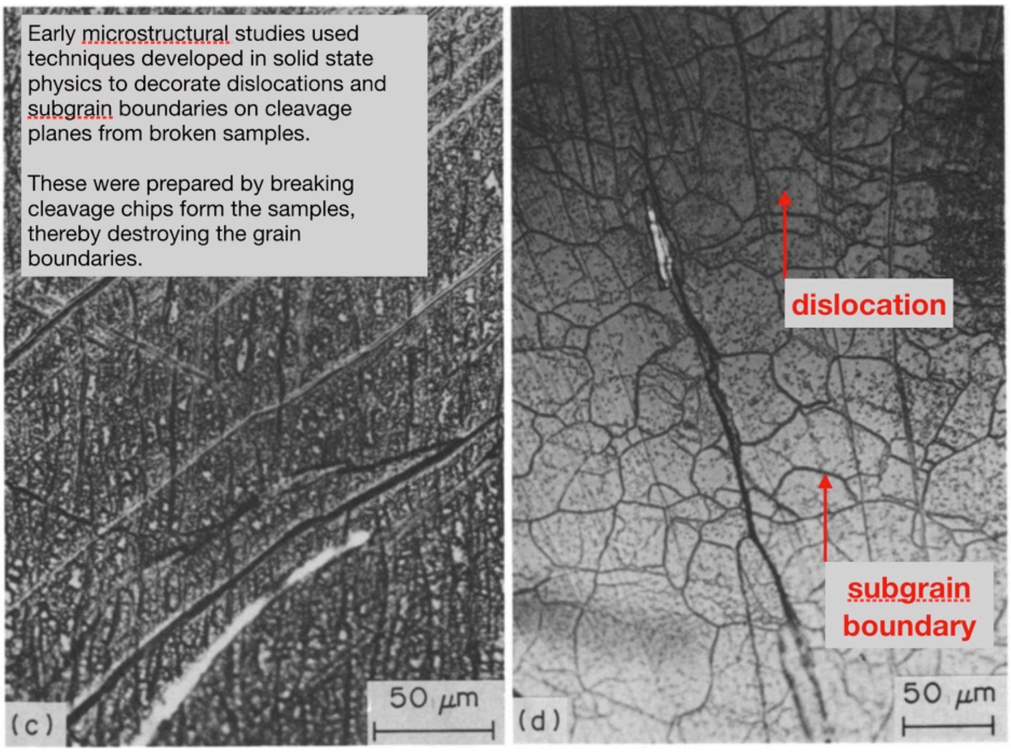



*Figure 7 Reflected light micrographs of cleavage chips from experimentally deformed Avery Island rock salt, showing dislocation etch pits and subgrain boundaries. These early studies did not have access to the grain boundary imaging techniques developed around 1984. After: Carter, N.L., Horseman, S.T., Russell, J.E., Handin, J., 1993. Rheology of rocksalt. Journal of Structural Geology 15, 1257–1271*

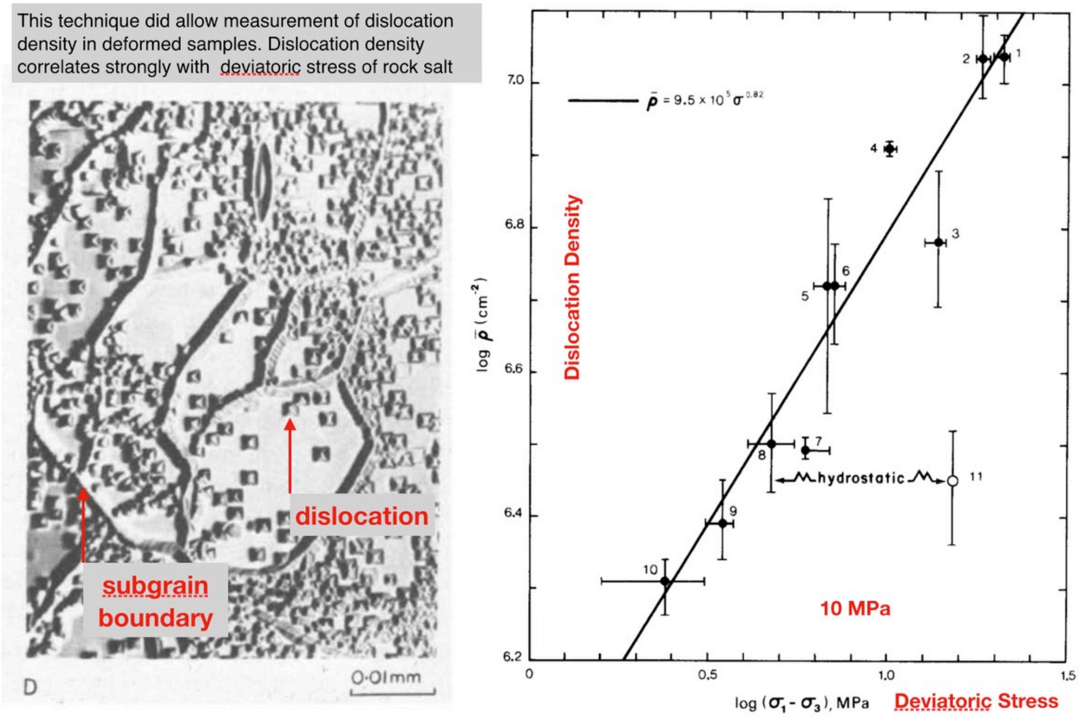



***Figure 8 Reflected light micrograph of a cleavage chip of experimentally deformed Avery island rock salt, showing dislocations and subgrain boundaries. Dislocation density correlated strongly with deviatoric stress. After: Carter, N.L., Hansen, F.D., 1983. Creep of rocksalt. Tectonophysics 92, 275–333.***

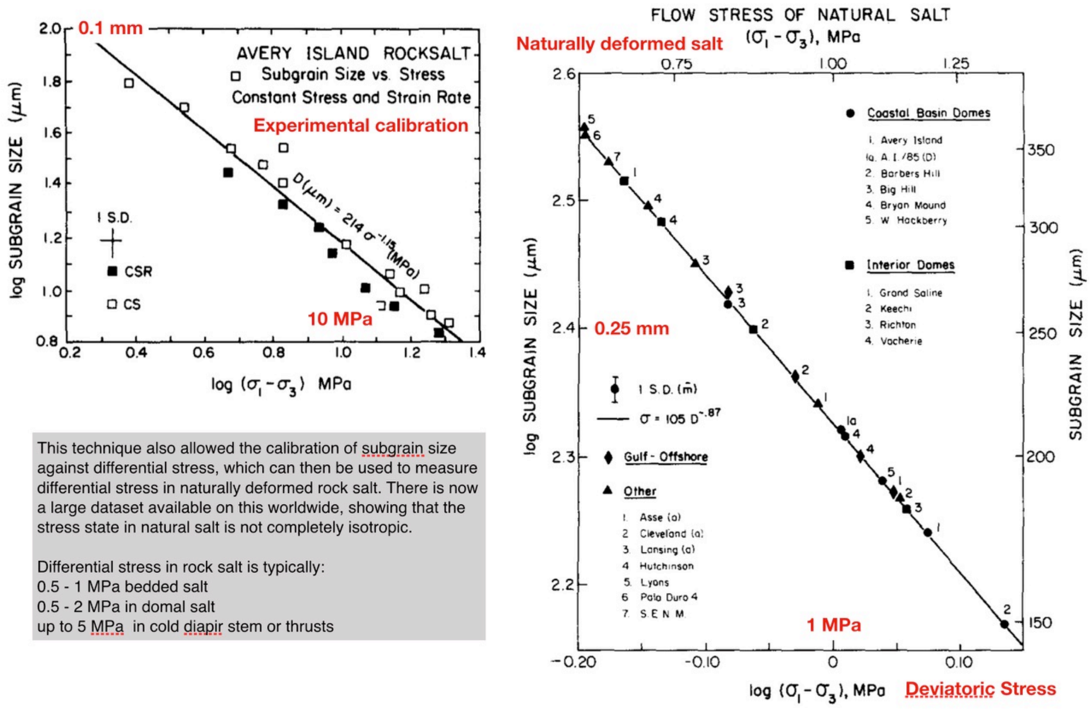



***Figure 9 Subgrain size correlates with deviatoric stress in deformed rock salt, providing the basis for measuring deviatoric stress in naturally deformed samples using microscopy: subgrain size piezometry. The graph on the right shows common values from different settings. This demonstrates that virgin in-situ stress in salt is not isotropic as assumed in current salt engineering studies. After: Carter, N.L., Horseman, S.T., Russell, J.E., Handin, J., 1993. Rheology of rocksalt. Journal of Structural Geology 15, 1257–1271***

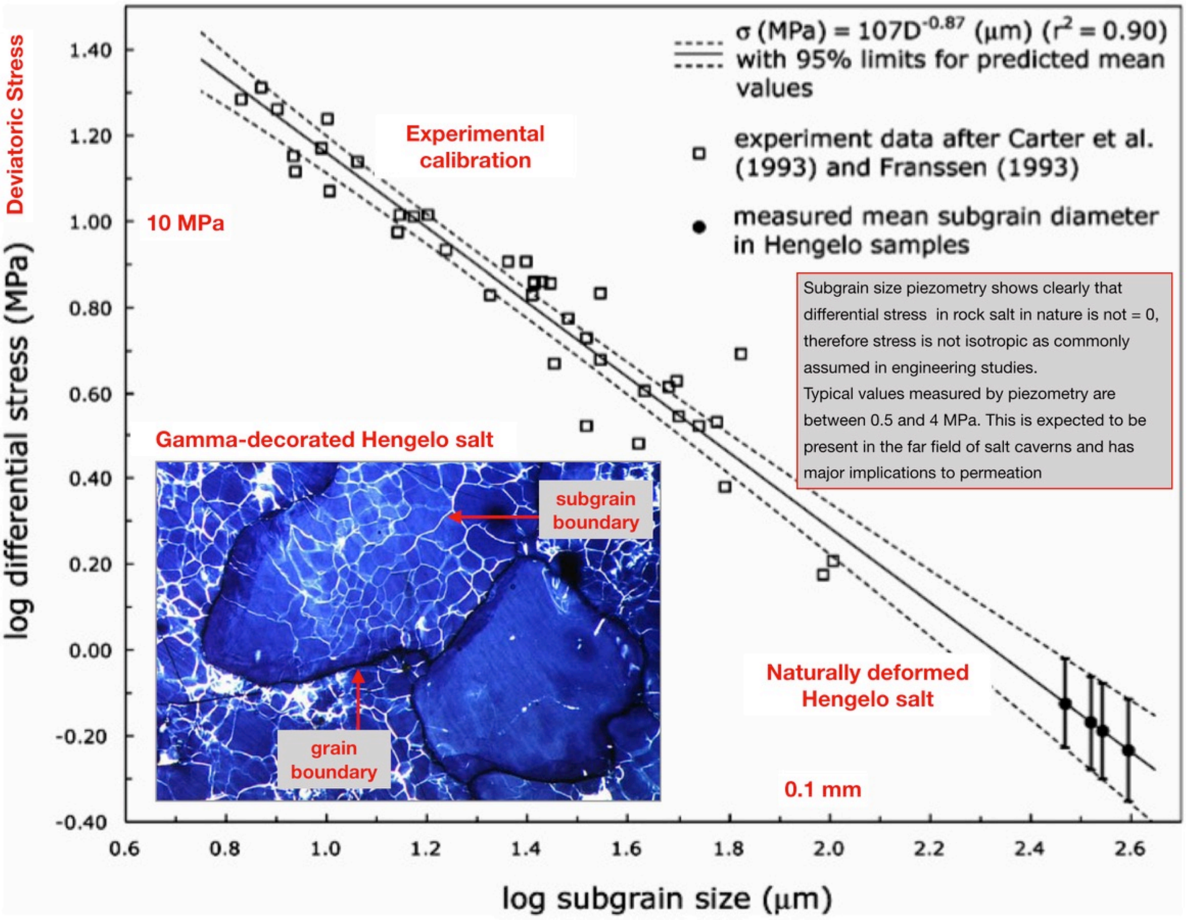



***Figure 10 Subgrain size piezometry using Gamma-decoration of microstructure in rock salt from a core from the Hengelo cavern field, the Netherlands. After: Schleder, Z., Urai, J.L., 2005. Microstructural evolution of deformation- modified primary Halite from Hengelo, the Netherlands. International Journal of Earth Sciences 94, 941–956.***

These experiments were done using artificially dried salt to switch off dynamic recrystallization. The transition from dislocation glide to dislocation climb controlled creep in **dry rock** salt was determined in the temperature range 22-350 C using the pressure sensitivity of stress to evaluate microphysical models.

The temperature at which the transition from glide to climb control takes place was found to lie in between 125 and 250 °C, at a deviatoric stress of about 16 MPa. At slow strain rates and low stresses normally relevant for in situ conditions, **dislocation creep of rock salt in nature was inferred to be controlled by dislocation climb**."

Muhammad, N., 2014. Deformation and transport processes in salt rocks:, PhD Thesis, Utrecht University.

Micrograph: Sample N142, T = 250 °C, $\dot{\varepsilon} = 1{\times}10^{-6}\ s^{-1}$, $\varepsilon_{max} = 0.16$, polygonised with clear grain boundaries, sub-grain size of 50 μm

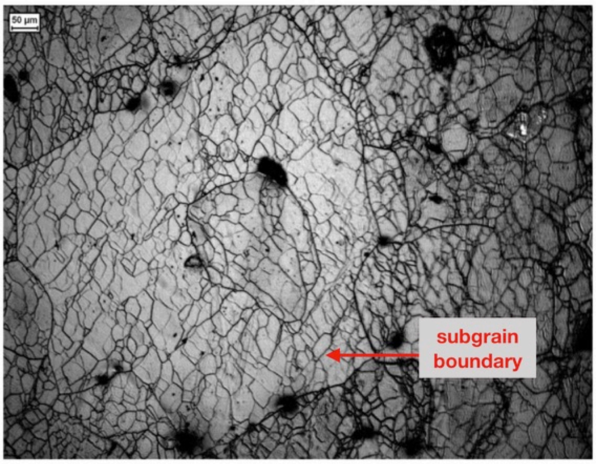



***Figure 11 Measurements of the pressure dependence of flow stress have shown that dislocation creep in nature is controlled by dislocation climb. After: Muhammad, N., C.J. Spiers, C.J. Peach & J.H.P. de Bresser, 2012. Effect of confining pressure on plastic flow of salt at 125 °C. In: Bérest, P., Ghoreychi, M., Hadj-Hassen, F., and Tijani, M. eds. Mechanical behaviour of salt VII, CRC press, pp. 57-64. and Muhammad, N., 2014. Deformation and transport processes in salt rocks, PhD Thesis, Utrecht University.***

Table 3.2. The updated Table of flow law parameters after Ter Heege *et al*. 2005a.

| Material/ composition | $H_2O$ [ppm] | $P$ [MPa] | $\dot{\varepsilon}$ [$s^{-1}$] | $T$ [°C] | $\sigma$ [MPa] | LOGA [$MPa^{-n}\ s^{-1}$] | Stress exponent $n$ | ΔU [$kJmol^{-1}$] | Source/ comments |
|---|---|---|---|---|---|---|---|---|---|
| Synthetic pure rocksalt | 5-10 | 50-600 | $4\times10^{-7}$ to $10^{-4}$ | 250-350 | 2.8-15.2 | -14.4 ±1.4 | 4.7 ±0.3 | 126 | This study (Chapter 2) |
| Synthetic pure rocksalt | 20–45 | 200 | $10^{-1}$ to $10^{-8}$ | 23-400 | 1.6-47 | 5.58 ±0.8 | 5.5 ±0.4 | 98 ±8 | Heard (1972) |
| Synthetic pure rocksalt | 20–45 | 200 | $10^{-1}$ to $10^{-8}$ | 23-400 | 1.6-47 | 0.7 ±0.4 | 5.8 ±0.2 | 96 ±3 | Heard & Ryerson (1986) |
| Natural (>95% rocksalt)a | ? | 14, 21 | $10^{-6}$ to $10^{-11}$ | 23-160 | 8.3-24 | 3.36-6.03 | 4.1 ±6.3 | 50-83 | Wawersik & Zeuch (1986) |
| Natural (>99% rocksalt)b | <100 | 2.5-20.7 | $10^{-6}$ to $10^{-9}$ | 50-200 | 6.9-20.7 | 3.8 | 5.3 ±0.4 | 68 ±4 | Carter *et al*. (1993) high $\dot{\varepsilon}$, $\sigma$ |
| Natural (>99% rocksalt)b | <100 | 2.5-20.7 | $10^{-7}$ to $10^{-9}$ | 100-200 | 2.5-10.3 | 4.09 | 3.4 ±0.1 | 52 ±1 | Carter *et al*. (1993) low $\dot{\varepsilon}$, $\sigma$ |
| Synthetic pure rocksalt | Dry | Uncof. | $10^{-3}$ to $10^{-7}$ | 250-780 | 0.4-14.8 | -0.76 ±0.2 | 5.7 ±0.3 | 129 ±8 | Franssen (1994) Low $T$ |
| Natural (rocksalt)c | ? | Unconf. + 15-20 | $10^{-3}$ to $10^{-11}$ | 30-250 | 1.7-40 | -- | 7 | 110 | Hunsche & Hampel (1999)d |
| Natural (>98% rocksalt)c | 500 | 3-30 | $3.5\times10^{-7}$ | 150 | 11-13 | -- | -- | -- | Peach *et al*. 2001 |
| Synthetic pure rocksalt | 9–46 | 50 | $10^{-4}$ to $10^{-7}$ | 75-200 | 7.2-22.4 | 1.56 ±0.54 | 5.6 ±0.5 | 80 ±6 | Ter Heege et al 2005 |

a: Range of parameters for natural rocksalt from five different locations: Salado (New Mexico), West Hackberry and Bayou Choctaw (Louisiana), Bryan Mound (Texas) and Asse (Germany).
b: Avery Island (Louisiana).
c : Asse Speisesalz (Germany).
d: In this study, mechanical data were fitted to a composite law. Stress exponent and activation energy quoted here are from their best fit to a power law equation.

unconf: unconfined

***Figure 12. After: Muhammad, N., 2014. Table of typical range of power law Deformation and transport processes in salt rocks, PhD Thesis, Utrecht University.***

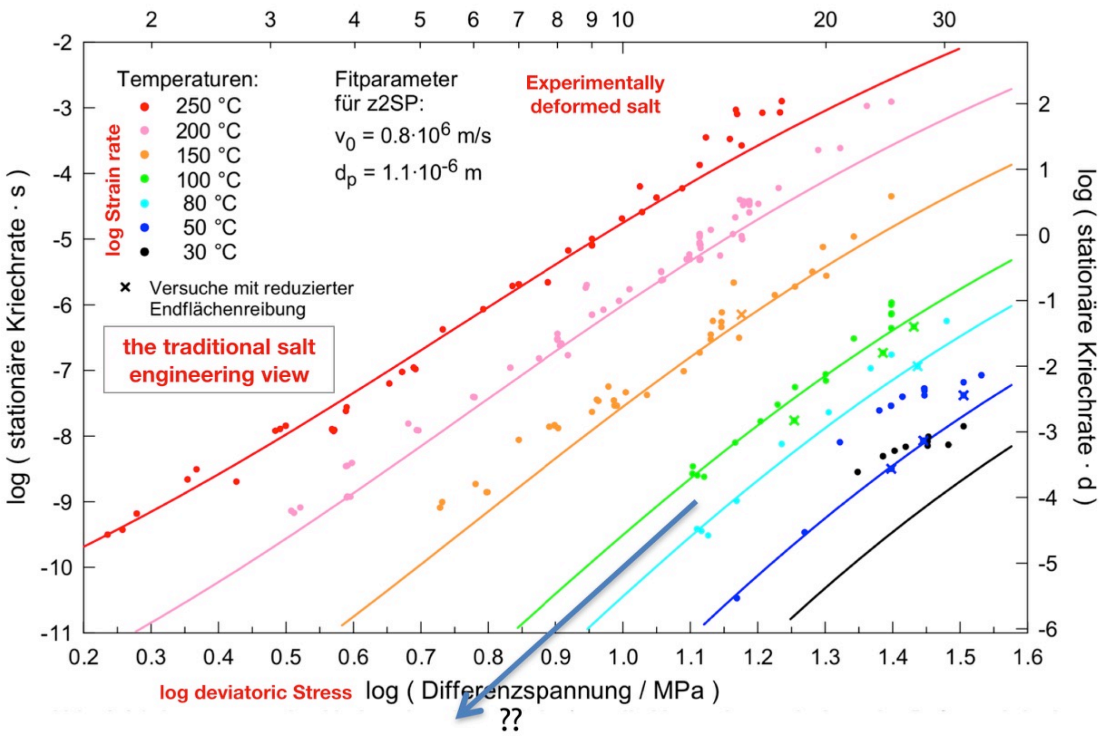



***Figure 13 Summary plot of steady state rate versus deviatoric stress in Zechstein rock salt, noting that these were done without microstructural study. This plot is based on an impressive number of creep experiments. The experimental facility is state of the art but there is no information on possible recrystallisation and effects of brine. Dilatant samples could have lost their inherent brine which stops pressure solution, and the dataset has to be extrapolated to allow description of salt creep at conditions relevant to salt caverns. This can be predicted using a materials science approach. The change in stress exponent which is missed in this dataset towards low differential stress is now increasingly recognised in engineering After: Hunsche, U., Schulze, O., Walter, F., Plischke, I.: Projekt Gorleben – Thermomechanisches Verhalten von Salzgestein. Abschlussbericht. Arbeitspaket 9G 213 811, Bundesanstalt für Geowissenschaften und Rohstoffe (BGR): Hannover, 2003.***

"Natural rock salt in deep underground structures is planned to act as the host material for the permanent storage of radioactive and toxic wastes. Dimensioning and safety analysis of such repositories require a model which allows to **predict the creep behaviour of rock salt on the basis of the microstructure and the physical mechanisms of deformation**.

Already before testing natural rock salt contains a subgrain structure. During deformation under constant stress the creep rate changes until a steady state is reached.

The same holds for the microstructural parameters subgrain size and spacing of dislocations inside subgrains. **The composite model of plastic deformation takes the observed heterogeneity of the dislocation structure explicitly into account.** Using this model in combination with appropriate kinetic laws for dislocation movement, transient as well as steady-state creep of rock salt can be calculated."

This is another compilation focussing on dislocation creep. NB: no measurements of microstructural parameters, are presented, such as water content, impurities, grain boundary structure, etc. It is not known if the in-situ water in the samples was preserved during the test ands if dynamic recrystallization and pressure solution operated. However, it is known that this can have large effects on mechanical properties when extrapolated to low differential stresses.

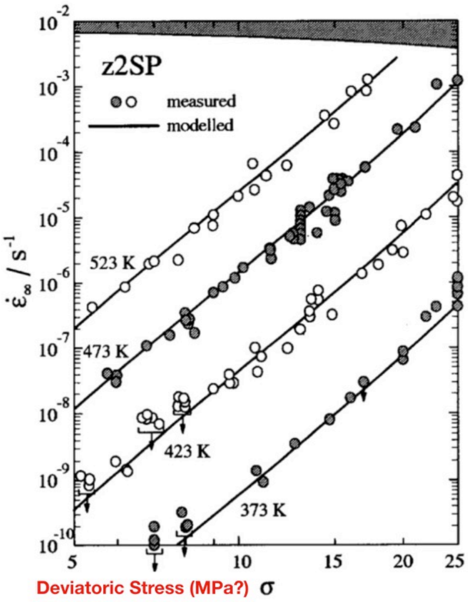



*Figure 14 Compilation and attempt to fit a creep law based only on dislocation dynamics, for Asse Speisesalz rock salt. Note that using this data to predict long-term deformation around salt caverns requires extrapolation, and extrapolation using the trend shown in this graph is not in agreement with other published studies and is not based on microphysical arguments, nor takes the well-known effects of water on grain boundaries into account. After: Weidinger, P., Hampel, A., Blum, W., Hunsche, U., 1997. Creep behaviour of natural rock salt and its description with the composite model. Materials Science and Engineering: A A234-236, 646–648.*

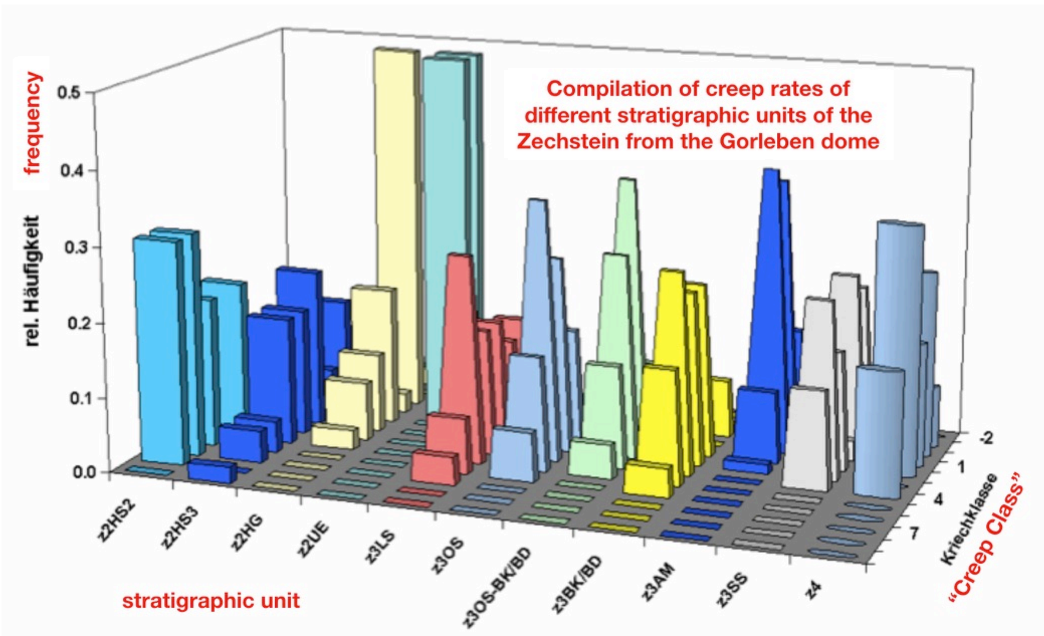



*Figure 15 Rock salt samples from different stratigraphic units have different creep properties as expected. In this compilation a series of "creep classes" with different parameters in the creep law are proposed for each unit. Kriechklasse 1 is "strong" – it creeps about 1000 times slower than Kriechklasse 9 under the same stress and temperature. NB: although this is an attractive approach, no measurements of microstructural parameters are presented, such as water content, impurities, grain boundary structure, etc. It is not known if the in-situ water in the samples was preserved during the test. Extrapolation of these trends to long term deformation around salt caverns is not supported by microphysical studies. After: Hunsche, U., Schulze, O., Walter, F., Plischke, I.: Projekt Gorleben – Thermomechanisches Verhalten von Salzgestein. Abschlussbericht. Arbeitspaket 9G 213 811, Bundesanstalt für Geowissenschaften und Rohstoffe (BGR): Hannover, 2003.*

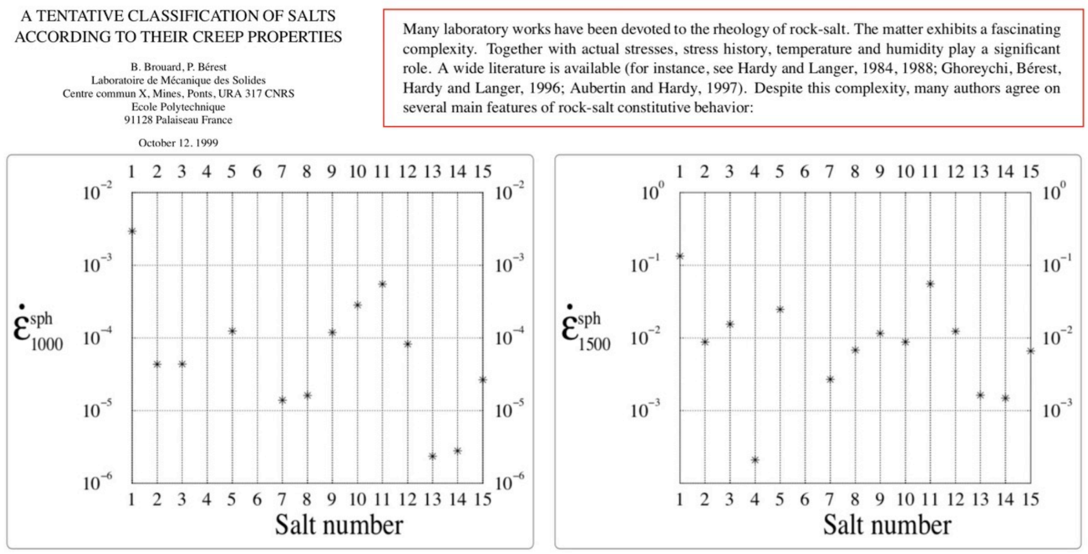



*Figure 16 Data collected by Brouard and Bérest show a similar picture: caverns in different rock salts creep about 100 - 1000 times slower than others under the same stress and temperature. This classification is an attractive approach, but at this point, these data cannot be correlated to microstructural features which can be measured in drill core. Also, it is not known how these data extrapolate to long term creep after abandonment. In this paper, published creep laws of a wide range of different salts were used to calculate cavity closure rates at different depths of 1000 and 1500m, with gas fill at 8 MPa. After: Brouard, B., Bérest, P., 1998. A tentative classification of salts according to their creep properties. Presented at the SMRI Spring 1998 Meeting, 19–22 April 1998, Solution Mining Research Institute, New Orleans, Louisiana, USA, 18–38.*

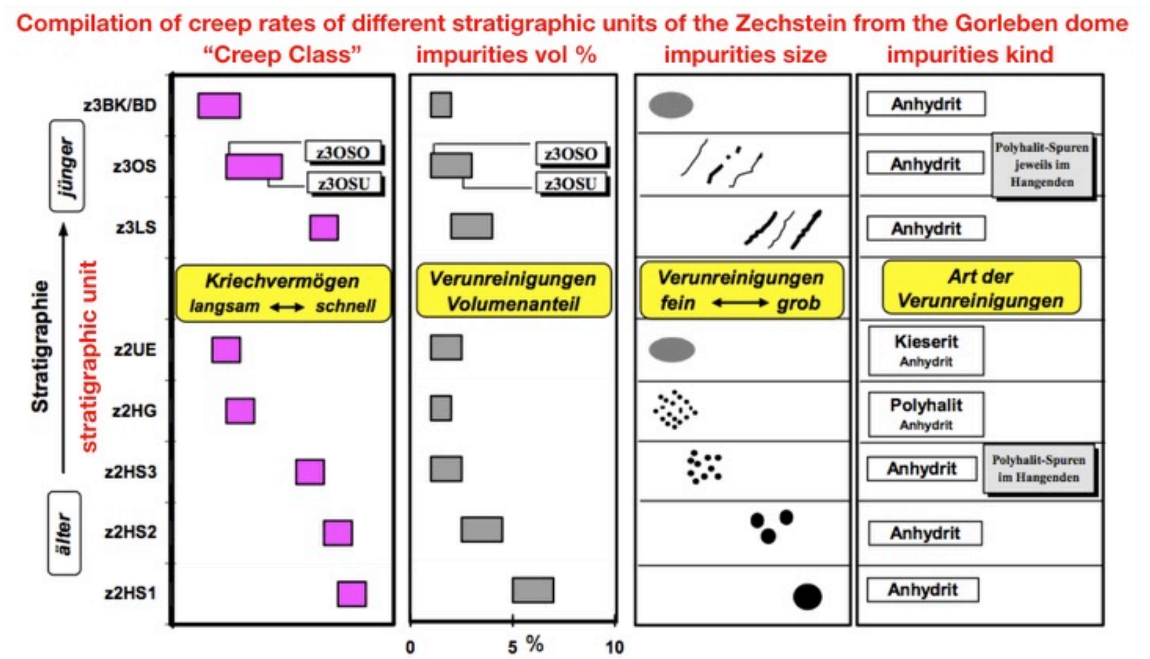



**Abb. 4.3.8: Korrelationen des Kriechvermögens und der Gefügeeigenschaften von Steinsalz (Salzstock Gorleben).**

*Figure 17 An attempt to classify creep properties of rock salt samples from different stratigraphic units based on composition and impurities. In this compilation a series of "creep classes" with different parameters in the creep law are used for each unit. Note that key microstructural parameters like grain size, solid solution impurities, grain boundary structure wer not measured in this study. After: Hunsche, U., Schulze, O., Walter, F., Plischke, I.: Projekt Gorleben – Thermomechanisches*

*Verhalten von Salzgestein. Abschlussbericht. Arbeitspaket 9G 213 811, Bundesanstalt für Geowissenschaften und Rohstoffe (BGR): Hannover, 2003.*

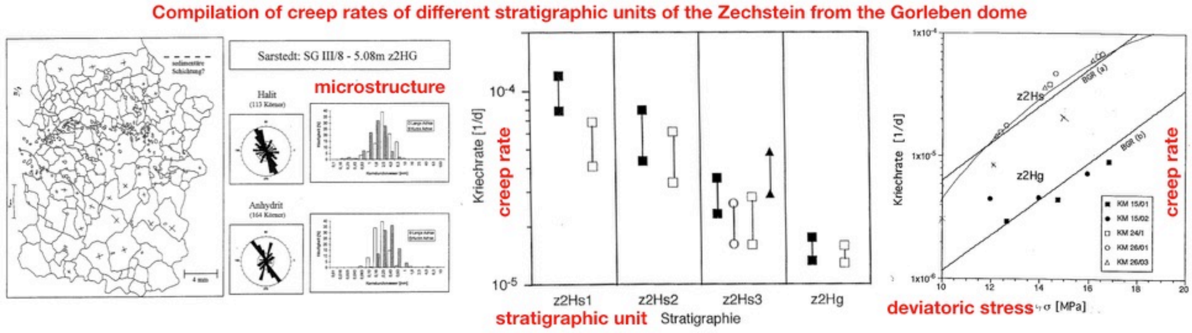



*Figure 18 An interesting attempt to correlate creep properties of rock salt samples from different stratigraphic units based on Microstructure (grain size and inclusions). After: Popp, T., 1994. Gefügekundliche Untersuchungen an Salzgestein der Staßfurt-Folge aus dem Salzstock Sarstedt. Research Report No. 112624. BGR, Hannover, Germany.*

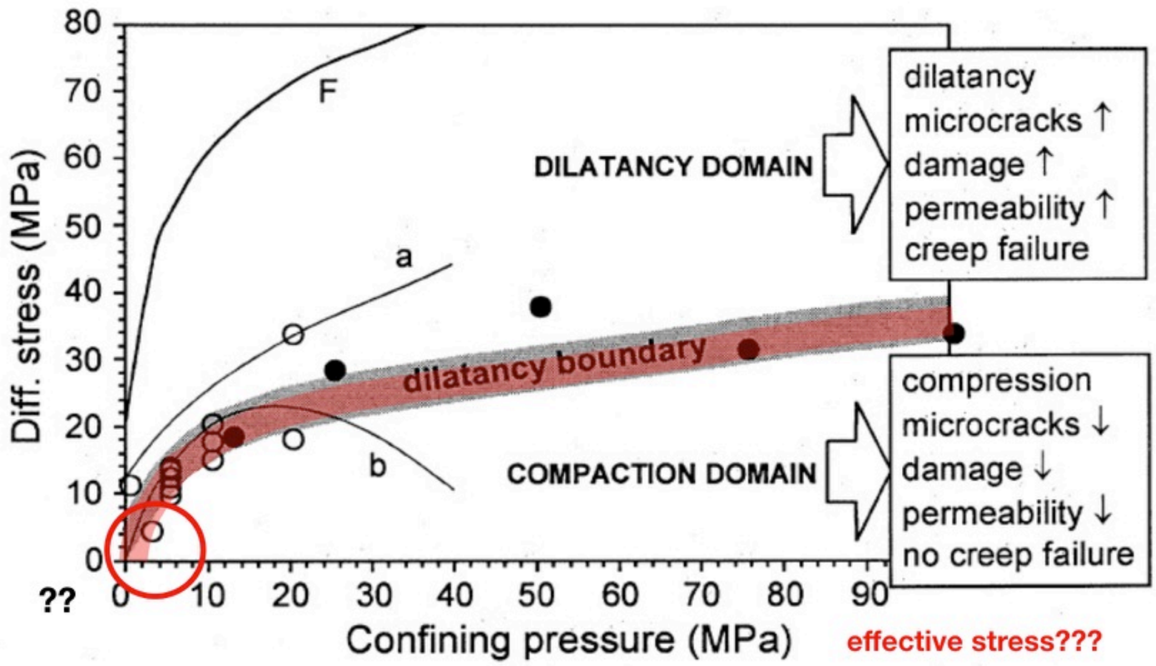



*Figure 19 The classic study on the dilatancy boundary in rock salt. Non-dilatant deformation is often used as a criterion for integrity in salt engineering. After: Popp, T., Kern, H., 2000. Monitoring the state of micro fracturing in rock salt during deformation by combined measurements of permeability and P- and S- wave velocities. Physics and Chemistry of the Earth, Part A: Solid Earth and Geodesy 25, 149–154.*

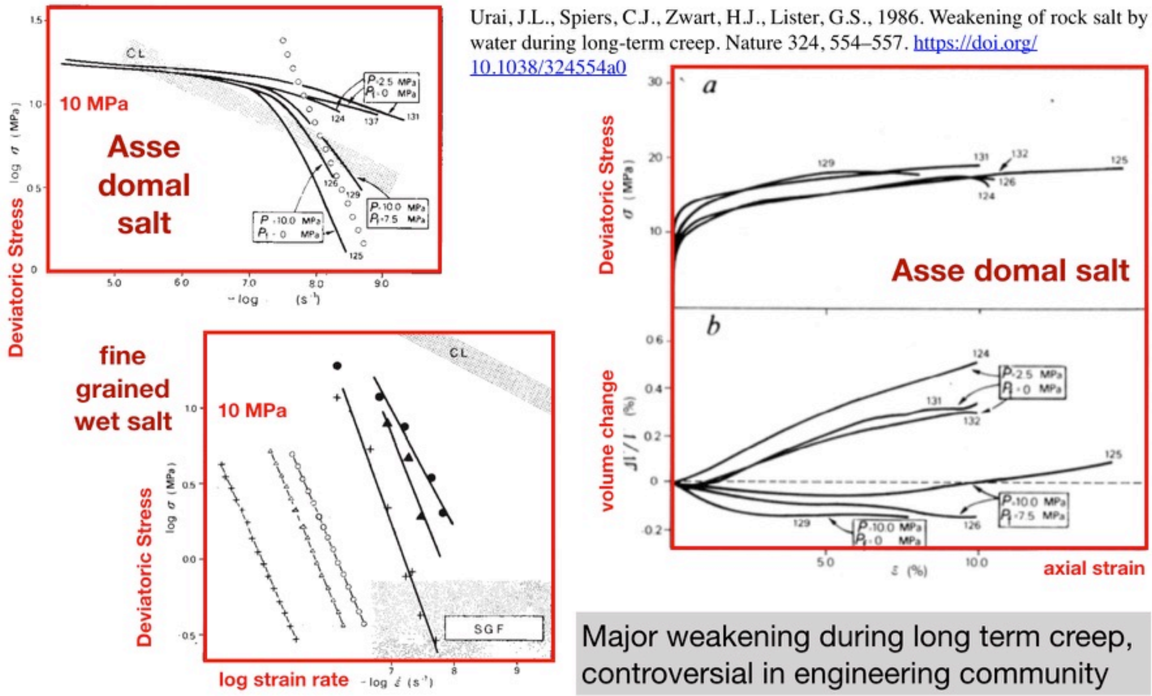



*Figure 20. A series of deformation experiments on Asse domal salt with very low water content present as small unconnected brine inclusions at grain boundaries has shown, that in the non-dilatant field, and after rapid deformation to about 10% strain, at lowering the deviatoric stress to values below 10 MPa these samples deformed many orders of magnitude faster than predicted for dislocation creep. Samples initially deformed in the dilatant field did not show this effect. This dramatic weakening was explained by the formation of thin brine films along the grain boundaries which dramatically increased their mobility, leading to recrystallization and grain refinement by grain boundary migration and the activation of pressure solution creep. The same effect was seen in wet, synthetic, fine grained rock salt samples which deform much faster than predicted by dislocation creep, and this deformation is well explained by pressure solution. This effect is expected to be prominent in the rock salt around caverns and play an important role during abandonment by making the rock salt around the cavern creep much faster than predicted. Urai, J.L., Spiers, C.J., Zwart, H.J., Lister, G.S., 1986. Weakening of rock salt by water during long-term creep. Nature 324, 554–557. - Urai, J.L., Spiers, C.J., 2007. The Effect of Grain Boundary Water on Deformation Mechanisms and Rheology of Rocksalt During Long-Term Deformation. The Mechanical Behavior of Salt – Understanding of THMC Processes in Salt. Hannover, Germany, 149–158.*

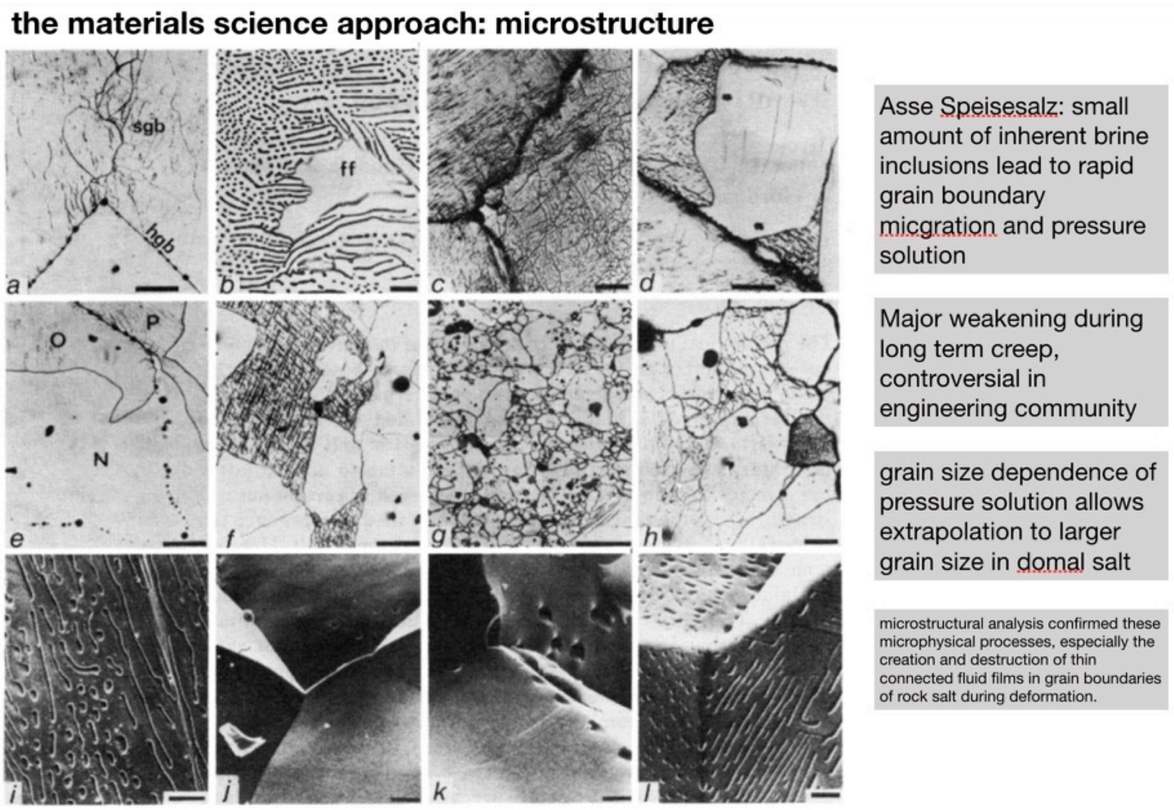



***Figure 21 A series of deformation experiments on Asse domal salt with very low water content present as small unconnected brine inclusions at grain boundaries has shown, that in the non-dilatant field, and after rapid deformation to about 10% strain, at lowering the deviatoric stress to values below 10 MPa, these samples deformed many orders of magnitude faster than predicted for dislocation creep. Samples initially deformed in the dilatant field did not show this effect. This dramatic weakening was explained by the formation of thin brine films along the grain boundaries which dramatically increased their mobility, leading to recrystallization and grain refinement by grain boundary migration and the activation of pressure solution creep. The same effect was seen in wet, synthetic, fine grained rock salt samples which deform much faster than predicted by dislocation creep, and this deformation is well explained by pressure solution. From: Urai, J.L., Spiers, C.J., Zwart, H.J., Lister, G.S., 1986. Weakening of rock salt by water during long-term creep. Nature 324, 554–557. - Urai, J.L., Spiers, C.J., 2007. The Effect of Grain Boundary Water on Deformation Mechanisms and Rheology of Rocksalt During Long-Term Deformation. The Mechanical Behavior of Salt – Understanding of THMC Processes in Salt. Hannover, Germany, 149–158.***

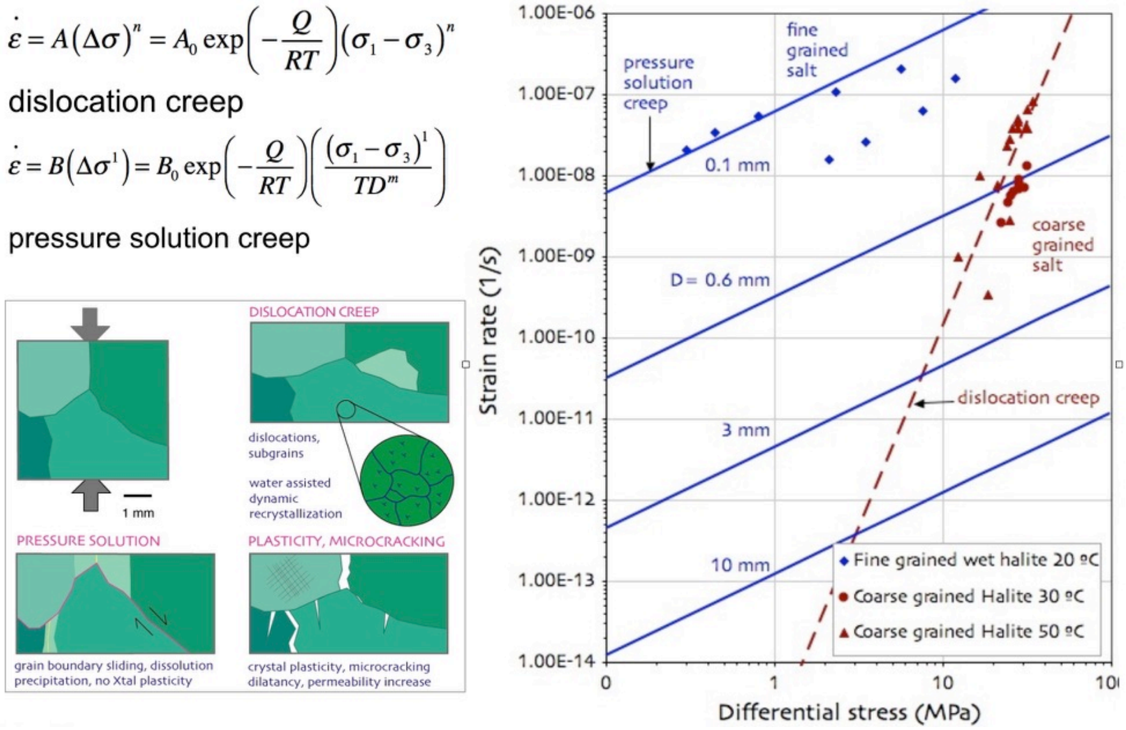



***Figure 22 This is a summary of the main microphysical deformation mechanisms in rock salt. They can all operate simultaneously depending on micro fabric and physical conditions, and they are associated with very different mechanical and transport properties. After: Urai, J.L., Schleder, Z., Spiers, C.J., Kukla, P.A., 2008. Flow and Transport Properties of Salt Rocks. Dynamics of Complex Intracontinental Basins: The Central European Basin System. Springer, 277–290.***

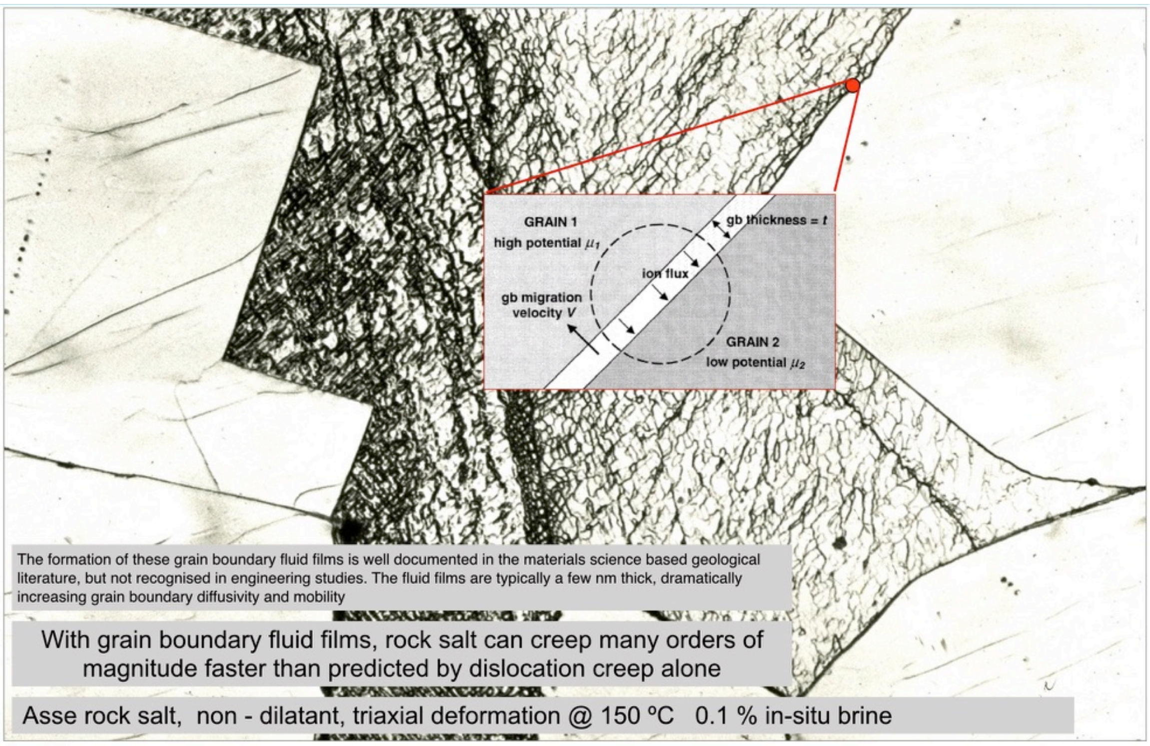



***Figure 23 Fluid filled grain boundaries in rock salt have a dramatically increased mobility and diffusivity, causing major changes in creep and permeation. These microphysical processes must be taken into account for reliable predictions of deformation around salt caverns over long term. After: Urai, J.L., Spiers, C.J., Zwart, H.J., Lister, G.S., 1986. Weakening of rock salt by water during long-term creep. Nature 324, 554–557.***

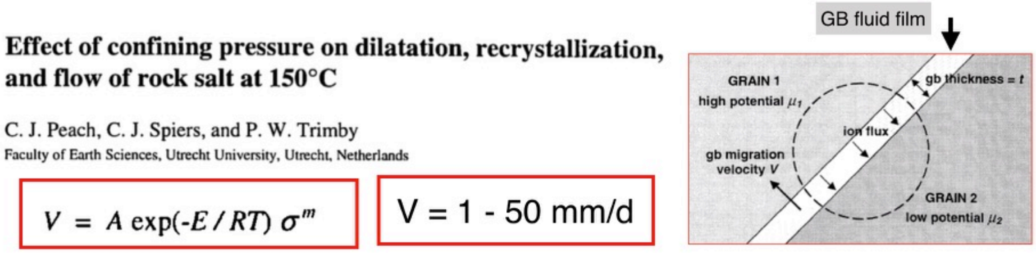



***Figure 24 Fluid filled grain boundary migration. Quantitative models of the properties and mobility of grain boundary fluid films have been published by a number of authors. In this study, calculations of grain boundary velocities show how rapid this process can be, in agreement with microscopic observations. After: Peach, C.J., Spiers, C.J., Trimby, P.W., 2001. Effect of confining pressure on dilatation, recrystallization, and flow of rock salt at 150°C. Journal of Geophysical Research: Solid Earth 106, 13315–13328***

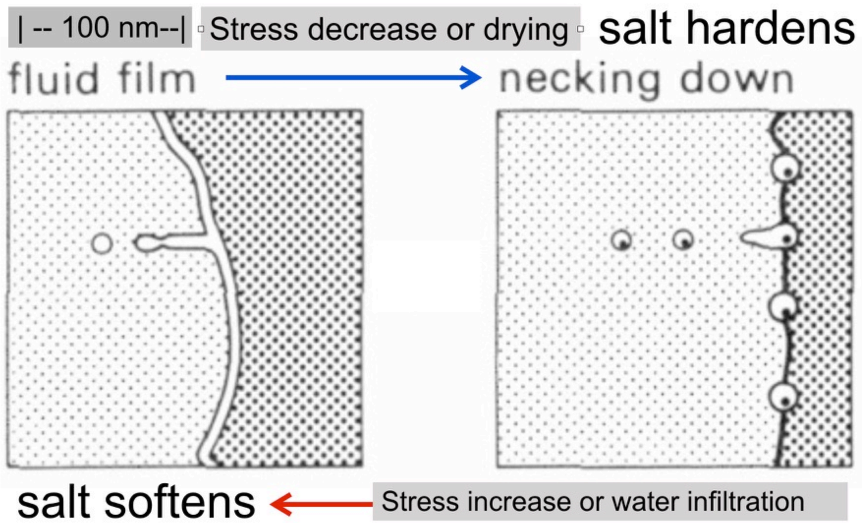
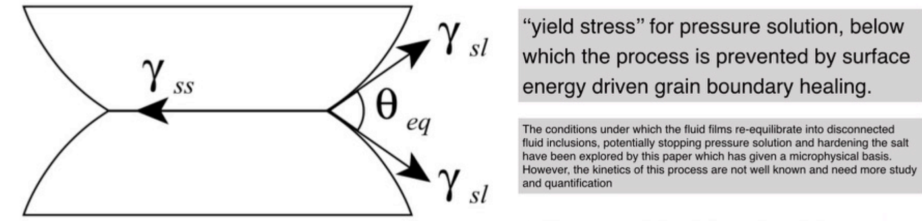



***Figure 25 Fluid filled grain boundary transitions. One interesting and important property of grain boundary fluid films in rock salt is that they are only stable when the deviatoric stress is sufficiently high. If the rock salt is recrystallized and deviatoric stress decreases, the films re-equilibrate into disconnected fluid inclusions, potentially stopping pressure solution and hardening the salt. This process is reversible. After: Drury, M.R., Urai, J.L., 1990. Deformation-related recrystallization processes. Tectonophysics 172, 235–253.***




$$(\sigma_n - P_f) < \sigma_{crit} = 2\alpha\sqrt{E\frac{\gamma_{sl}}{\delta}\left(\cos\frac{\theta}{2} - \cos\frac{\theta_{eq}}{2}\right)}$$

***Figure 26 The conditions under which the fluid films re-equilibrate into disconnected fluid inclusions, potentially stopping pressure solution and hardening the salt have been explored by this paper which has given a microphysical basis for the process. However, the kinetics of this process are not well known and need more study and quantification. After: van Noort, R., H. J. M. Visser, and C. J. Spiers (2008), Influence of grain boundary structure on dissolution-controlled pressure solution and retarding effects of grain boundary healing, J. Geophys. Res., 113, B03201***

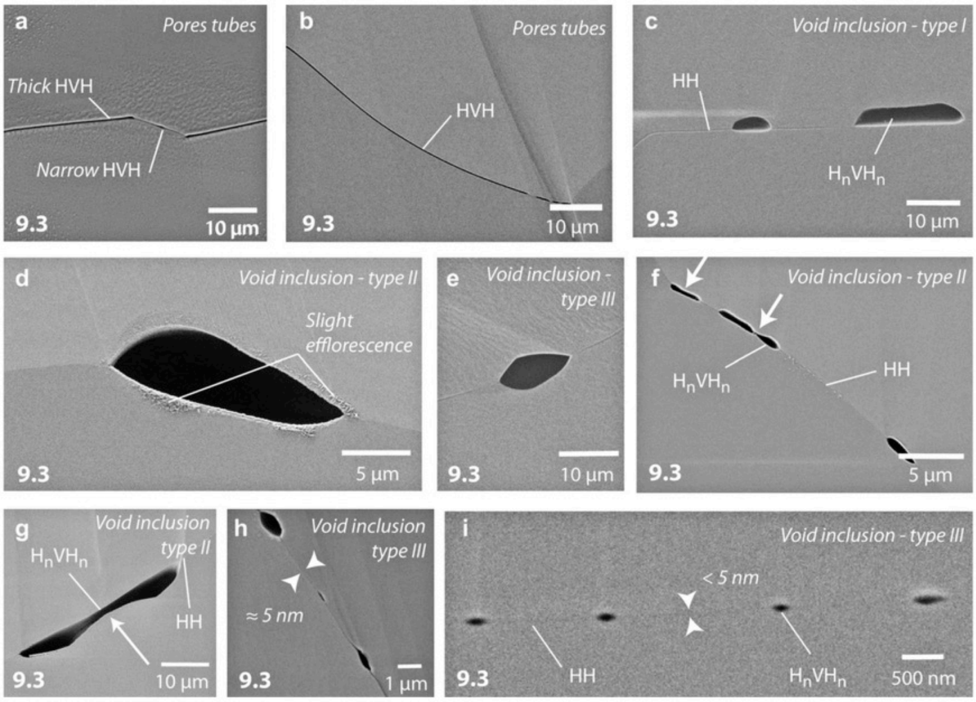



***Figure 27 Examples of re-equilibrated grain boundaries with disconnected brine inclusions, preventing pressure solution and hardening the salt. These images were prepared by Broad ion Beam polishing and Scanning electron microscopy. After: Desbois, G., Urai, J.L., de Bresser, J.H.P., 2012. Fluid distribution in grain boundaries of natural fine-grained rock salt deformed at low differential stress (Qom Kuh Salt Fountain, Central Iran): Implications for rheology and transport properties. Journal of Structural Geology 43, 128–143.***

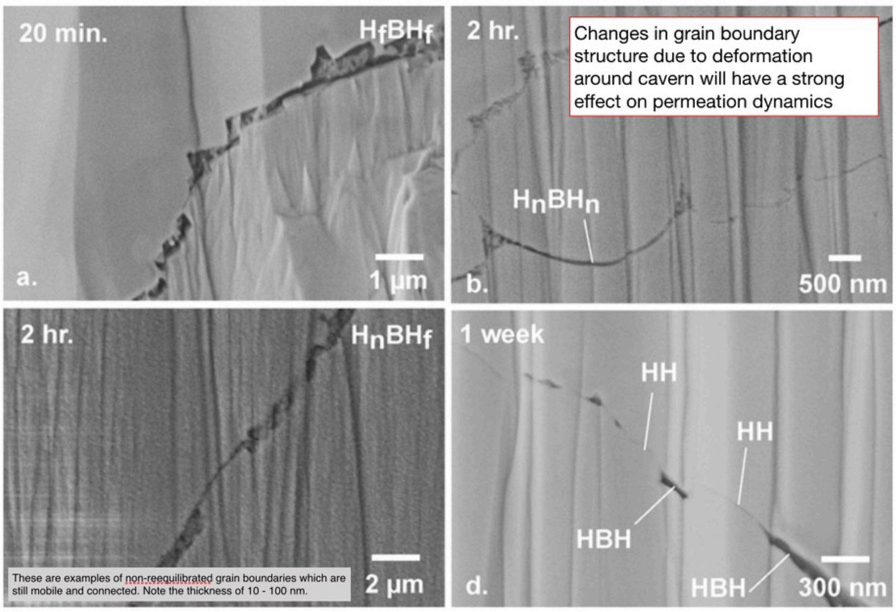



***Figure 28 Cryo-BIB-SEM images of brine-filled grain boundaries in rock salt in various stages of reequilibration. Desbois, G., Urai, J.L., Kukla, P.A., Wollenberg, U., Pérez-Willard, F., Radí, Z., Riholm, S., 2012. Distribution of brine in grain boundaries during static recrystallization in wet, synthetic Halite: Insight from broad ion beam sectioning and SEM observation at cryogenic temperature. Contributions to Mineralogy and Petrology 163, 19–31.***

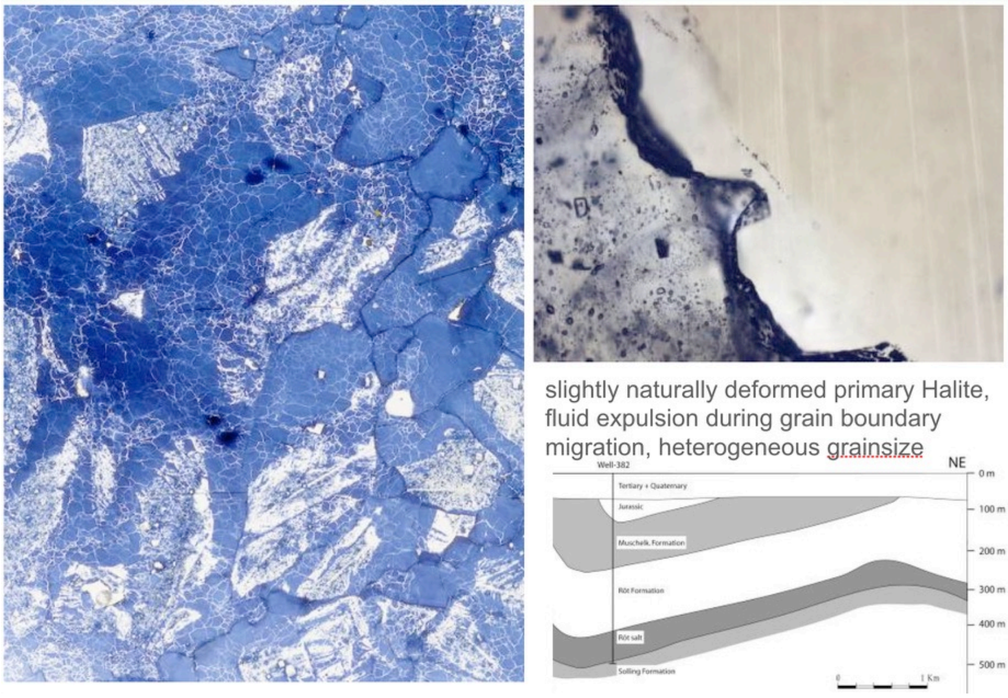



***Figure 29 Primary and early diagenetic fabric of Hengelo rock salt is overprinted by tectonic strain forming subgrains which indicate low deviatoric stress. This fabric is in turn partly overgrown by migrating grain boundaries which remove the primary fluid inclusions from the crystals. Similar processes are predicted to have occurred in many engineering tests on rock salt, but were not detected because no microstructural analysis was carried out. After: Schleder, Z., Urai, J.L., 2005. Microstructural evolution of deformation- modified primary Halite from Hengelo, the Netherlands. International Journal of Earth Sciences 94, 941–956.***

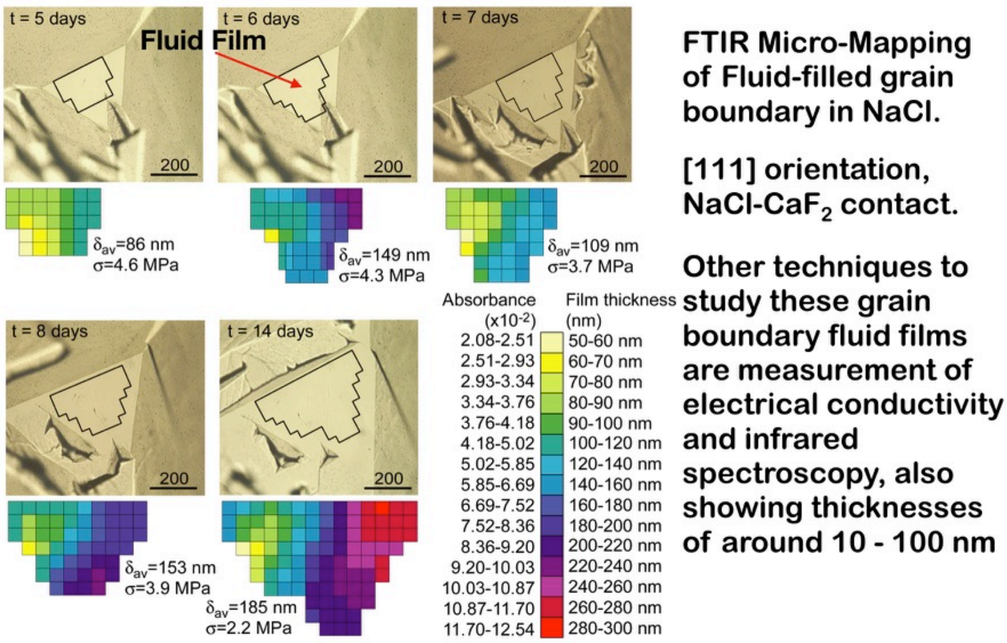



***Figure 30 - Fourier Transform Infrared Imaging of active- fluid-filled grain boundary in NaCl undergoing pressure solution -- After: de Meer, S., Spiers, C.J., Peach, C.J., Watanabe, T., 2002. Diffusive properties of fluid-filled grain boundaries measured electrically during active pressure solution. Earth and Planetary Science Letters 200, 147–157. de Meer, S., Spiers, C.J., Nakashima, S., 2005. Structure and diffusive properties of fluid-filled grain boundaries: An in-situ study using infrared (micro) spectroscopy. Earth and Planetary Science Letters 232, 403–414.***

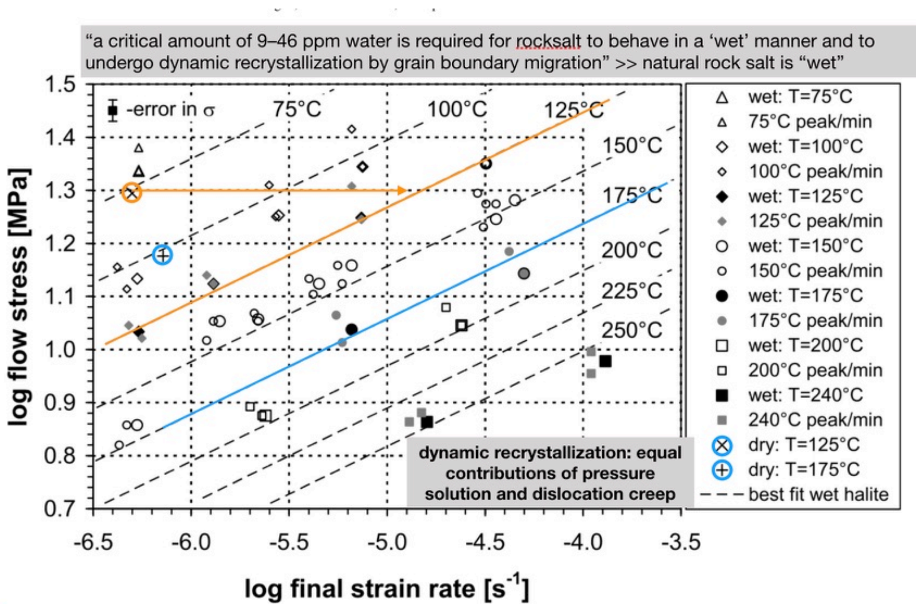



*Figure 31 Dedicated studies of the effect of small amounts of water in rock salt (> 10 ppm, which is always present in nature, so that rock salt in-situ is always "wet") have shown a clear difference with artificially dried samples which creep about three orders of magnitude slower than wet samples under the same conditions. There is extensive dynamic recrystallisation as shown by detailed microstructural analysis and the samples deform by equal contributions of dislocation and pressure solution creep. After: Ter Heege, J.H., de Bresser, J.H.P., Spiers, C.J., 2005. Rheological behaviour of synthetic rocksalt: the interplay between water, dynamic recrystallization and deformation mechanisms. Journal of Structural Geology 27, 948–963*

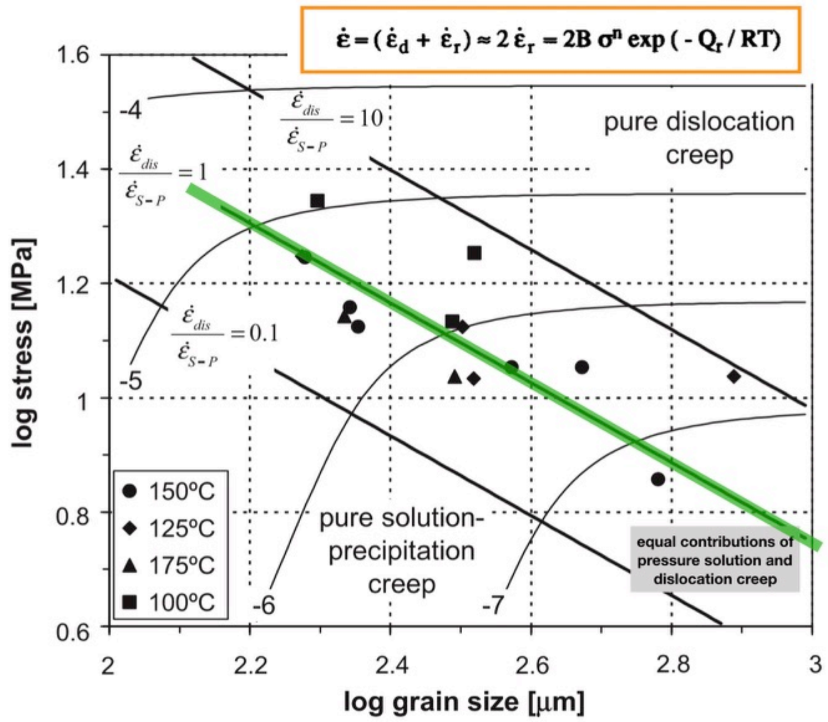



*Figure 32 During extensive dynamic recrystallisation the samples deform by equal contributions of dislocation and pressure solution creep. This is the basis of the well-known microphysical field boundary model. The dynamically recrystallized grainsize is a function of deviatoric stress: the lower the deviatoric stress, the higher the dynamically recrystallized grainsize. After: de Bresser, J.H.P., Ter Heege, J.H., Spiers, C.J., 2001. Grain size reduction by dynamic recrystallization: can it result in major rheological weakening? International Journal of Earth Sciences 90, 28–45.*

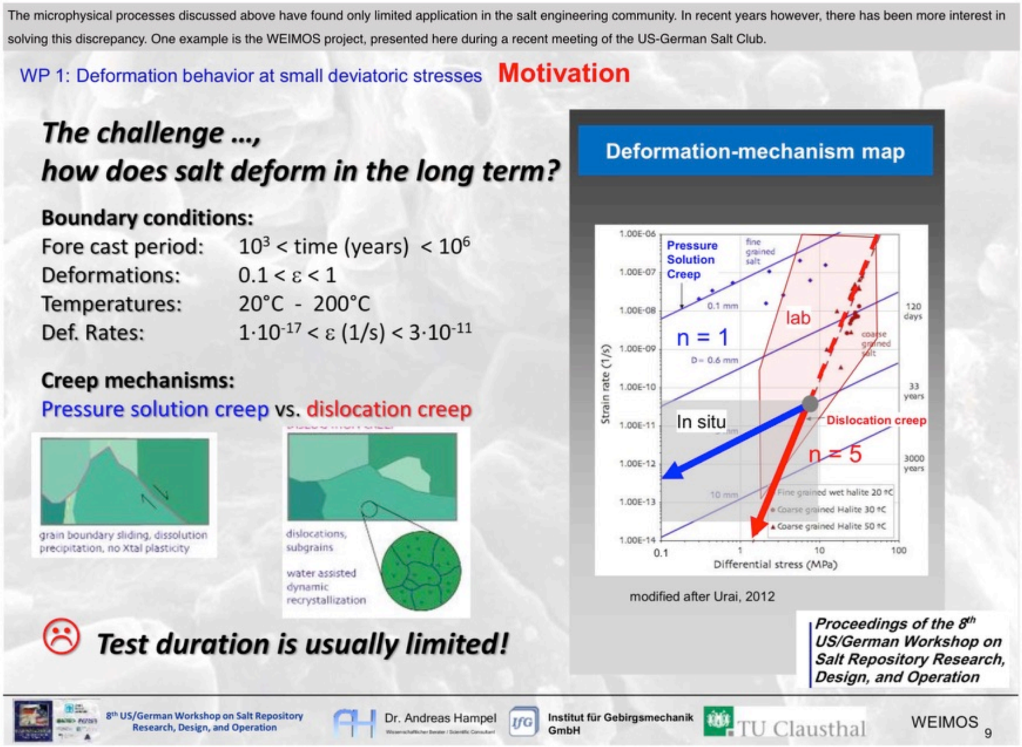



***Figure 33 Recent introduction WEIMOS project – showing that fluid assisted grain boundary processes are now starting to be taken seriously in salt engineering. After: Current Status of Research in the Joint Project WEIMOS Andreas Hampel (Consultant), Till Popp (IfG), Kai Herchen (TUC) - 8th US/German Workshop on Salt Repository Research, Design, and Operation- Middelburg, The Netherlands September 5-7, 2017***

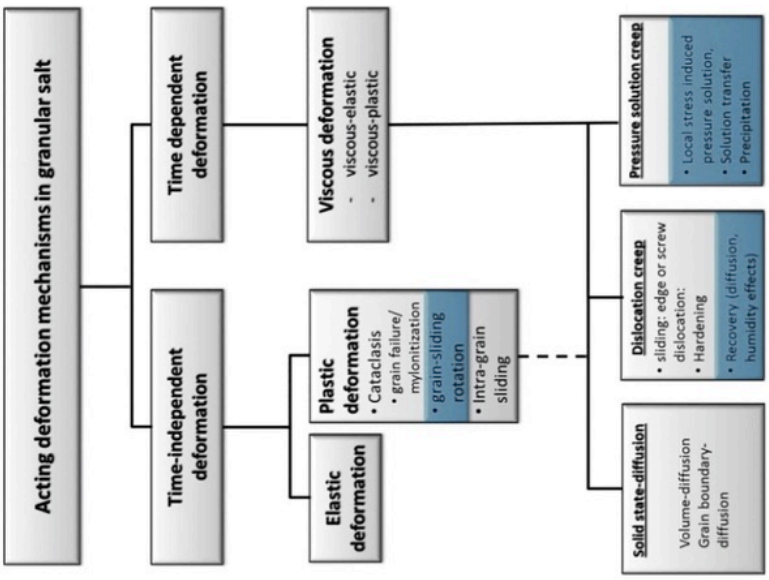



***Figure 34 Overview of deformation mechanisms in rock sal, including pressure solution. -- After: Hansen, F.D., Popp, T., Wieczorek, K., Stührenberg, D., 2015. Salt reconsolidation applied to repository seals. In: Roberts, L., Mellegard, K., Hansen, F. (Eds.), The Mechanical Behaviour of Salt VIII, Rapid City, South Dakota. Taylor & Francis Group, London, pp. 179–189.***

Spiers et al., (1996- 2007)

$$\sigma_e = \sigma - P_f$$

**Dissolution Control:**

$$\dot{\varepsilon}_s = I_s \times \frac{\sigma_e}{d} \times f_s(\phi)$$

**Diffusion Control:**

$$\dot{\varepsilon}_d = [DCS] \times \frac{\sigma_e}{d^3} \times f_d(\phi)$$

**Precipitation Control:**

$$\dot{\varepsilon}_p = I_p \times \frac{\sigma_e}{d} \times f_p(\phi)$$

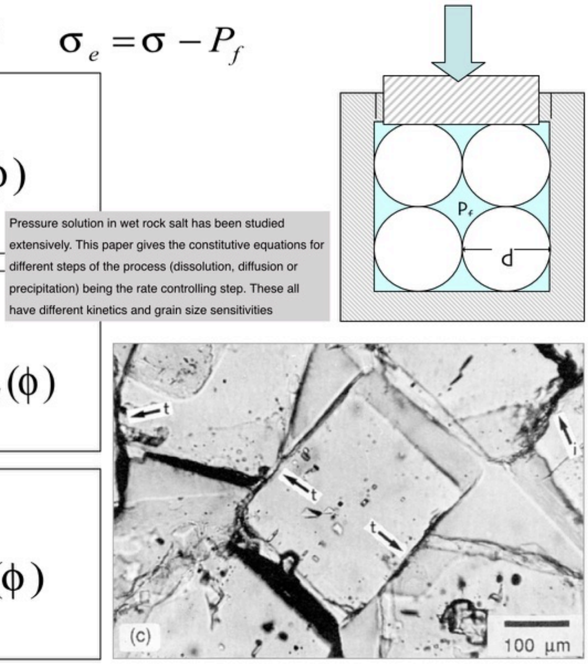



*Figure 35 Pressure solution in wet rock salt has been studied extensively. This paper gives the constitutive equations for different steps of the process (dissolution, diffusion or precipitation) being the rate controlling step. These all have different kinetics and grain size sensitivities but are all linearly dependent on deviatoric stress. After:. Spiers, C.J., Schutjens, P.M.T.M., Brzesowsky, R.H., Peach, C.J., Liezenberg, J.L., Zwart, H.J., 1990. Experimental determination of constitutive parameters governing creep of rocksalt by pressure solution. In: Knipe, R.J., Rutter, E.H. (Eds.), Deformation Mechanisms, Rheology and Tectonics, Geological Society Special Publications. The Geological Society, London, United Kingdom, 215–227*

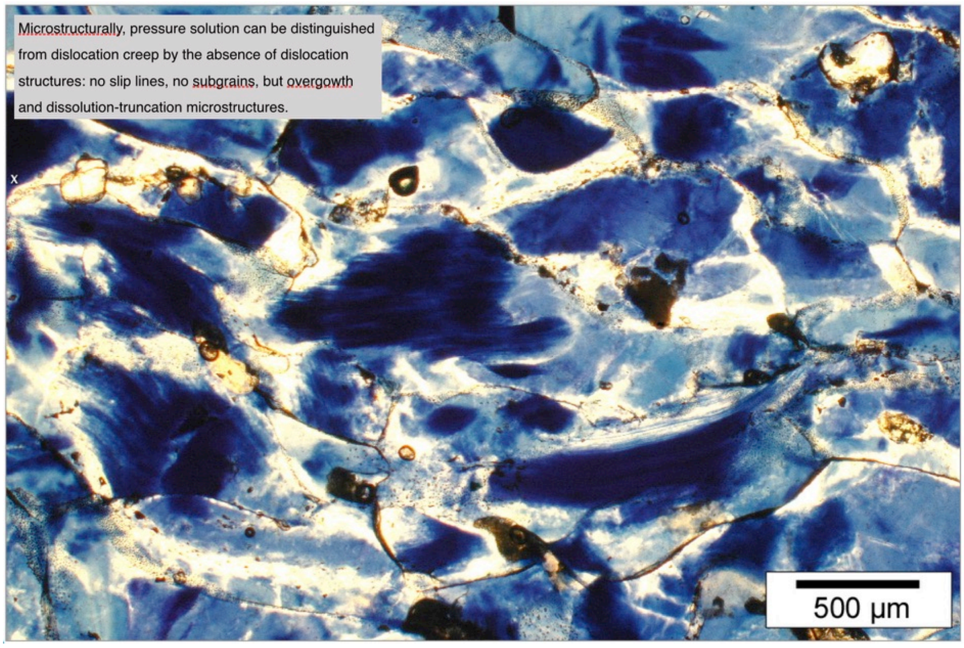



*Figure 36. Microstructurally, pressure solution can be distinguished from dislocation creep by the absence of dislocation structures: no slip lines, no subgrains, but overgowth and dissolution-truncation microstructures. After: Schleder, Z., Urai, J.L., 2007. Deformation and recrystallization mechanisms in Mylonitic shear zones in naturally deformed extrusive Eocene-Oligocene rocksalt from Eyvanekey Plateau and Garmsar Hills (Central Iran). Journal of Structural Geology 29, 241–255.*

Muhammad, N., 2014. Deformation and transport processes in salt rocks:, PhD Thesis, Utrecht University.

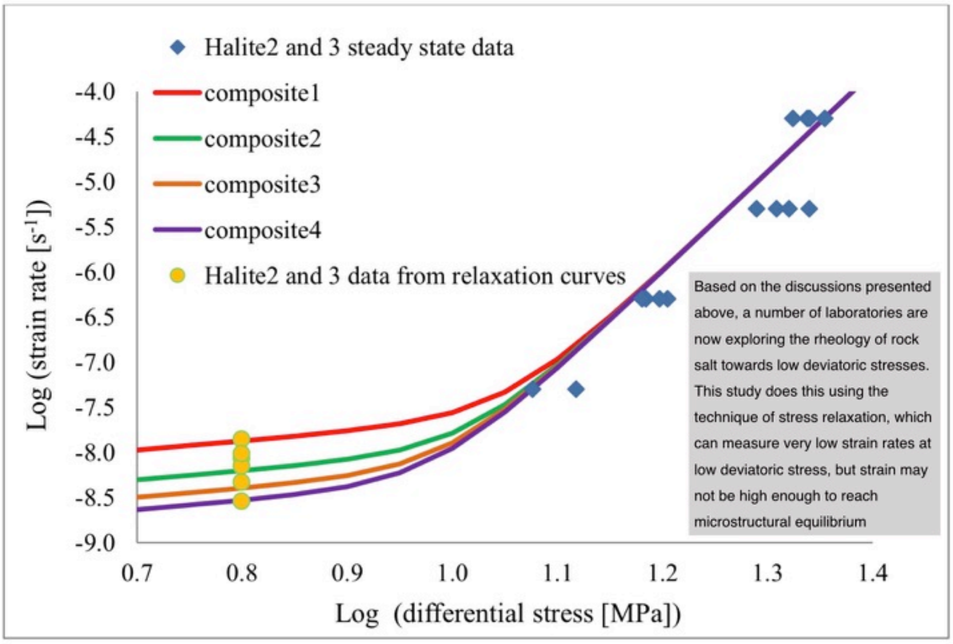



***Figure 37 Based on the discussions presented above, a number of laboratories are now exploring the rheology of rock salt towards low deviatoric stresses. This study does this using the technique of stress relaxation, which can measure very low strain rates at low deviatoric stress, but strain may not be high enough to reach microstructural equilibrium. After: Muhammad, N., 2014. Deformation and transport processes in salt rocks:, PhD Thesis, Utrecht University.***

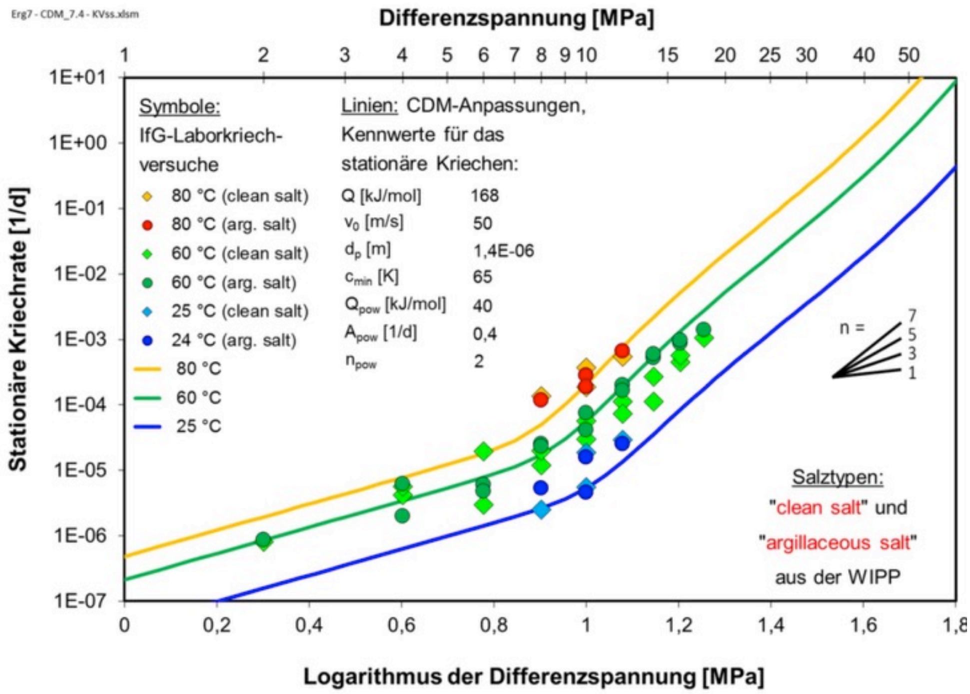



***Figure 38 In the German Verbundproject, a change in stress exponent towards low deviatoric stress is also implemented, unfortunately without microstructural analysis of the samples so it is difficult to compare the results with microphysical parameters and experiments with pressure solution. After: Hampel (2016): Verbundprojekt - Vergleich aktueller Stoffgesetze und Vorgehensweisen anhand von Modellberechnungen zum thermo-mechanischen Verhalten und zur Verheilung von Steinsalz– Teilvorhaben1 – Dr. Andrea s Hampel Bundesministerium für Wirtschaft und Energie (BMWi) Projektträger (PTKA-WTE) (KIT)***

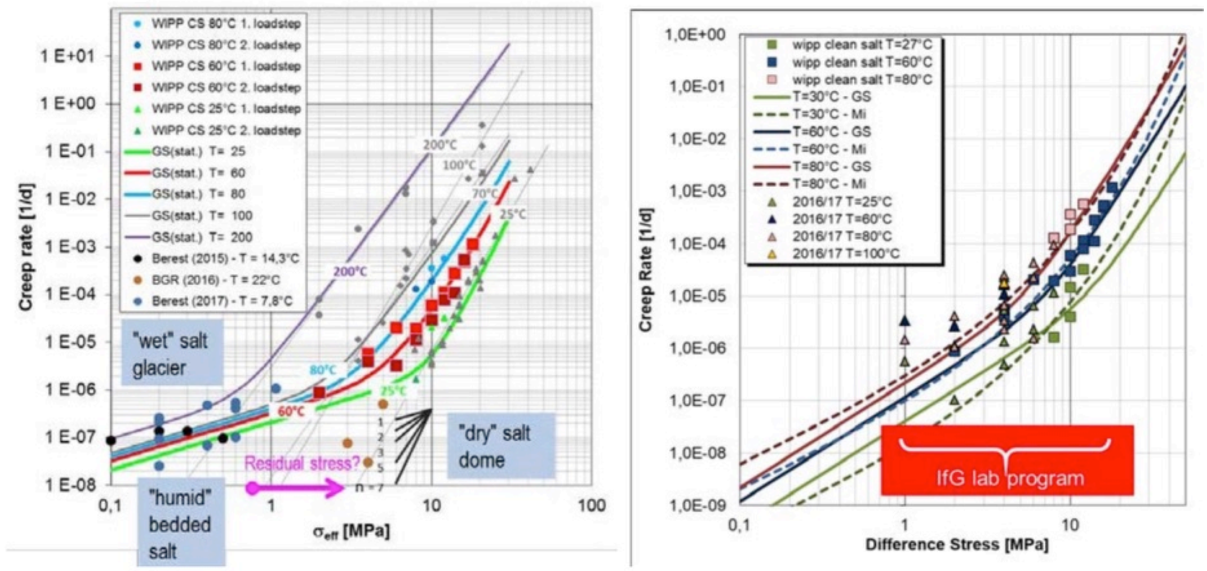



*Figure 39: "Owing to intense interest shown by the salt repository community in creep of salt at low stresses, the US/German workshop leadership extended an invitation to Professor Pierre Bérest, who along with a group of colleagues from the Solution Mining Research Institute, has executed some clever low-deviatoric creep tests with control of temperature and stress to tight specifications (SMRI Research Report RR2017-1). The data base obtained from in situ observations and laboratory tests on salt indicates that both dislocation (e.g., Carter & Hansen, 1983) and diffusional creep mechanisms (Urai & Spiers, 2007; Spiers et al., 1990) can be important in salt under long-term conditions." After: Bérest, 2018: CREEP AT LOW DEVIATORIC STRESS. In: Proceedings of the 8th US/German Workshop on Salt Repository Research, Design, and Operation - Spent Fuel and Waste Disposition - Prepared for US Department of Energy - Spent Fuel and Waste Science and Technology - Hansen, Steininger, Bollingerfehr, Kuhlman, Dunagan, - February 7, 2018 - SFWD-SFWST-2018-000485*

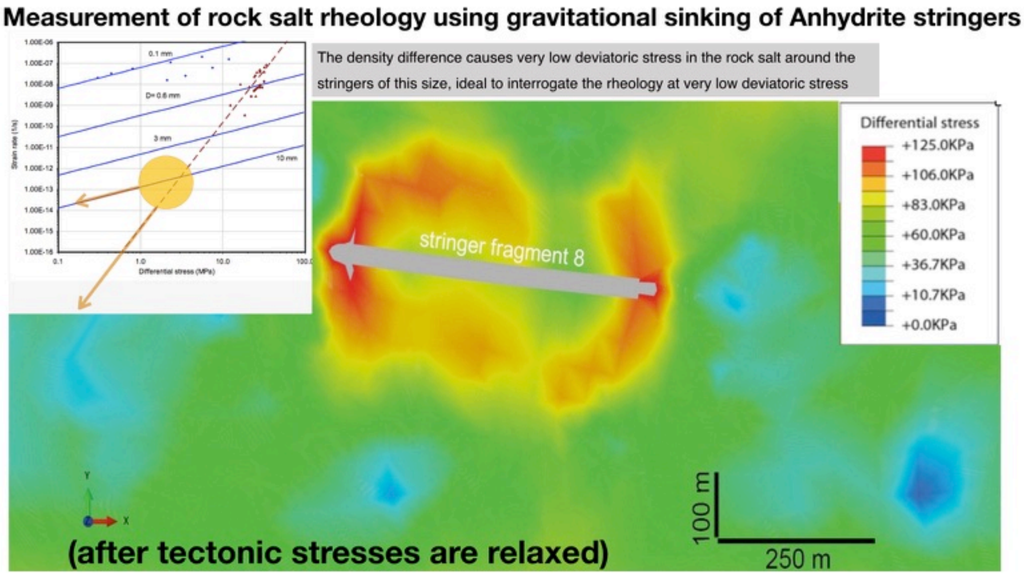



*Figure 40 Measurement of the rheology of rock salt at very low deviatoric stresses and strain rates. Finite element model of the deviatoric stress around an anhydrite inclusion in rock salt, caused by the density difference. Note that these deviatoric stresses are very small, allowing to interrogate the in-situ deformation of rock salt at very low deviatoric stress by measuring the gravitational sinking rate. Li, S., Abe, S., Urai, J.L., Strozyk, F., Kukla, P.A., van Gent, H.W., 2012. A method to evaluate long-term rheology of Zechstein salt in the Tertiary. SaltMech7 - The Mechanical Behaviour of Salt VII. Taylor & Francis Group, Paris, France, 215–220.*

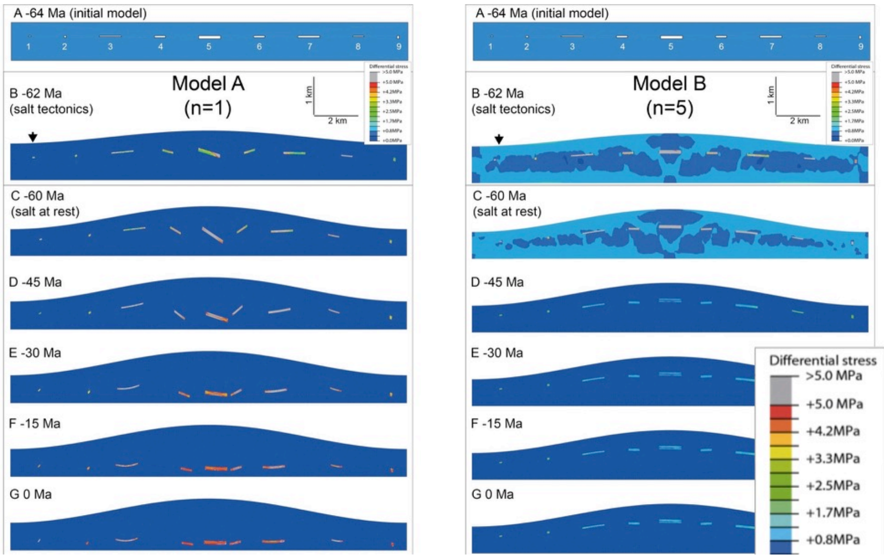



***Figure 41. results of two series of numerical models with the two different rheologies (dislocation creep vs pressure solution) has shown, that in this case, pressure solution cannot have been active because the stringers would all have sunk to the bottom of the Zechstein by now. At present, the long-term rheology of Zechstein rock salt seems to be dominated by dislocation creep. It is an interesting and as yet unexplained microstructural observation that despite the high rate of fluid-assisted grain boundary migration observed in experiments, most naturally deformed rock salt is not completely recrystallized and preserves subgrains. An explanation for this is that below some critical difference in driving force for cross-boundary solution-precipitation transfer, surface energy driving forces cause necking or healing of grain boundary fluid films to form isolated fluid inclusion), thus rendering the boundaries immobile and switching off pressure solution. After: Li, S., Abe, S., Urai, J.L., Strozyk, F., Kukla, P.A., van Gent, H.W., 2012. A method to evaluate long-term rheology of Zechstein salt in the Tertiary. SaltMech7 - The Mechanical Behaviour of Salt VII. Taylor & Francis Group, Paris, France, 215–220.***

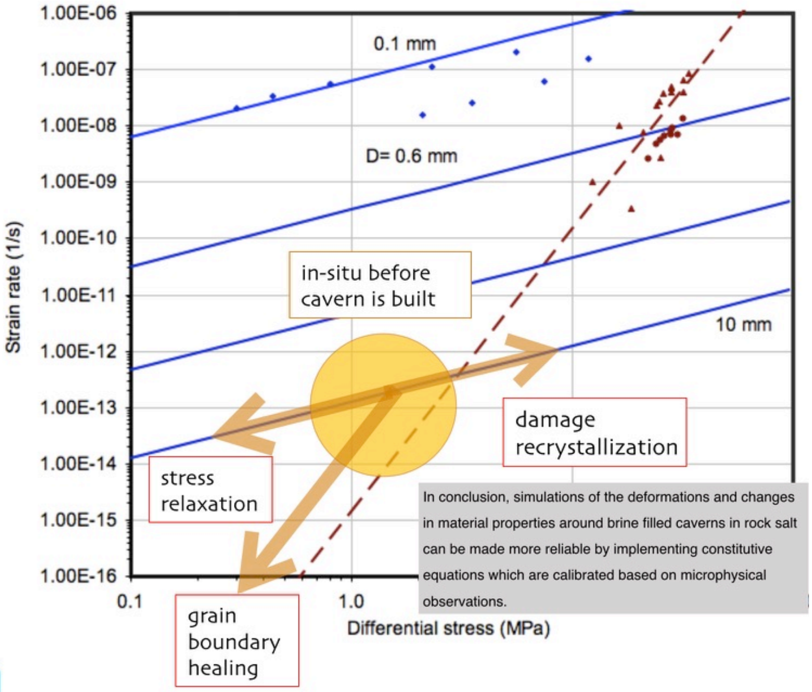



***Figure 42 Based on integration of microphysics and rock mechanics in rock salt geoscience, we now have a reasonably complete set of microphysically based constitutive models which describe the complex rheology of rock salt at low deviatoric stress and low strain rate. Here we propose that integrating this understanding in the salt engineering design will significantly increase the reliability of predictions of the evolution of abandoned caverns.***

Table 1. Microstructure descriptors

| Descriptors | Definition |
|---|---|
| Grain orientation | Grains are represented by virtual ellipses with same second moment. The orientation is the angle between the major axis of the ellipse and the horizontal axis of the image. |
| Branch orientation | The angle between a branch and the horizontal axis of the image |
| Grain area | The area of a grain |
| Branch length | The length of segments linking the centroids of two grains in contact |
| Roundness | The ratio of the area of a particle over the area of a circle whose diameter equals to the length of the virtual ellipse's major axis |
| Elongation | The ratio between the minor and major axes of the virtual ellipses representing the grains |
| Local solid | Solid volume fraction over a |

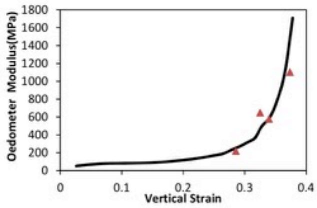



Fig. 4. Calibration of the elastic properties (solid line: experimental results; red dots: calibrated model)

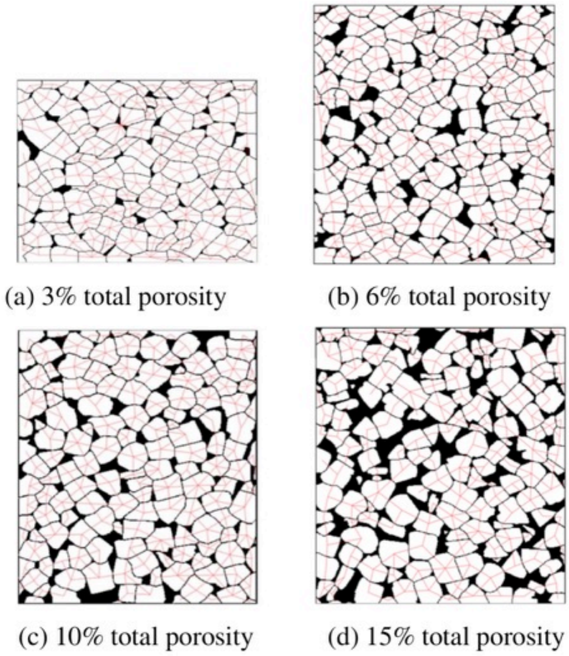



Fig. 2. Microstructure images of salt samples (white area: salt grains, black area: voids, red lines: branches linking the centers of two grains in contact)

***Figure 43 The upscaling of grain scale microphysics to constitutive laws for rock mechanics applications in materials science is usually done using microstructure-based numerical models. This paper computes the elastic properties of rock salt samples from microstructure. After: Xianda, S., Arson, C., Ding, J., Chester, F.M., Chester, J.S., 2017. Experimental characterization of microstructure development for calculating fabric and stiffness tensors in salt rock. Presented at the 51st US Rock Mechanics / Geomechanics Symposium, 25–28 June 2017.***

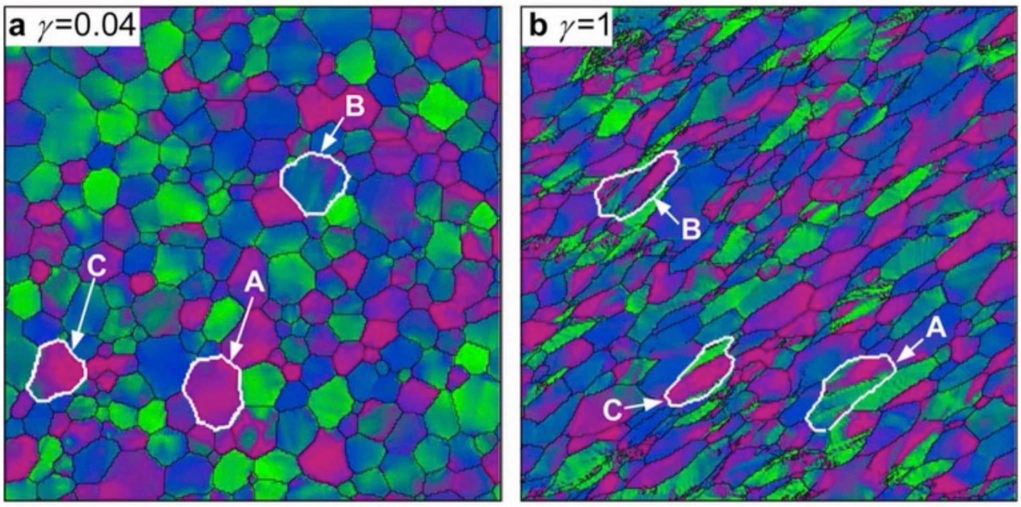



***Figure 44. The upscaling of grain scale microphysics to constitutive laws for rock mechanics applications is usually done using microstructure-based numerical models. This paper computes the crystal-scale distortions, changes in crystal orientations and rotation recrystallization using ELLE, with full flield crystal plasticity simulations of a rock salt polycrystals. After: Gomez-Rivas, E., Griera, A., Llorens, M.-G., Bons, P. D., Lebensohn, R. A., & Piazolo, S. (2017). Subgrain rotation recrystallization during shearing: Insights from full-field numerical simulations of halite polycrystals. Journal of Geophysical Research: Solid Earth, 122, 8810–8827.***


*Figure 45 Anhydrite is strong and brittle based on short term rock mechanics experiments, although these ductile fold structures in salt show that over long term it must have been deforming with a viscous constitutive law. It is not clear how, especially in the case of caverns in bedded salt, Anhydrite rheology affects permeation, pressure build-up, and possible fracture propagation. From the Bernburg cavern.*

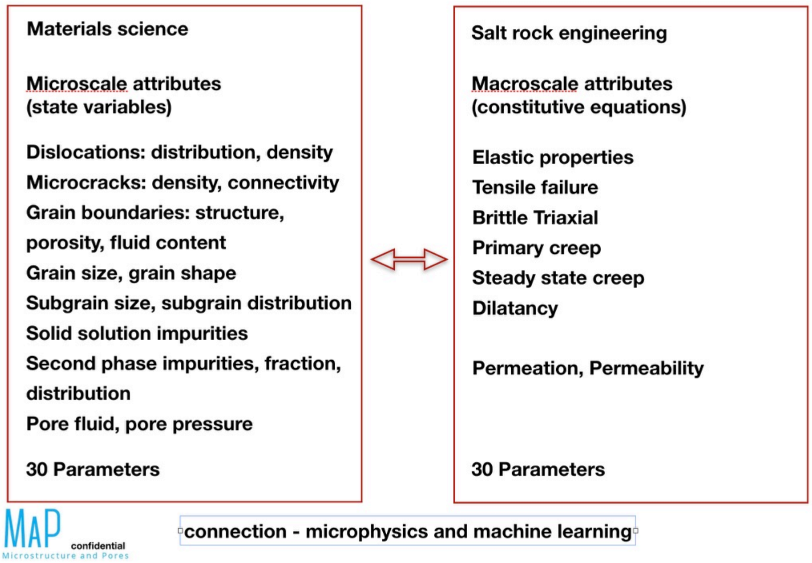



*Figure 46. Coupling of microscale and macroscale properties of rock salt. At present, mechanical properties of salt for cavern design are exclusively determined by laboratory measurement of stress-strain relations. The measurements are time consuming and therefore usually incomplete for prediction of long-term behavior. Over the past decades there were attempts to predict this using microstructure, which would allow property determination in the whole core. We have reviewed some of these. For dislocation creep, important parameters which affect creep properties are: solid solution impurities, second phase impurities, grain size, grain shape, subgrain size and density, dislocation density, crystallographic fabric and grain boundary mobility (the presence or absence of continuous fluid films on the grain boundaries), pore pressure (if present). For pressure solution creep, important parameters which affect creep properties are grain boundary mobility (the presence or absence of continuous brine films on the grain boundaries) and grain size, and to lesser extent solid solution impurities and pore pressure and porosity. Here we show how such a correlation could be achieved. Once the complete set of material variables is defined, these are measured for a sufficient number of samples, for which also the bulk properties are measured. Then based on microphysical models and upscaling using micromechanical models, first order correlations are explored. Finally, the dataset is analyzed using modern machine learning techniques which give sufficient training data, will allow predicting the bulk properties from microstructure.*

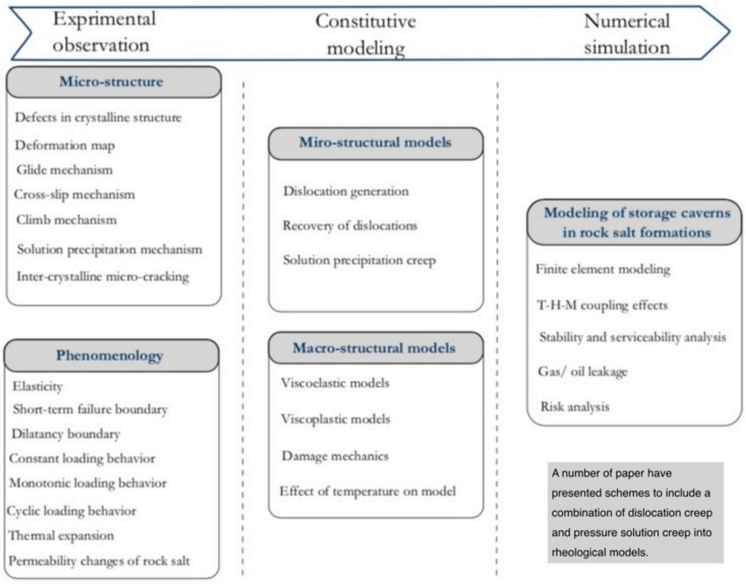



Figure 2.1: Three main steps to analyze the energy storage caverns

***Figure 47 – Proposed scheme to integrate constitutive modelling, microphysical understanding, and numerical modelling in the engineering prediction of salt caverns. After: Kavan Khaledi 2017 Constitutive Modeling of Rock Salt with Application to Energy Storage Caverns. Dissertation, Civil and Environmental Engineering, Ruhr-Universitaät Bochum***

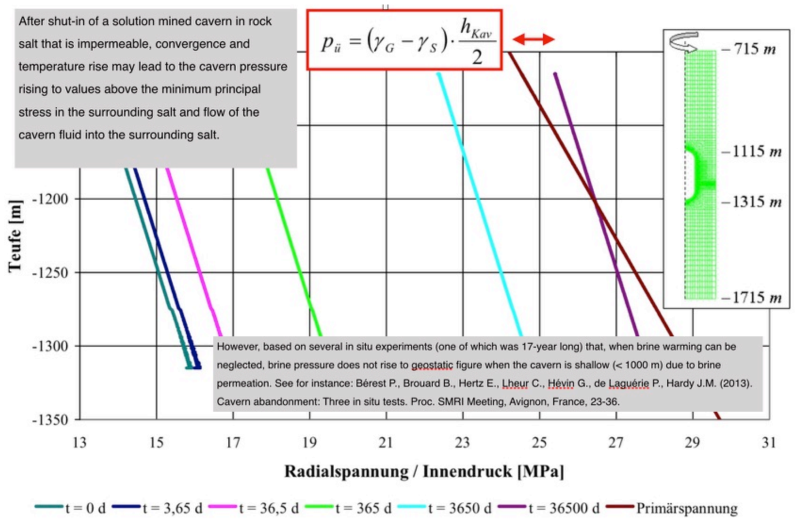



*Bild 3.2 Darstellung des konvergenzbedingt ansteigenden Kaverneninnendrucks in einer verschlossenen solegefüllten Kaverne, Lux (2006a)*

***Figure 48 – After shut-in of a solution mined cavern in rock salt that is impermeable, convergence and temperature rise may lead to the cavern pressure rising to values above the minimum principal stress in the surrounding salt and flow of the cavern fluid into the surrounding salt. After: Wolters, R., Düsterloh, U., 2009. Weiterentwicklung der EDV-Software INFIL zur Simulation des druckgetriebenen Infiltrationsprozesses von Fluiden in ein nicht permeables Barrieren-Gebirge (Salinar). Final Report No. 02C1355. Technical University Clausthal, Clausthal, Germany. However, based on several in situ experiments (one of which was 17-year long) that, when brine warming can be neglected, brine pressure does not rise to geostatic figure when the cavern is shallow (< 1000 m) due to brine permeation. See for instance: Bérest P., Brouard B., Hertz E., Lheur C., Hévin G., de Laguérie P., Hardy J.M. (2013). Cavern abandonment: Three in situ tests. Proc. SMRI Meeting, Avignon, France, 23-36.***

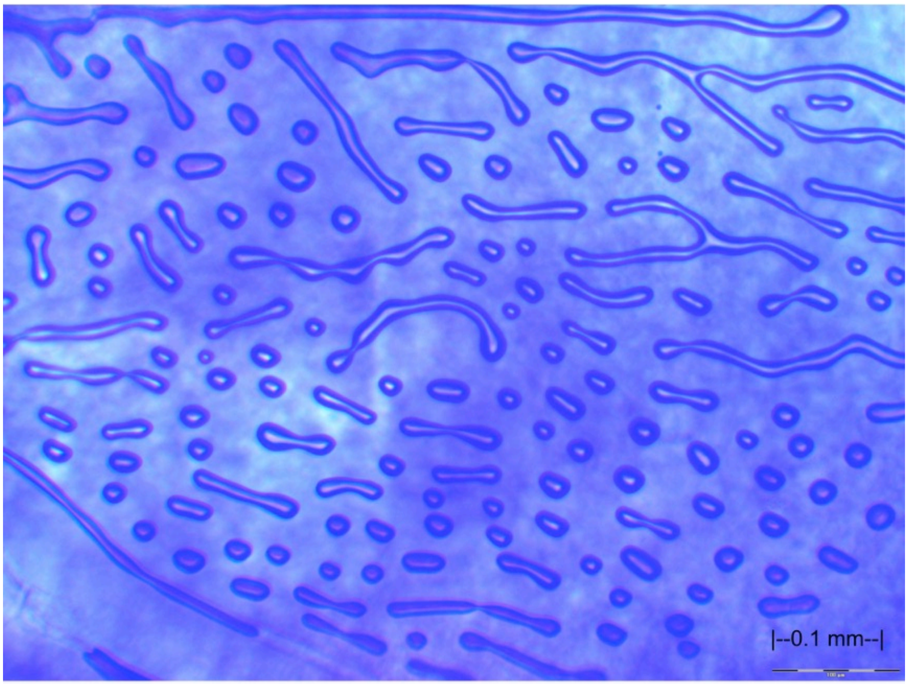



*Figure 49. In deeply buried, clean Zechstein salt, the grain boundaries typically contain a small amount of brine in isolated, thermodynamically equilibrated micropores, separated by solid state grain boundaries. This salt therefore has a very low permeability, and can be considered practically impermeable, as long as this grain boundary structure is not disturbed. This is illustrated by this optical microscope image of a grain boundary in natural Zechstein salt, showing isolated, thermodynamically equilibrated micropores, separated by solid state grain boundaries. Marc Sadler, MSc Thesis, RWTH Aachen University.*

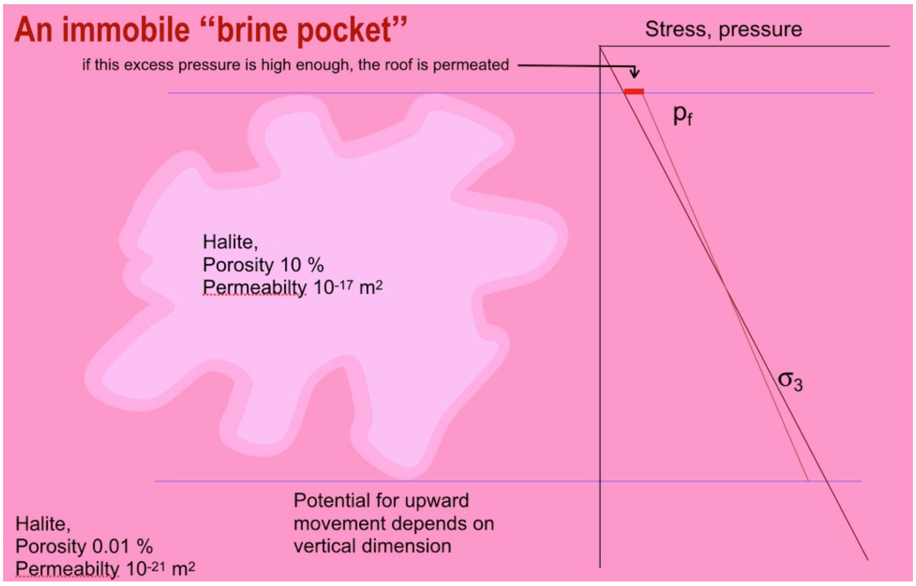



*Figure 50. In drilling Zechstein salt, “brine pockets” can pose major problems. These are thought to consist of slightly porous and permeable volumes of rock salt, containing brine at close to lithostatic pressure. However, when the vertical dimension of these pockets is small enough, the pockets can be geologically stable, even though the pressure at the top is slightly larger than the minimum principal stress in the salt. These brine pockets are natural analogues of brine migrating upwards from abandoned caverns. NB: In principle, in the presence of a thermal gradient, convection can take place leading to dissolution at the warmer face of the pocket. See for example: M.H.A.A. Zijp, M.A. Huijgen, M. Wilpshaar, R. Bouroullec, J.H. ter Heege (2018) Stringers in Salt as a Drilling Risk. Report, TNO2018 R10975*

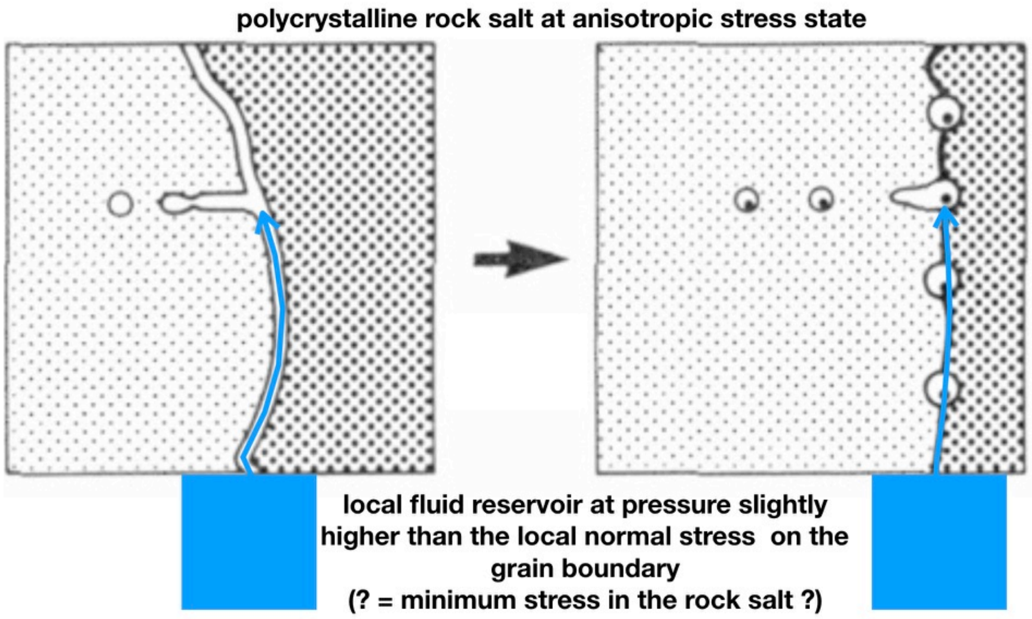



***Figure 51 At the microscale, the problem to be solved is the permeation of salt grain boundaries by high pressure (saturated) brine. It is clear from this diagram that this is a complex microphysical problem, and also that the process is very different between the two different grain boundary structures shown in this diagram and discussed in Part 1 of this report. microcracking, grain boundary healing, dilsocation creep, pressure solution all play important but unexplored roles, together with grain-scale stress.***

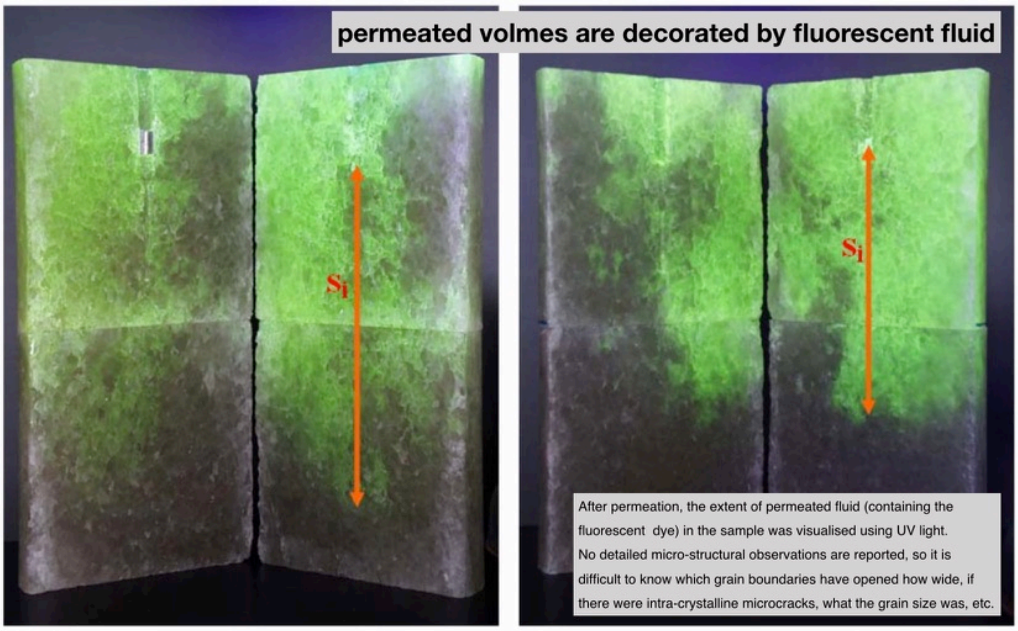



***Figure 52. Typical results of fluid permeation experiments in the Clausthal Laboratory. After: Lux, K.-H., 2005. Zum langfristigen Tragverhalten von verschlossenen solegefüllten Salzkavernen – ein neuer Ansatz zu physikalischer Modellierung und numerischer Simulation: Theoretische und laborative Grundlagen. Erdöl, Erdgas, Kohle 121, 414–422. and Wolters, R., Lux, K.-H., Düsterloh, U., 2011. Fluid infiltration processes into rock salt barriers resulting from fluid pressure build-up due to convergence, thermal expansion and gas generation. In: Li, X., Jing, L., Blaser, P. (Eds.), Impact of Thermo-Hydro-Mechanical Chemical (THMC) Processes on the Safety of Underground Radioactive Waste Repositories. European Commission, Luxembourg, 209–218. https://doi.org/10.2777/2***

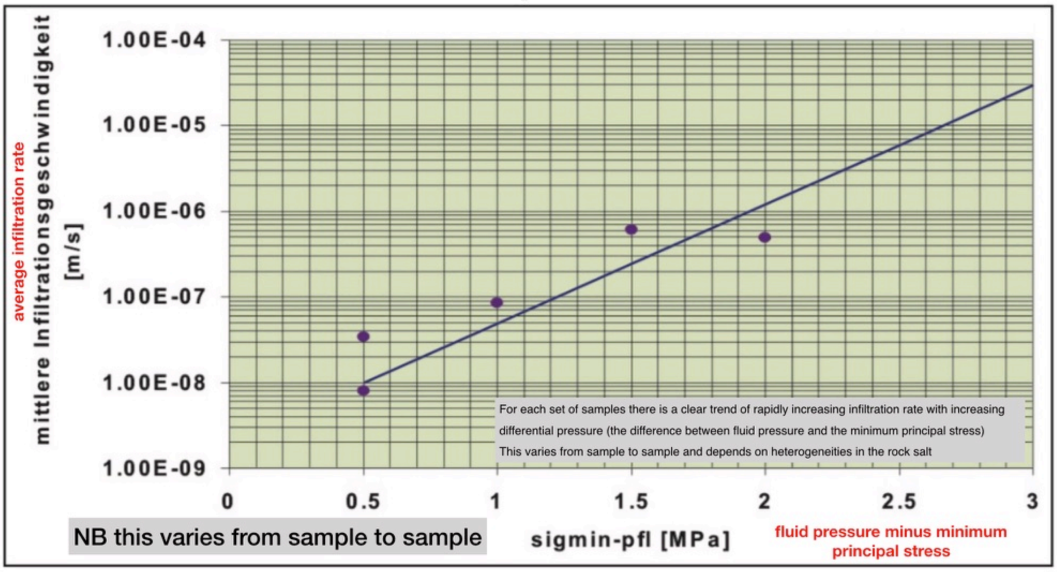



*Figure 53. Plot of average infiltration rate against the difference between fluid pressure and minimum principal stress applied to the salt sample (both numbers taken as negative, explaining the somewhat puzzling sign). An increase in overpressure from 0.5 to 2 MPa results in two orders of magnitude, nonlinear increase. Note also the differences between samples which are supposed to be very similar. After: Lux, K.-H., 2006. Zum langfristigen Tragverhalten von verschlossenen solegefüllten Salzkavernen – ein neuer Ansatz zu physikalischer Modellierung und numerischer Simulation: Rechnerische Analysen und grundlegende Erkenntnisse. Erdöl, Erdgas, Kohle 122, 150–158.*

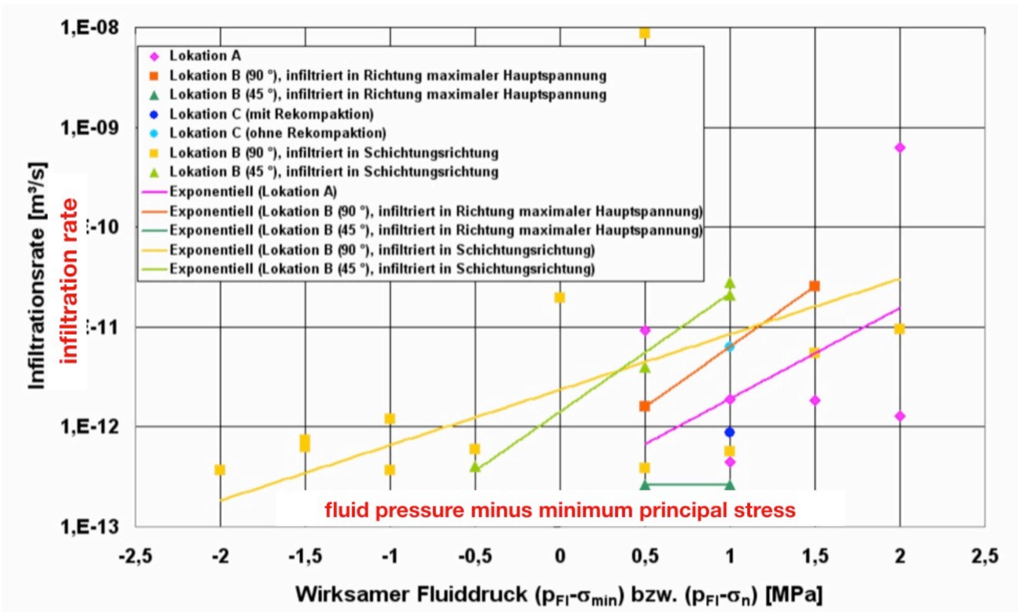



*Figure 54 The results of infiltration and permeation by the Clausthal Group show show that the kinetics of permeation vary with rock salt heterogeneity, microstructure and impurities. In this plot the large differences between samples are summarized. In some cases, infiltration is possible at excess pressure below zero. Because of the lack of microphysical basis, it is not possible to reliably extrapolate these results to low excess pressures. However, it is clear that in heterogeneous rock salt these differences will lead to strongly localized infiltration and permeation. Although this project does not aim to model this process at the cavern scale, here we propose that therefore permeation after cavern abandonment will be strongly heterogeneous and localised. After: Wolters, R., Düsterloh, U., 2009. Weiterentwicklung der EDV-Software INFIL zur Simulation des druckgetriebenen Infiltrationsprozesses von Fluiden in ein nicht permeables Barrieren-Gebirge (Salinar). Final Report No. 02C1355. Technical University Clausthal, Clausthal, Germany.*

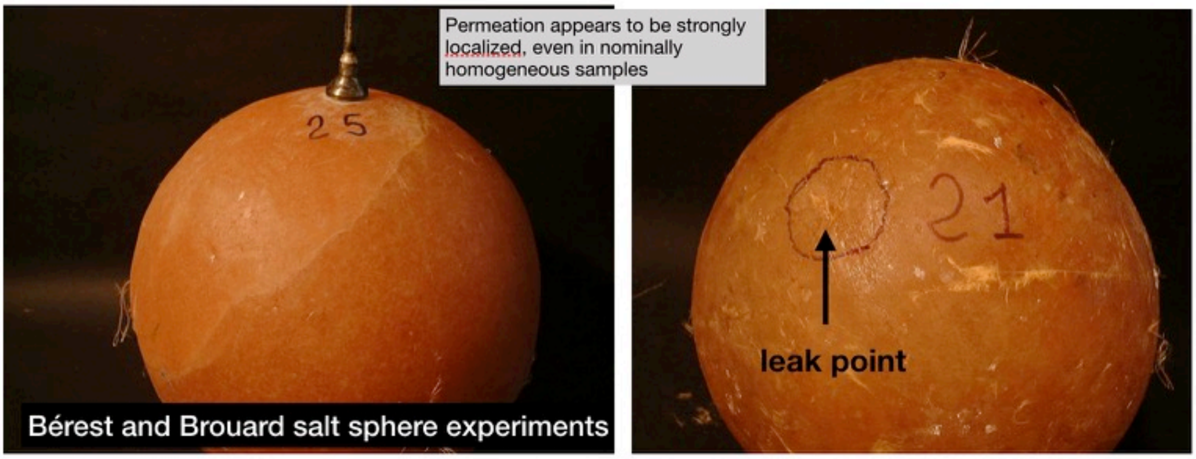



*Figure 55 The Paris salt sphere permeation experiments, apparently strongly localized. Bérest, P., Brouard, B., de Greef, V., 1999. Salt permeability testing RFP 98-1 and 2: The influence of permeability and stress on spherical hollow salt samples. Research Report. Laboratoire de M´ecanique des Solides (Ecole polytechnique), Paris, France.*

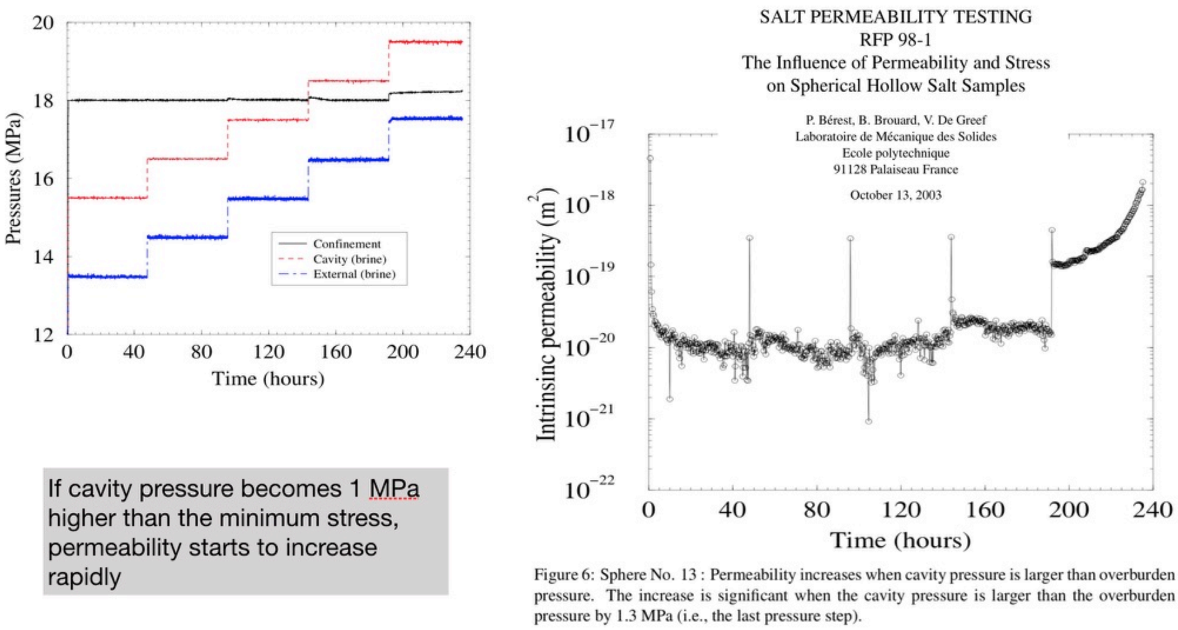



*Figure 56 In the Paris experiments, if cavity pressure becomes 1 MPa higher than the minimum stress, permeability starts to increase rapidly. Bérest, P., Brouard, B., de Greef, V., 1999. Salt permeability testing RFP 98-1 and 2: The influence of permeability and stress on spherical hollow salt samples. Research Report. Laboratoire de M´ecanique des Solides (Ecole polytechnique), Paris, France.*

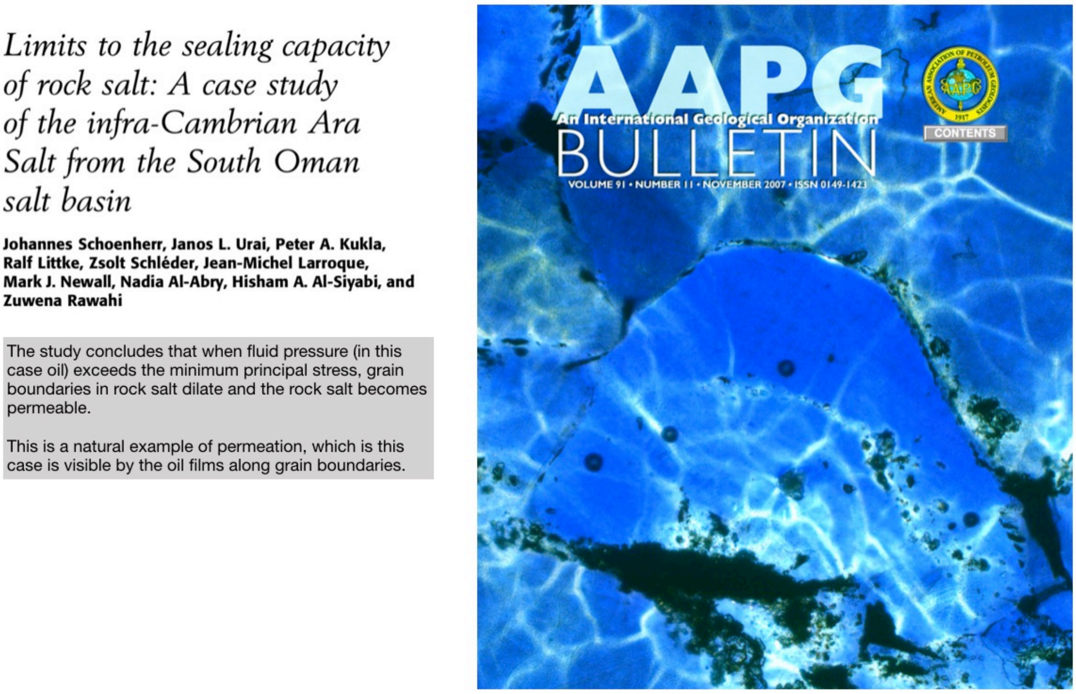



***Figure 57. When fluid pressure (in this case oil) exceeds the minimum principal stress, grain boundaries in rock salt dilate and the rock salt becomes permeable. This is a natural example of permeation, which in this case is visible by the oil films along grain boundaries. After: The study concludes that when fluid pressure (in this case oil) exceeds the minimum principal stress, grain boundaries in rock salt dilate and the rock salt becomes permeable. After: Schoenherr, J., Urai, J.L., Kukla, P.A., Littke, R., Schleder, Z., Larroque, J.-M., Newall, M.J., Al-Abry, N., Al-Siyabi, H.A., Rawahi, Z., 2007. Limits to the sealing capacity of rock salt: A case study of the infra-Cambrian ara salt from the south Oman salt basin. AAPG Bulletin 91, 1541–1557.***

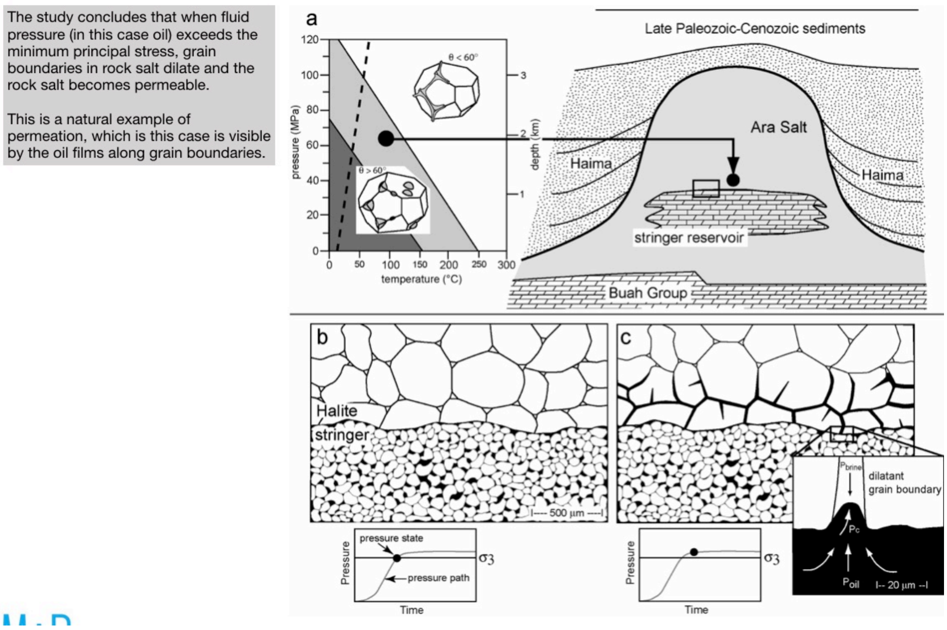



***Figure 58. Micro- and macromechanical model of the premeation of roock salt by oil and by brine in nature. This is a natural example of permeation, which is this case is visible by the oil films along grain boundaries. The study concludes that when fluid pressure (in this case oil) exceeds the minimum principal stress, grain boundaries in rock salt dilate and the rock salt becomes permeable. After: Schoenherr, J., Urai, J.L., Kukla, P.A., Littke, R., Schleder, Z., Larroque, J.-M., Newall, M.J., Al-Abry, N., Al-Siyabi, H.A., Rawahi, Z., 2007. Limits to the sealing capacity of rock salt: A case study of the infra-Cambrian ara salt from the south Oman salt basin. AAPG Bulletin 91, 1541–1557.***

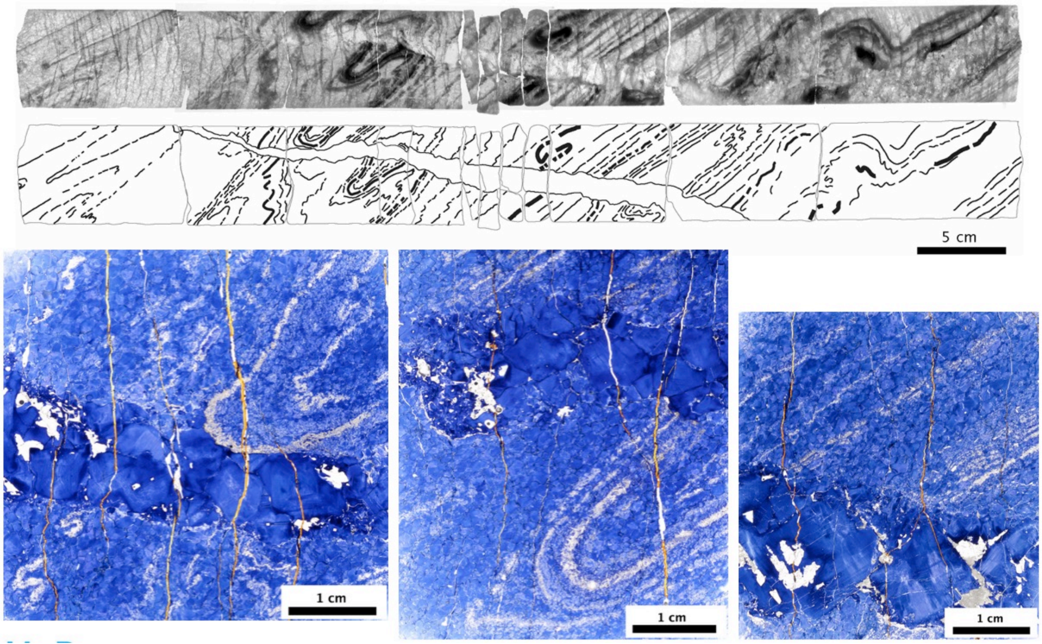



*Figure 59. Natural example of hydrofracturing in rock salt, followed by healing of the fracture. After: Schleder, Z., Urai, J.L., Nollet, S., Hilgers, C., 2008. Solution-precipitation creep and fluid flow in Halite: A case study of Zechstein (Z1) rocksalt from Neuhof salt mine (Germany). International Journal of Earth Sciences 97, 1045–1056.*

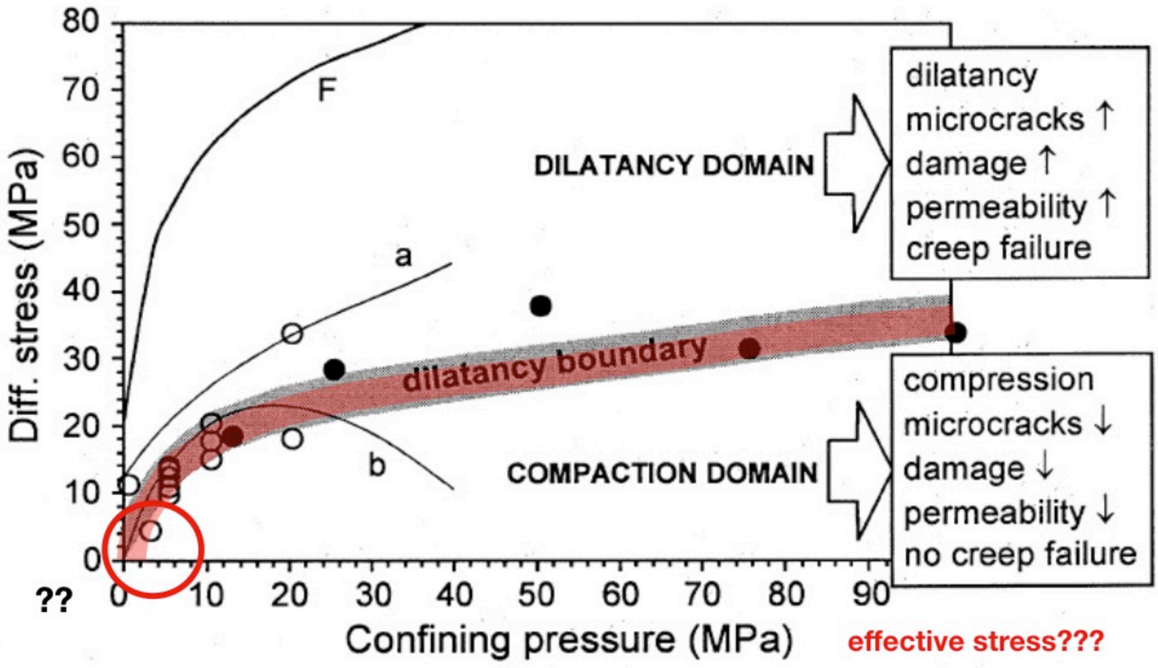



*Figure 60. From many triaxial tests the dilatancy boundary in rock salt is reasonably known at high differential stress. However, the effects of dilatancy on microstructure mechanical properties are not well implemented in the salt engineering community. Here there are major effects if samples can lose their in-situ brine content by evaporation. In addition, the relevant part of this diagram for the permeation problem considered here (low effective stress, low differential stress) is poorly known and needs more study, including microstructural investigations. After: Popp, T., Kern, H., 2000. Monitoring the state of microfracturing in rock salt during deformation by combined measurements of permeability and P- and S- wave velocities. Physics and Chemistry of the Earth, Part A: Solid Earth and Geodesy 25, 149–154.*

This paper is a good example of measurement of dilatancy, permeability integrated with microstructural observation and microphysical modelling using a connected microcrack model.

The results are indirectly applicable to the abandonment problem, because dilatancy is created by the imposed stress, and fluid flow is by gas, in dry rock salt and no fluid effects are studied.

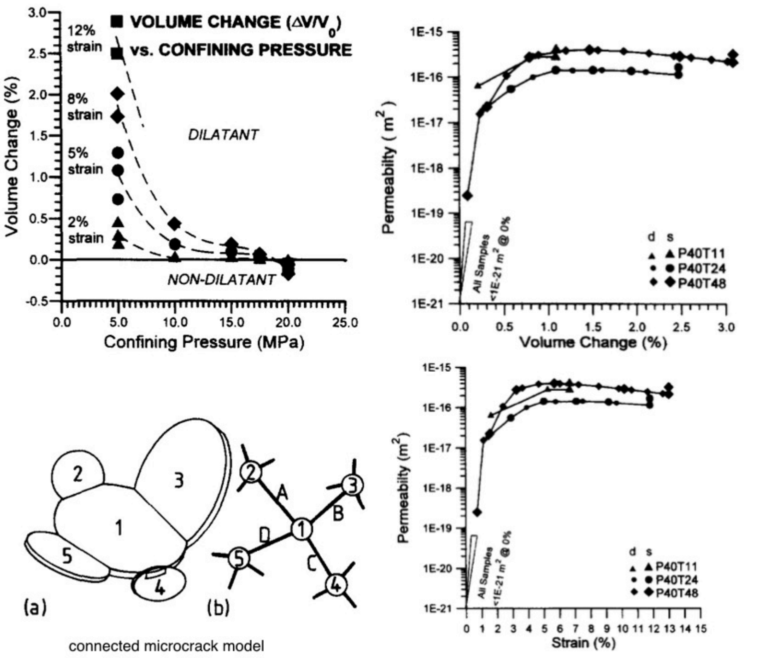



*Figure 61. Materials science based measurement of the evolution of dilatancy and permeability in rock salt. After: Peach, C.J., Spiers, C.J., 1996. Influence of crystal plastic deformation on dilatancy and permeability development in synthetic salt rock. Tectonophysics 256, 101–128.*

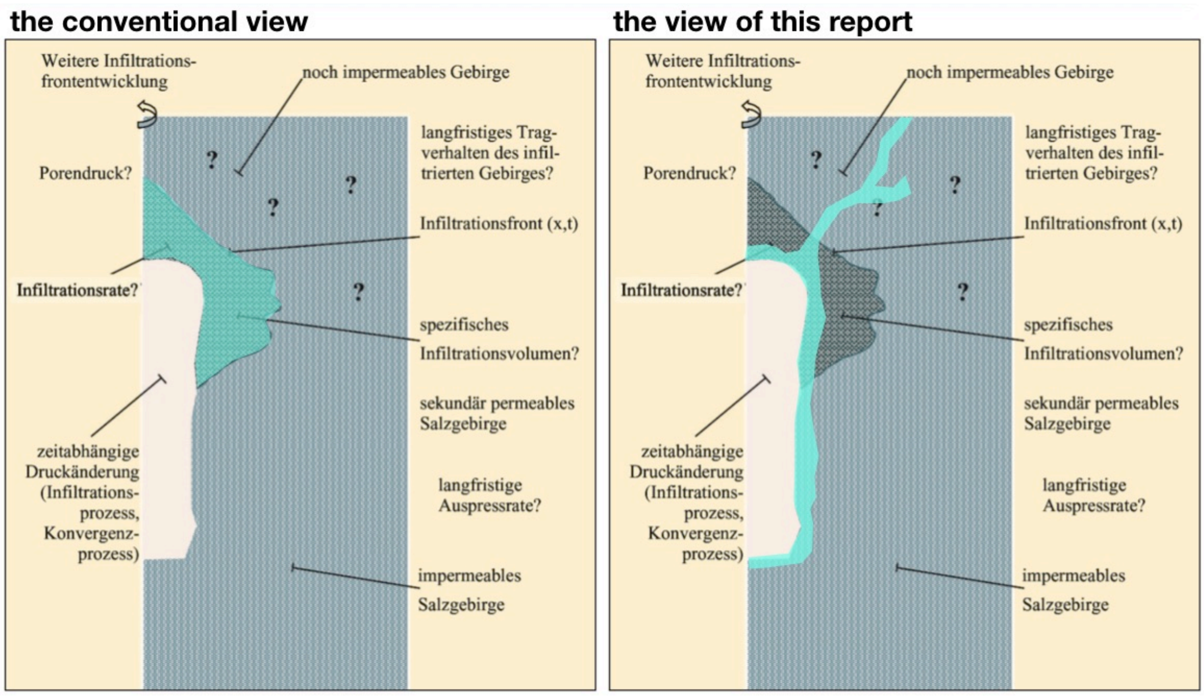



*Figure 62. Schematic illustration of the geometry of the infiltrated volume and the relevant processes and parameters. Let picture After: Lux, K.-H., 2006. Zum langfristigen Tragverhalten von verschlossenen solegefüllten Salzkavernen – ein neuer Ansatz zu physikalischer Modellierung und numerischer Simulation: Rechnerische Analysen und grundlegende Erkenntnisse. Erdöl, Erdgas, Kohle 122, 150–158. Right picture: the preferential fingering proposed in this review.*

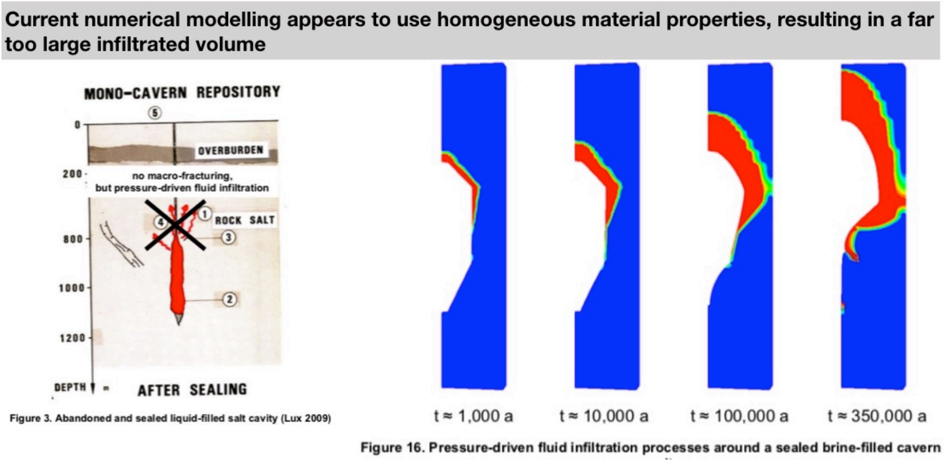



*Figure 63 Although this part of the project does not aim to model this process at the cavern scale, (this will be done in the second part of the project) here we propose that permeation after cavern abandonment will be much more heterogeneous and localisedthan predicted by current models. After: Wolters, R., Lux, K.-H., Düsterloh, U., 2017. Rock-mechanical investigations regarding the proof of long-term safety of abandoned salt production cavities using hazardous waste as backfill material. Presented at the SMRI Spring 2017 Technical Conference, 23–26 April 2017, Solution Mining Research Institute, Albuquerque, New Mexico, USA, 1–19.*

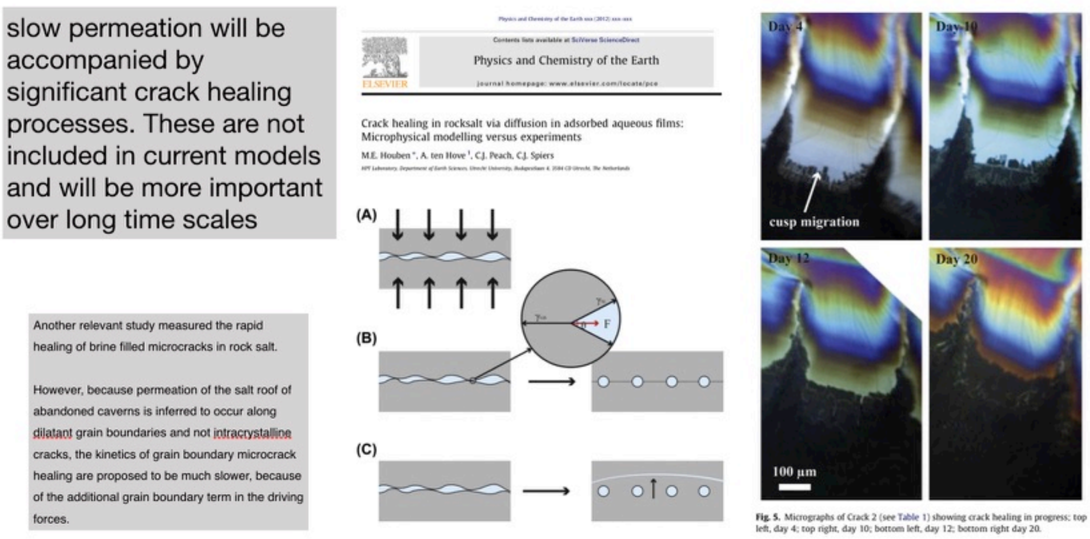
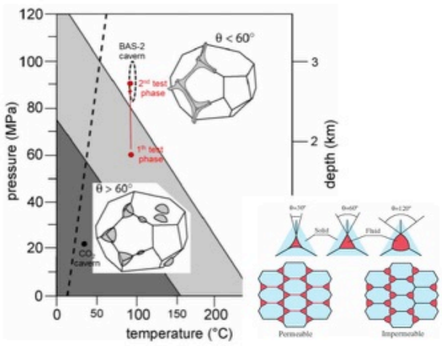



*Figure 64 Crack sealing experiments observed in transmitted light. Note that these are not grain boundaries but cracks in single crystals which heal much faster than grain boundaries. The kinetics of grain boundary healing are not well known and need further research to quantify and predict over long time scales. After: Houben, M.E., ten Hove, A., Peach, C.J., Spiers, C.J., 2013. Crack healing in rocksalt via diffusion in adsorbed aqueous films: Microphysical modelling versus experiments. Physics and Chemistry of the Earth, Parts A/B/C 64, 95–104. https://doi.org/10.1016/j.pce.2012.10.001*





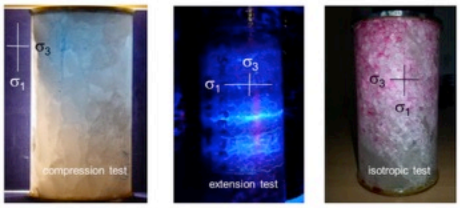



Figure 4: Pressure driven percolation after overcoming the minimum principle stress $\sigma_3$ directed to the maximum principle stress $\sigma_1$ within an anisotropic stress field (left and center) and with a homogenous diffusion along all grain boundaries at isotropic stress (right)

"Pressure driven percolation is the most important failure mechanism responsible for loss of integrity of salt rock barriers at fluid pressures higher than minimum principle stress which act as the percolation threshold. Only after overcoming a percolation threshold the pressure-driven opening and interconnection of flow paths along grain boundaries is initiated in the salt rock and induces a directional percolation in the direction of the maximum principal stress."

NB: the value of the equilibrium dihedral angle is the key to this process. No microstructural study was undertaken to measure this parameter.
The conclusions of this study must be reevaluated critically

***Figure 65 A recent paper by Ghanbarzadeh et al (2015) proposed that thermodynamically controlled reequilibration of grain boundary fluid connectivity could provide an additional process of permeation. Ghanbarzadeh, S., Hesse, M.A., Prodanovi, M., Gardner, J.E., 2015. Deformation-assisted fluid percolation in rock salt. Science 350, 1069–1072. This model was critically discussed by Brückener et al., albeit without microstructural inverstigations, which bring their conclusions in question. Brückner, D., Minkley, W., 2016. Fluid percolation and the tightness of salt. Presented at the SMRI Spring 2016 Technical Conference, 25–26 April 2016, Solution Mining Research Institute, Galveston, Texas, USA, 1–11.***

discrete element modelling of percolation

Stability and integrity of salt caverns under consideration of hydro-mechanical loading

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Stability and integrity of salt caverns under consideration of hydro-mechanical loading
ByW. Minkley, M. Knauth, T. Fabig & N. Farag (2015)

Mechanical Behaviour of Salt VIII
Edited By Lance Roberts, Kirby Mellegard, Frank Hansen

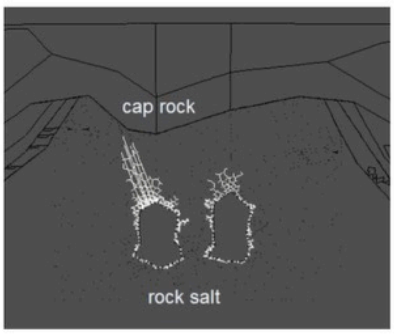



Figure 19. Pressure-driven percolation in brine-filled caverns, 10000 years after sealing of the caverns with reduced percolation threshold along the bedding planes in saliniferous strata (above left cavern S2).

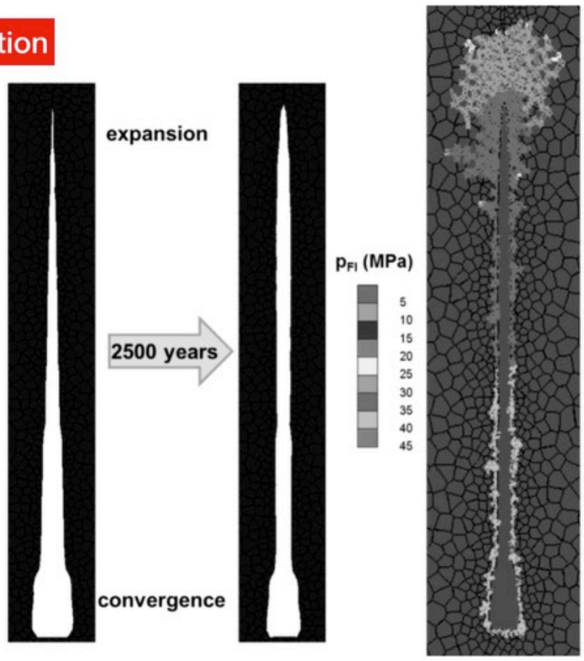
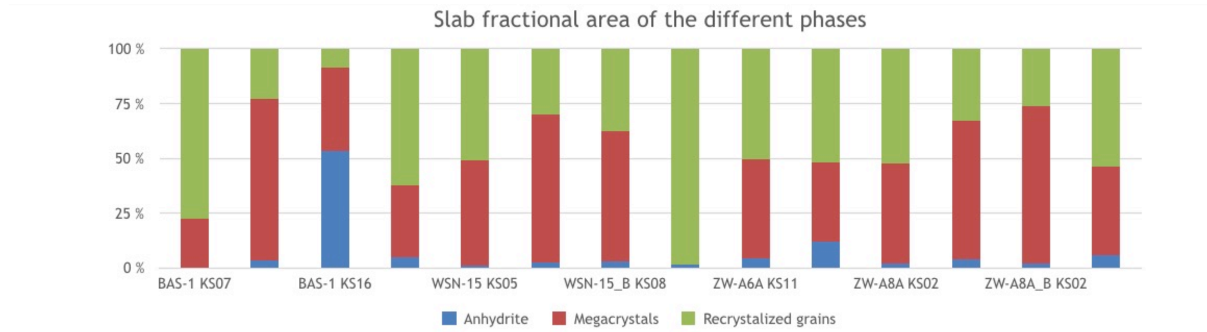



Figure 17. Computed deformation caused by brine pressure, and pressure-driven percolation of a sealed cavern in a depth of 1350–1900 m.

*Figure 66. Discrete element modelling of permeation in salt. After: Stability and integrity of salt caverns under consideration of hydro-mechanical loading ByW. Minkley, M. Knauth, T. Fabig & N. Farag (2015) in: Mechanical Behaviour of Salt VIII Edited By Lance Roberts, Kirby Mellegard, Frank Hansen*


Source: https://www.nlog.nl/kaart-boringen, Vergunningen steenzout 02/01/2019

*Figure 67 Site locations core material used in the microstructural study*




| | Barradel | | | | Winschoten | | | Zuidwending | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| SAMPLE | BAS-1 KS07 | BAS-1 KS15 | BAS-1 KS16 | BAS-2 KS14 | WSN-15 KS05 | WSN-15 KS08 | WSN-15_B KS08 | ZW-A2B KS06 | ZW-A6A KS11 | ZW-A6A_B KS11 | ZW-A8A KS02 |
| Anhydrite | 0.0% | 3.2% | 53.4% | 4.7% | 0.9% | 2.3% | 2.8% | 1.5% | 4.5% | 12.1% | 2.1% |
| Single Crystals | 22.3% | 74.0% | 37.8% | 33.2% | 48.0% | 67.6% | 59.7% | 0.0% | 45.1% | 36.1% | 45.8% |
| Recrystallized grains | 77.7% | 22.8% | 8.8% | 62.1% | 51.1% | 30.2% | 37.6% | 98.5% | 50.4% | 51.8% | 52.1% |

MaP confidential
Microstructure and Pores

*Figure 68 Phase mapping of core slab photos showed that the core samples were heterogeneous and predominantly consisting of three phases: Halite mega-crystals alternating with recrystallized Halite polycrystalline layers and with Anhydrite layers. This causes an overall bimodal grain size distribution of the Halite fraction. While hundreds of recrystallized grains were measured in the polycrystalline layers, only a few, but large single crystals (mega-crystals) were found. The size of the mega-crystals was not measured as it exceeded in most cases the size of the thin section and even the slab.*


*Figure 69 Core slab (KS_15) of well BAS-1 from 2845 m depth in transmitted light showing deformed (layer parallel shear, foliation) Anhydrite layers boundinghalite megacrystals. Red frame indicating thin section location.*

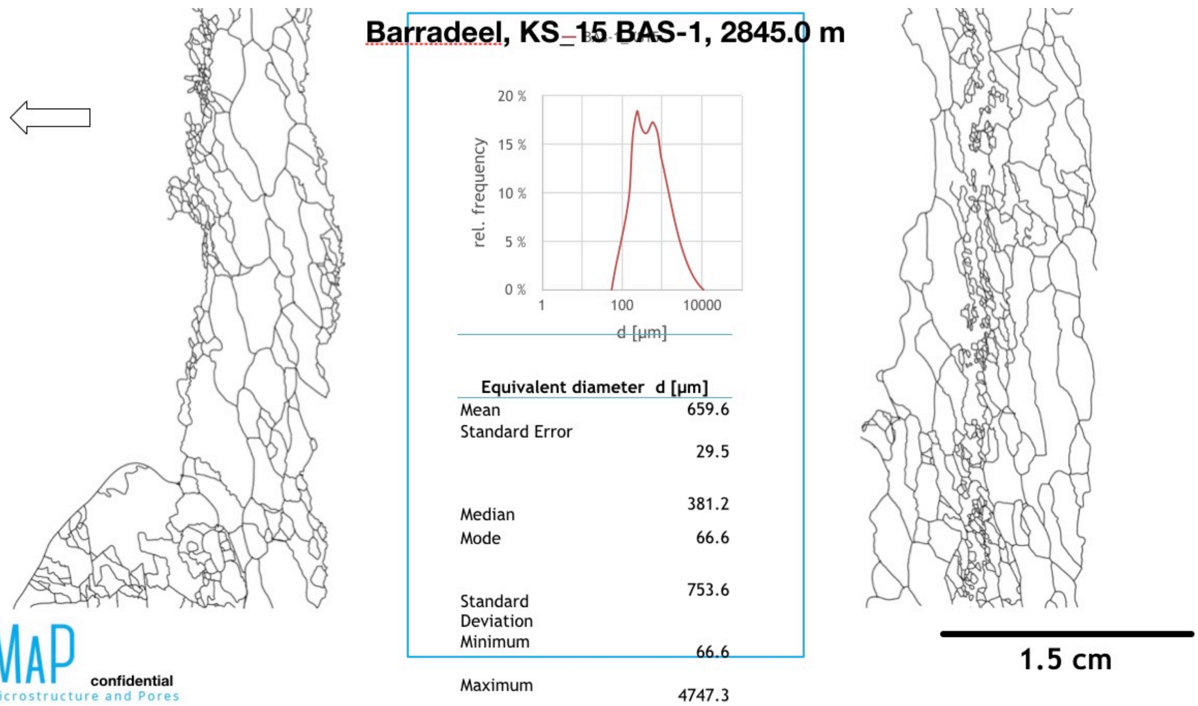



*Figure 70 Barradeel, KS_15 BAS-1, 2845.0 m, Grain Size analysis. Grain size distribution of the recrystallized areas was measured using micrographs acquired in reflected light. The grain boundaries were exposed using chemical etching facilitating semi-automated image analysis. The recrystallized grain size was log-normally (with visible influence of small grain size) distributed with decreasing grain size towards the interbedded anhydate layers and close to Halite single crystals. Halite megacrystals (one crystal in this thin section) were excluded from the statistics as they cross-cut the thin section.*

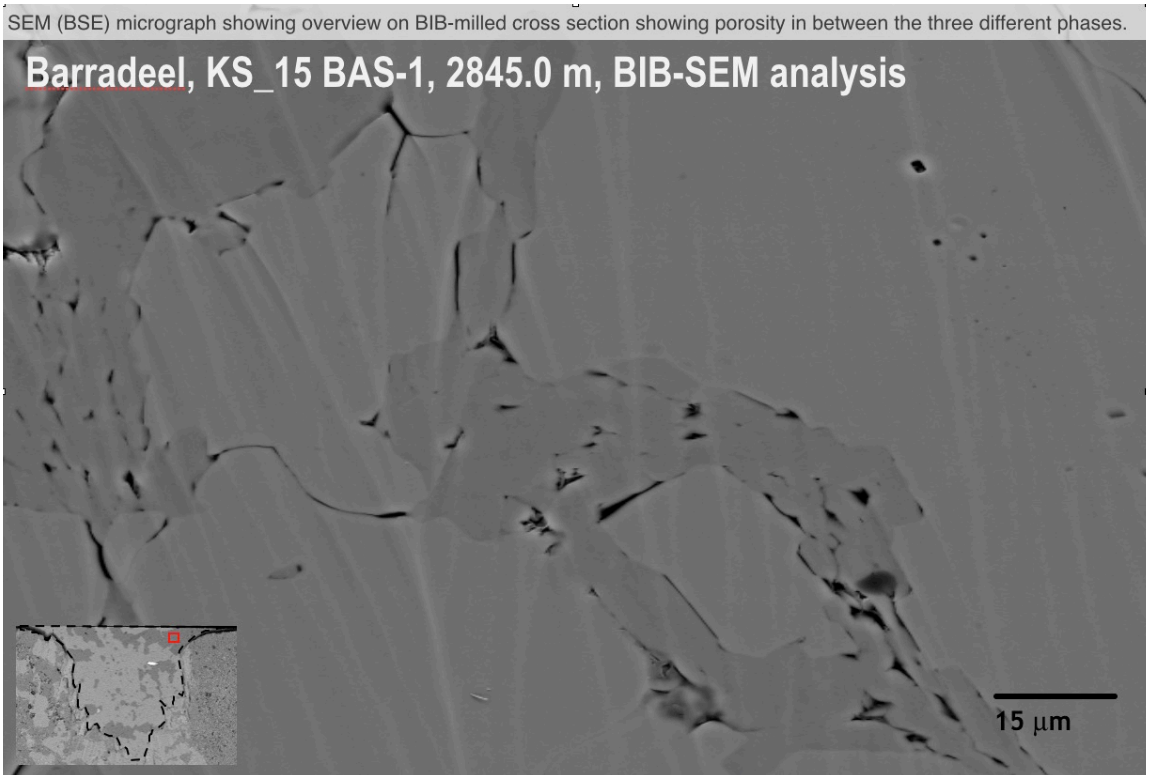



*Figure 71 SEM (BSE) micrograph showing overview on BIB-milled cross section showing porosity in between the three different phases.*

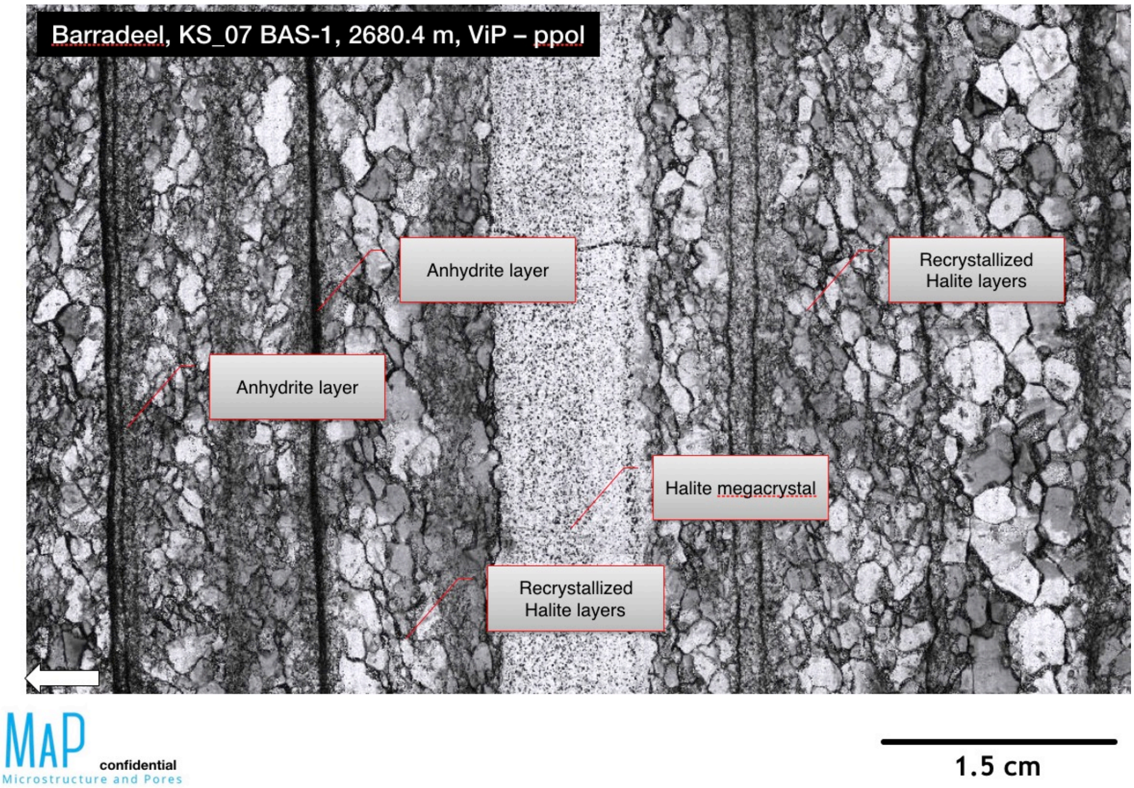
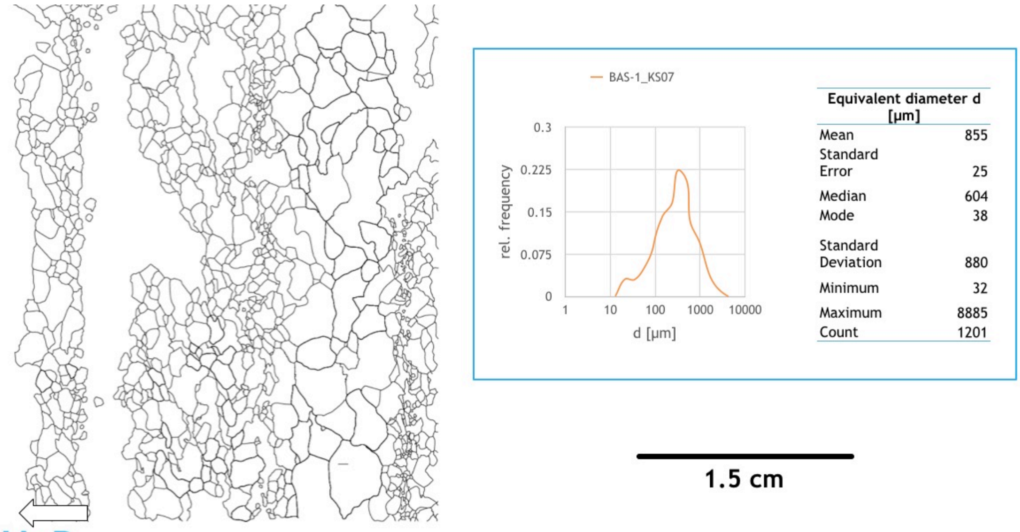



*Figure 72 ViP micrograph acquired in transmitted plane polarized light was used to investigate the grain boundary fluid morphology, as well as the nature and distribution of second phase impurities such as Anhydrite grains, enclosed to Halite*

***grains and grain boundaries. This overview of the entire cross-section allows fast discrimination of Halite single crystals and recrystallized areas.***




MAP confidential
Microstructure and Pores

***Figure 73 Barradeel, KS_07 BAS-1, 2680.4 m, grain size analysis. Grain size distribution of the recrystallized areas was measured using micrographs acquired in reflected light. The grain boundaries were exposed using chemical etching facilitating semi-automated image analysis. The recrystallized grain size was log-normally distributed with decreasing grain size towards the anhydrite layers and close to Halite megacrystals. Halite megacrystals (one crystal in this thin section) were excluded from the statistics because they are larger than the thin section. It is clear that pressure solution creep and fluid permeation in this rock salt will be highly heterogeneous.***

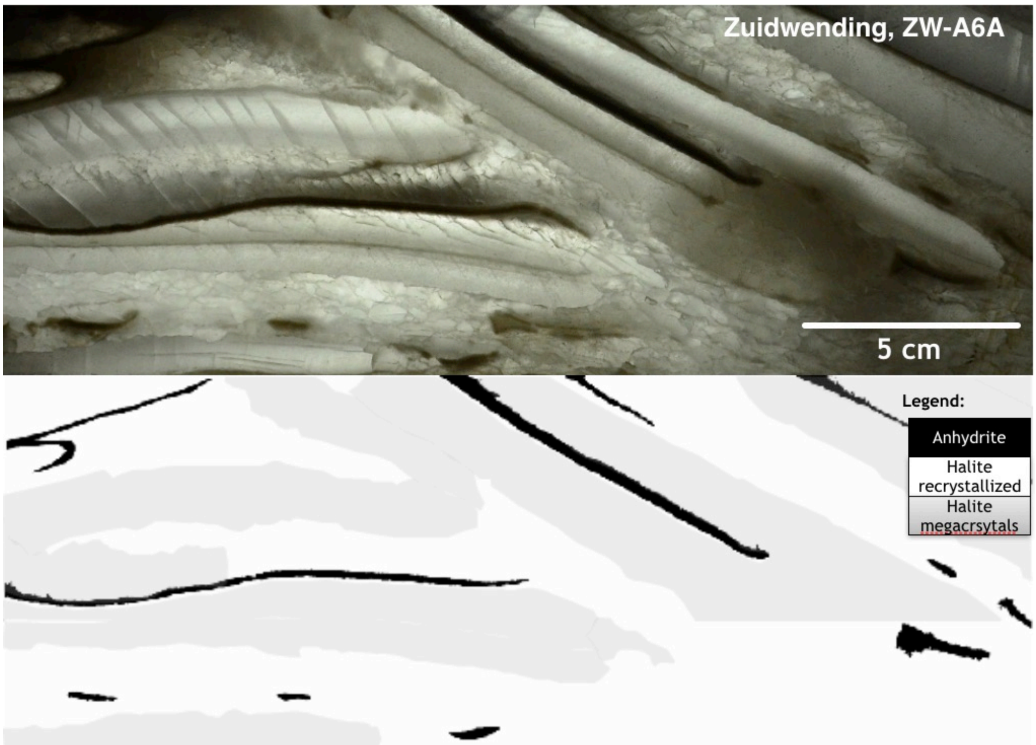
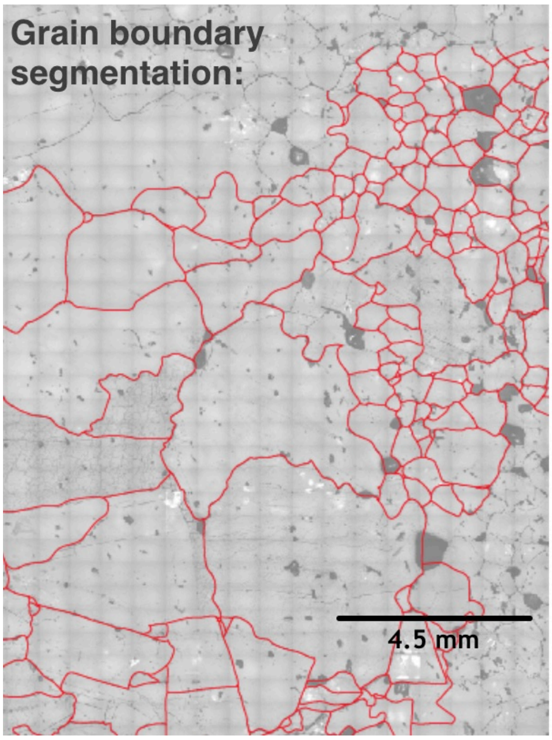
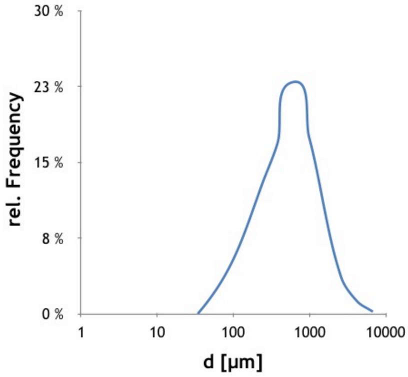



***Figure 74 Photographs of slabs using incident and transmitted light (upper picture) were used to estimate areal fractions of the different phase at the scale of the core and to select thin section locations. The slab photos were segmented (lower image) using semi-automated image processing and analysis to derive the area fraction of Anhydrite, recrystallized Halite,***

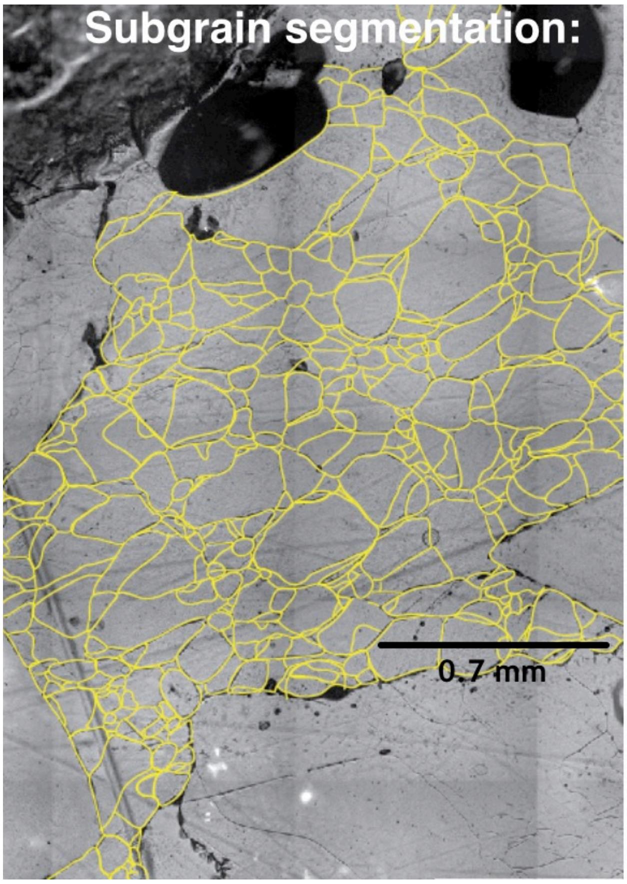
***and Halite megacrystals. It is clear that pressure solution creep and fluid permeation in this rock salt will be highly heterogeneous.***

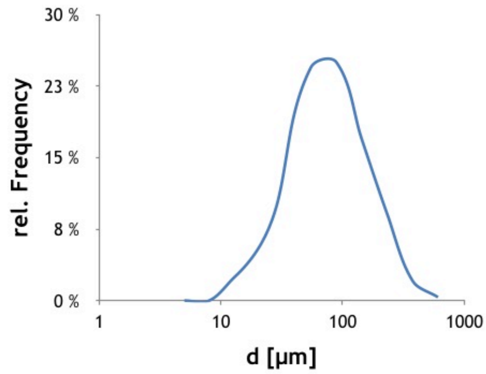



| Equivalent diameter d [µm] | |
|---|---|
| Mean | 593 |
| Standard Error | 13 |
| Median | 430 |
| Mode | 43 |
| Standard Deviation | 579 |
| Minimum | 43 |
| Maximum | 4769 |
| Count | 1912 |

***Figure 75. Zuidwending, ZW-A6A ViP reflected light image with traced grain boundaries. Grain size is computed from the spherical equivalent diameter (d) of the traced area and presented as the relative frequency [%] as a function of d [µm]. Dynamically recrystallized grain size is inversely proportional to the stress difference and thus often used as a piezometer. Grain size is also a key parameter to determine the rate of pressure solution.***




| Equivalent diameter d [µm] | |
|---|---|
| Mean | 76.3 |
| Standard Error | 1.5 |
| Median | 58.7 |
| Mode | 9.0 |
| Standard Deviation | 60.0 |
| Minimum | 9.0 |
| Maximum | 516.4 |
| Count | 1682 |

***Figure 767 Zuidwending, ZW-A6A The steady state subgrain size is inversely proportional to the stress difference and thus often used as a piezometer (e.g., Schleder et al., 2005). It is interpreted to represent the maximum deviatoric stress seen by the salt, after recrystallization. The present day virgin deviatoric stress can be lower due to stress relaxation in the tectonically quiet Tertiary. Subgrain size is calculated as the spherical equivalent diameter (d) and presented as the relative frequency [%] as a function of d [µm].***

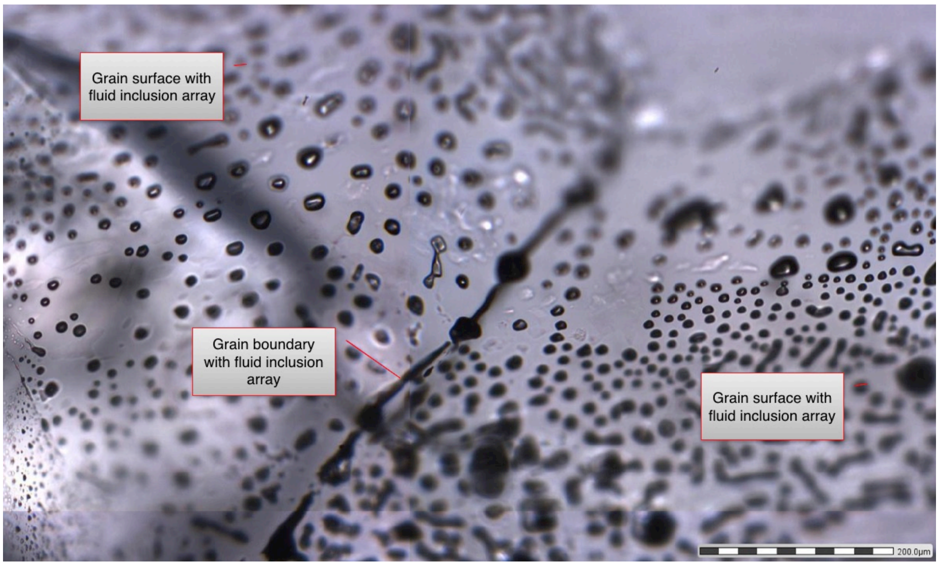



***Figure 77 Zuidwending, KS_11 ZW-A6A, 599.4 m, grain boundary structure. ViP micrograph showing triple junction of recrystallized Halite grains with fluid inclusion arrays on the grain boundaries.***

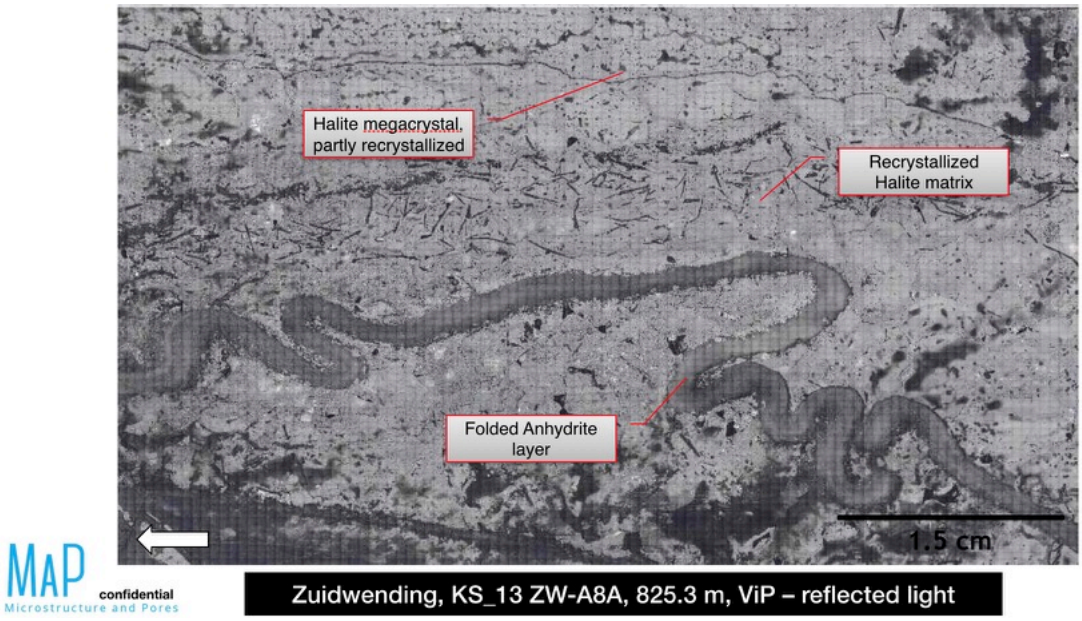



*Figure 78. Folded anhydrite layers ViP micrograph acquired in reflected light was used to measure grain- and subgrain size in the recrystallized parts of the thin section. For this purpose, the thin section surface was etched using the protocol proposed by Urai et al. (1987). Image showing overview of the entire cross-section, with folded Anhydrite layers and Halite single crystals embedded into recrystallized Halite matrix.*

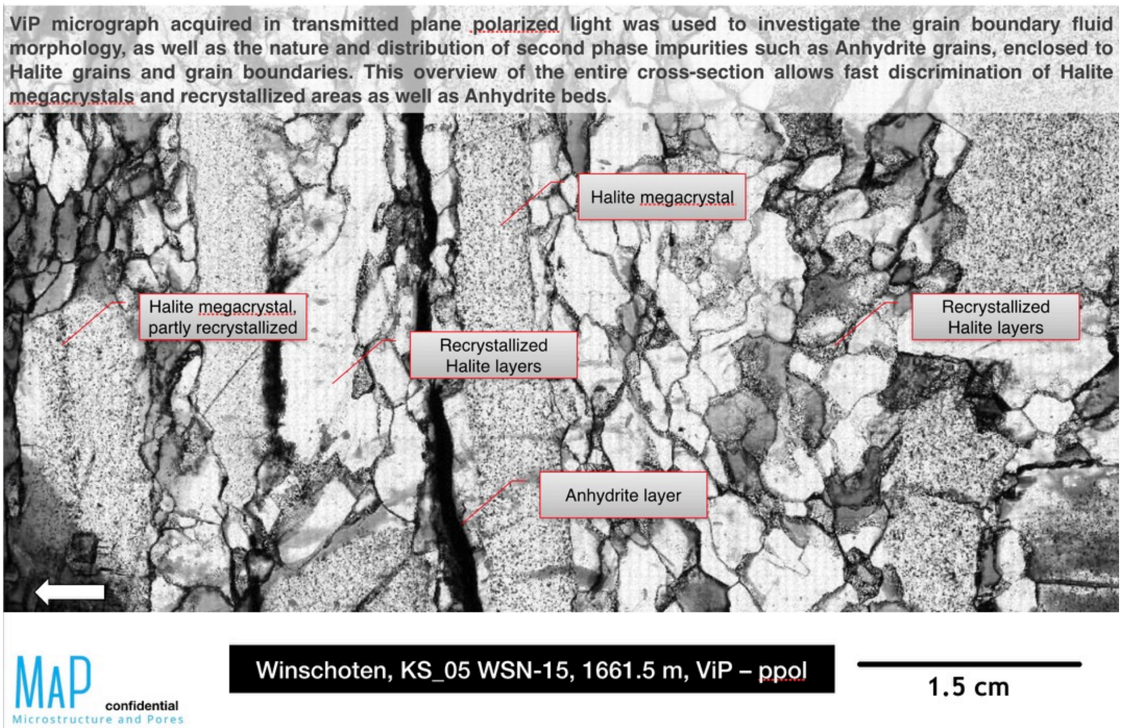



*Figure 79. Typical microstructure of the Winschoten domal salt core, formed by large strain deformation and recrystallization of a layered rock salt with halite megacrystals, which have been replaced by newly recrystallized, smaller Halite grains. ViP micrograph acquired in transmitted plane polarized light was used to investigate the grain boundary and subgrain boundary network, as well as the nature and distribution of fluid inclusions and second phase impurities. This overview of the entire cross-section allows rapid, semi-automatic mapping of grain boundaries and second phases and recrystallized areas as well as Anhydrite layers. Grain size is computed from the spherical equivalent diameter (d) of the traced area and presented as the relative frequency [%] as a function of d [µm]. Dynamically recrystallized grain size is inversely proportional to the stress difference and thus often used as a piezometer. Grain size is also a key parameter to*

***determine the rate of pressure solution. It is clear that pressure solution creep and fluid permeation in this rock salt will be highly heterogeneous.***

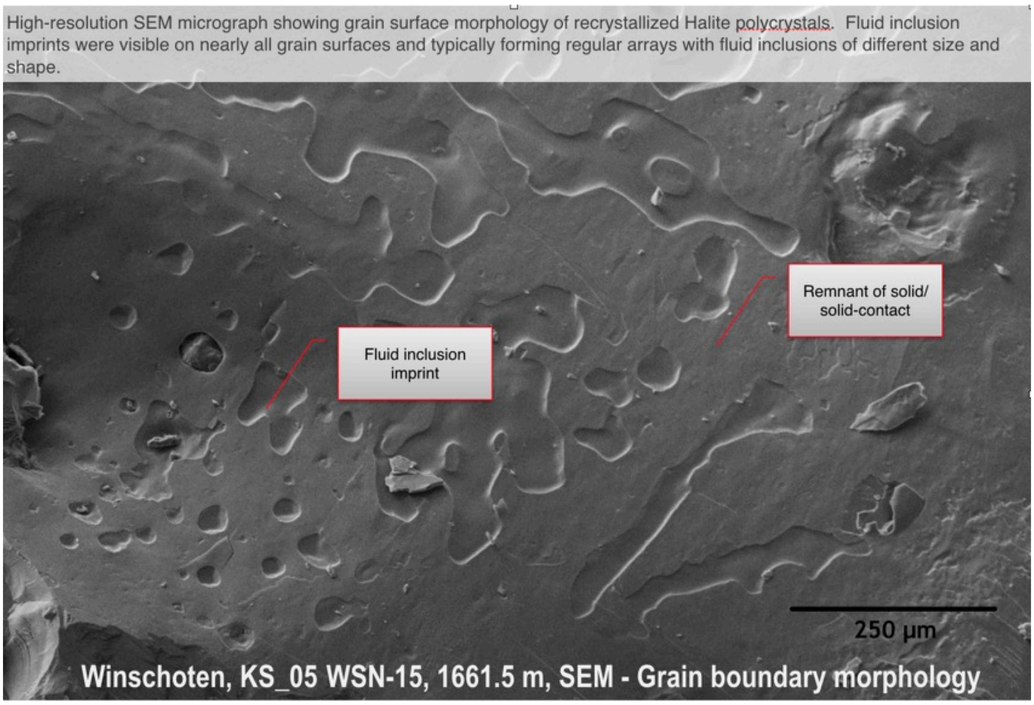



***Figure 80. Typical grain boundary structure in the recrystallized part of the Winschoten core. In this SEM SEII image, the image is of a grain's surface after it was separated from another grain along a grain boundary.***