The effect of grain boundary water on deformation mechanisms and rheology of rocksalt during long-term deformation

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ABSTRACT: Reliable modeling of the deformation of rocksalt under the very low strain rates characterizing long term engineering conditions or natural halokinetic requires extrapolation of experimentally-derived flow laws to rates much lower than those attainable in the laboratory. This extrapolation must be based on an understanding of the microscale deformation mechanisms operating under these conditions, from studies of natural laboratories. 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 under conditions of long-term deformation, grain boundary dissolution-precipitation processes, such as solution-precipitation creep (or “pressure solution”) and dynamic recrystallization, play a significant role. In this contribution, we briefly review the microphysics of grain boundary water related, solution-precipitation processes in halite, together with the flow behaviour associated with these processes, and we discuss the contribution of these mechanisms to the strain rate during long-term creep.

1 DEFORMATION MECHANISMS AND RHEOLOGY OF HALITE IN EXPERIMENTS

1.1 Deformation mechanisms

Polycrystalline halite rocks (rocksalt) consist of grains of halite (NaCl), with a diameter between 0.01 mm and several dm, containing impurities in solid solution, secondary mineral phases and fluids trapped in inclusions, grain boundaries or in pores. Under deviatoric stress, halite rocks can deform by a range of processes. The deformation mechanisms known to operate at temperatures relevant for engineering and natural halokinetic conditions (20-200 °C) are summarized in Fig. 1.

At very low effective confining pressures and high deviatoric stresses, inter- and intragranular microcracking, grain rotation and intergranular slip are important strain accumulating processes alongside crystal plasticity, and the mechanical properties and dilatational behaviour are dependent on the effective mean stress or effective confining pressure (Cristescu & Hunsche 1988, Peach & Spiers 1996, Cristescu 1998, Peach et al. 2001). At high enough deviatoric stress, the material fails in a (semi)brittle manner, with failure described by a pressure (effective mean stress) dependent failure envelope. With increasing effective mean stress, microcracking and dilatancy are suppressed and crystal plastic processes dominate.

Fig. 2 Reflected light optical micrograph of experimentally deformed halite (Asse Speisesalz, 150 °C, 50 MPa, 3x10⁻⁵ s⁻¹), showing dislocation slip lines, incipient subgrains and minor grain boundary microcracking. Image is approximately 1 mm wide.

Fig. 1. Schematic drawing of the microstructural processes that can operate during deformation of halite at temperatures in the range 20-200°C. Different shades of green represent crystals with different orientation. See text for explanation.
At temperatures in the range 100-200 °C, dislocation creep is important in laboratory experiments, and polycrystalline halite can deform to large strains by this mechanism (Fig. 2), even at confining pressures as low as 10 MPa. During this process subgrains are formed in the halite grains, and the diameter of the subgrains is correlated with the deviatoric stress (Carter et al., 1993). If the polycrystal contains small but significant amounts of water in the form of saturated brine inclusions or 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 (Schenk & Urai 2004, Schenk et al. 2006 - see Fig. 3). This recrystallization process involves grain boundary migration by solution-precipitation transfer across grain boundary water/brine films (Fig 3b), and is driven by chemical potential differences across grain boundaries related to the dislocation density differences between old deformed grains and newly growing grains (Peach et al 2001). In strongly deformed, wet halite, the migration process is very rapid, reaching rates up to 10 nm/s at room temperature (Schenk et al., 2006).

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 (Schenk & Urai, 1999, Spiers et al., 2004 - Fig. 4). 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 (Spiers et al., 1999).

Solution - precipitation creep is an important deformation mechanism in most rocks in the Earth’s crust (Renard & Dysthe 2003), but is especially rapid in halite. Early reports, theoretical treatments and reviews are given by Durney (1976), Rutter (1976), Sprunt & Nur (1977), Rutter (1983) and Tada & Siever (1996). Recent theoretical treatments of the process are given by Lehner (1990) and Kruzhankov & Stöckhert (1998). In brief, the differences
in chemical potential $\mu$ 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 (Fig 4). 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.

The above processes have been documented in laboratory experiments and in naturally deformed salt from a wide range of settings (Urai et al. 1987, Spiers & Carter 1998, Trimby et al. 2000, Ter Heege et al. 2005, Schléder & Urai (in press); Schleder et al. (subm). The relative importance of each process depends strongly on variables such as temperature, confining pressure, grain size, solid solution impurities and second phase content, and, importantly, on the presence of sufficient water in grain boundaries to enable solution-precipitation phenomena (Fig 5).

Fluid assisted grain boundary migration and solution-precipitation processes do not operate in dry salt, i.e. synthetic samples made of carefully dried material (< 5 ppm water, ter Heege et al., 2005) or natural samples deformed under dilatant conditions which allow intergranular water to evaporate (Peach et al. 2001). The fields in which different deformation mechanisms are dominant can be represented in deformation mechanism maps (Fossum & Fredrich 2002, Ter Heege et al. 2005 - Fig. 6).

Published deformation mechanism maps tend to differ in the mechanisms included, depending on the time scales of interest.

1.2 Rheological behaviour

The rheology of a given crystalline material depends on the dominant deformation mechanism, which in turn depends on the time scale and hence deformation rate of interest. For halite, summaries of behaviour observed in experiments have been published by, for example, Urai et al. (1986), Cristescu & Hunsche (1988), Hunsche & Hampel (1999), Fossum & Fredrich (2002) and Ter Heege et al. (2005).

Considering steady state, non-dilatant deformation, the main classes of equations for the creep strain rate of halite are written

$$
\dot{\varepsilon}_{DC} = A \exp\left(-\frac{Q_{DC}}{RT}\right) (\sigma_1 - \sigma_3)^{m}
$$

for dislocation creep, and

$$
\dot{\varepsilon}_{PS} = B \exp\left(-\frac{Q_{PS}}{RT}\right) \frac{(\sigma_1 - \sigma_3)}{TD^{m}}
$$

for solution-precipitation creep, with the total strain rate being the sum of the two

$$
\dot{\varepsilon} = \dot{\varepsilon}_{DC} + \dot{\varepsilon}_{PS}
$$

In these equations, written in a form appropriate for axially symmetric compressive deformation, $A$ and $B$ are material parameters, $Q_{DC}$ and $Q_{PS}$ repre-
sent (apparent) activation energies for dislocation and pressure solution creep, \( R \) is the gas constant, \( T \) is absolute temperature, \( \sigma_1 \) and \( \sigma_3 \) are the maximum and minimum principle compressive stresses, \( D \) is grain size, and \( n \) and \( m \) are the exponents of stress and grain size respectively. Two important differences between equations (1) and (2) are firstly the dependence of strain rate on stress (\( n = 1 \) for solution-precipitation creep or pressure solution while \( n > 1 \) for DC creep – see Fig. 7) and secondly the dependence of strain rate on grain size. Note that for dislocation creep deformation is grain size independent, while the exponent \( m = 3 \) makes pressure solution creep strongly grain size dependent.

Fig. 7. Steady state strain rate data of fine-grained wet halite samples deforming by solution-precipitation processes, showing a stress exponent \( n \) close to 1. Gray bands represent the theoretical flow law for this process (from Spiers et al., 1990).

At differential stresses below 15-20 MPa and strain rates below \( 10^8 \) s\(^{-1} \), both uniaxial and triaxial experiments on natural and synthetic rocksalt show power law dislocation creep behaviour with a stress exponent \( n \) of 5-6 at the higher stresses and 3.5-4.5 at lower stresses (Wawersik & Zeuch 1986, Carter et al. 1993, Spiers & Carter 1998, Hunsche & Hampel 1999). The apparent activation energy for creep is unusually low, taking values of 50-70 kJmol\(^{-1} \). Intragranular microstructural signatures including wavy deformation band (slip/sub-boundary) structures indicate that cross-slip of screw dislocations may be the rate controlling process at differential stresses (\( \sigma_1 - \sigma_3 \)) above 10-15 MPa (\( n = 5-6 \)), while well formed equiaxed subgrains indicate that climb-controlled recovery becomes dominant at lower stresses (\( n = 3-4 \)). A large amount of published data on natural rocksalt deformed in the laboratory indicates that for a given differential stress, the rate of dislocation creep can vary by approximately three orders of magnitude, caused by differences in concentration of impurities in solid solution, amount and distribution of secondary mineral phases, grain-size, subgrain size, dislocation density and fluids in grain boundaries.

Deformation experiments, performed in the dislocation creep field at confining pressures high enough to suppress dilatancy (> 10-20 MPa), have shown that “wet” samples containing more than 10-20 ppm of water (brine) at grain boundaries undergo rapid dynamic recrystallization by fluid assisted grain boundary migration, alongside dislocation creep. Compared with dry rocksalt samples (<10 ppm water), this recrystallization process leads to a reduction in average dislocation density, an associated reduction of flow strength by 25-50% and power law flow behaviour with an \( n \)-value of about 4.5 (Peach et al., 2001, ter Heege et al., 2005). In addition, during dynamic recrystallization of wet salt (>10-20 ppm), the grain size evolves such that a systematic relation between flow stress and grain size is established, with deformation occurring close to the boundary between the dislocation and solution-precipitation or pressure solution creep fields (ter Heege et al. 2005, Fig. 8).

Fig 8. Dynamically recrystallized grain size versus stress data for synthetic halite samples, superposed on a deformation mechanism map, showing that these samples deform in the transition region between dislocation creep and solution-precipitation creep (from ter Heege et al., 2005).

Solution-precipitation creep has been widely recorded in laboratory experiments on wet, fine grained (< 500 \( \mu \)m) polycrystalline halite at temperatures in the range 20-200°C, leading to quite rapid linear viscous deformation of dense salt (equation 2) and compaction of porous material (e.g. Urai et al. 1986, Spiers et al., 1990, Spiers & Brzesowsky 1993, Renard et al. 2002, 2004). The process has also been observed at individual halite grain contacts under stress (e.g. Gratier 1993, Schütjens & Spiers, 1999, De Meer et al., 2005) and in salt aggregates containing second phases (Renard et al., 2001,
De Meer et al. (2005), Schenk et al. (2006). Possible evidence for pressure solution has also been reported by Berest et al. (2005) in low stress creep experiments on coarse, natural rocksalt, in which much faster rates of deformation were observed than expected by extrapolating conventional dislocation creep laws obtained at higher stresses and strain rates (cf. Fig 5). In addition, rapid long-term deformation of pillars and galleries in potash mines has been attributed to solution-precipitation creep in halite (Campos de Orellana 1998, Lee and de Souza 1998), though such in-situ experiments have not yet been sufficiently documented to allow full quantification of the processes operating (Bekendam and Urai, this volume).

However, 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 typically 1 cm), and it is therefore not usually not found in engineering descriptions of salt rheology (Cristescu & Hunsche 1988, Hunsche & Hampel (1999), Fossum and Fredrich 2002). Nonetheless, predictions made using equation (2) suggest that provided the 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 (see Figures 5 and 6).

As indicated above, at low confining pressures and high deviatoric stresses, flow of rocksalt is accompanied by grain boundary microcracking and rapid permeability increase (Cristescu & Hunsche 1988, Peach & Spiers 1996). The mechanical conditions under which this occurs have been accurately delineated by Cristescu and Hunsche 1988 and Cristescu 1998. While the onset of microcracking has a minor direct effect on creep behaviour, 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 (Peach et al 2001). On the other hand, under conditions where microcracking allows free brine or water vapour access to the interior of a creeping salt sample, then both recrystallization and solution-precipitation creep effects can be strongly enhanced.

Note 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. This is at least partly due to the difficulties of imaging the fine-scale (1-100 nm) structure of wetted grain boundaries during deformation. Approaches applied here include in-situ infrared and electrical resistivity measurements, interference microscopy, and electron microscopy of frozen boundaries using cryo-SEM (Hickman & Evans 1995, Watanabe & Peach 2002, Spiers et al. 2004; De Meer et al. 2005, Schenk et al. 2006).

2 NATURAL LABORATORIES

Studies of rocksalt deformation in nature are essential for reliable extrapolation of laboratory data to describe the flow of salt during slow, human-induced or natural flow, because such studies provide a detailed understanding of the deformation mechanisms and microstructural processes that operate at strain rates well below those accessible in laboratory experiments. In recent years, major advances in this field have been reported, based on developments in microstructural and textural/orientation analysis using electron backscatter diffraction (EBSD), microstructure decoration by gamma-irradiation, Cryo-SEM and other methods. Samples from a wide range subsurface and surface locations have been studied (e.g. Schleder & Urai 2005, in press, Schleder et al., submitted).

In addition there has recently been much progress in measuring the surface displacement field 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 (Weinberger et al., 2006, Urai and Kenis, unpublished data). Insight into the in-situ rheology of salt on the time scale of years has also been gained by simply adjusting the flow laws used in numerical models of mining-related subsidence to obtain a match with surface displacement evolution. Studies of this type, conducted in relation to deep solution mining operations at Barradeel in the Netherlands (2-3 km depth), suggest salt flow behaviour involving dislocation creep (n value of 3-4) combined with a linear viscous flow law comparable to that expected for solution-precipitation creep (Breunese et al. 2006).

![Fig. 9. Subgrain-size versus differential stress data from experimentally deformed halite, providing the basis for measurement of in-situ differential stress in core samples. Solid dots with error bars are the application of this technique to Hengelo Halite (after Schleder & Urai 2005).](image-url)
Microstructural studies of naturally deformed salt show that low temperature dislocation glide and dislocation creep processes, solution-precipitation creep and water-assisted dynamic recrystallization are all of major importance (Urai et al., 1987, Spiers & Carter 1998, Schleder & Urai 2005, in press). The relative importance of these processes varies strongly, as a function of grain size, impurity content, stress path and fluid chemistry. Differential stress, as measured using laboratory-calibrated sub-grain-size piezometry (Fig. 9), is usually less than 2 MPa in rock salt deforming in nature, in agreement with in-situ stress measurements and geologic flow rates (Spiers & Carter 1998, Schleder & Urai 2005). Higher stresses, up to 5 MPa are recorded in the near-surface parts of diapir stems where salt is extruded to the surface (Schleder & Urai in press).

Microstructural studies also show, in agreement with recent experiments, that during fluid-assisted dynamic recrystallization of salt in nature (water content >10 ppm), the grain size adjusts itself so that the material deforms close to the boundary between the dislocation and pressure solution creep fields. Power law flow, as measured in recrystallizing samples (ter Heege et al. 2005), with an n-value of about 4.5 is therefore proposed to be a good representation of this behavior. In samples which are sufficiently fine grained, solution-precipitation creep (equation 2), is found to be dominant both in salt glaciers recrystallized after extrusion to the surface, and in very fine grained primary rock salt in the subsurface (Schleder and Urai 2005, in press). At geologic strain rates, such salt will be orders of magnitude weaker than would be predicted from extrapolation of short-term experiments on coarse-grained halite (see Fig. 5).

The rather high variability of flow strength in layers of halite in nature is in good agreement with the small-scale folding ubiquitously observed in layers of naturally deformed salt. This has not yet been incorporated in numerical models of salt tectonics, which typically assume much more homogeneous material properties and accordingly produce much less heterogeneous strain fields.

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 halite is not completely recrystallized and preserves subgrains. The most likely explanation for this is that below some critical difference in driving force for crossboundary solution-precipitation transfer, surface energy driving forces cause necking of grain boundary fluid films to form isolated fluid inclusions (or possibly some other structural change), thus rendering the boundaries immobile.

In the following, we consider a number of recent examples of how microstructural studies of natural salt can elucidate operative deformation processes and rheology in nature.

2.1 Evidence for dilatancy and fluid flow in halite rocks in the deep subsurface

Intact rocksalt has an extremely low permeability (< 10^-21 m²), which imparts excellent sealing capacity. However, we know from experiments that at sufficiently low effective mean stress, dilatancy and extensional (Mode I) fracture can occur in deforming salt, producing significant permeability (Peach & Spiers, 1996). If this occurs in nature, fluid flow will be possible. Evidence for fluid flow, in the form of saturated brines of different chemical composition, and of hydrocarbons is frequently found in naturally deformed salt (Schleder et al., submitted, Schönherr et al., submitted). Microstructural evidence suggests that both diffuse dilatancy and Mode I fracturing can occur at very low effective stress conditions, i.e. at near-lithostatic fluid pressures and differential stress of a few MPa, in the deep subsurface, followed by crack healing through precipitation from supersaturated solutions.

2.2 Deformation mechanisms in the Hengelo Salt

As an example of a study of deformation mechanisms in natural salt, we briefly review the work done recently by Schleder & Urai (2005) on the bedded salt mined by AKZO at Hengelo in the Netherlands. Deformation of the Hengelo halite in the geologic past has taken place probably during Cretaceous tectonic inversion in the area.

![Fig. 10. Typical micrograph of Hengelo Halite, decorated to show the microstructure, with subgrains (white lines), grain boundaries (dark bands), showing clear evidence for “overgrowth” due to solution-precipitation processes such as pressure solution and and grain boundary migration. Mean grain-size in Hengelo samples is between 5 and 25 mm. Width of image is 7 mm (from Schleder & Urai, 2005).](a)

The microstructure of core samples from the subhorizontal, bedded Main Röt Evaporite Member...
(AKZO well 382, depth interval of 420-460 m) was studied by transmitted and reflected light microscopy of gamma-irradiation decorated samples. Primary microstructures compares favourably with those found in recent ephemeral salt pans. In addition, in all layers the grains are rich in deformation-related substructures such as slip bands and subgrains indicating strains of a few percent (Fig. 10). The study of gamma-irradiation decorated thin sections showed that the main recrystallization mechanism was grain boundary migration (Fig 10). This process removes primary fluid inclusions and produces clear, strain-free (subgrain- and slip band free) new grains. Differential stresses as determined by subgrain size piezometry (Fig 9) were 0.45 – 0.97 MPa. The corresponding deformation mechanisms are inferred to be a combination of dislocation creep, dynamic recrystallization and solution-precipitation processes. Solution-precipitation processes are activated by the small amount of brine present in grain boundaries. Inserting the above differential stress values determined from subgrain size into published flow laws for dislocation creep and for pressure solution (Carter et al 1993, Spiers & Carter 1998) yields transitional strain rates between $10^{-12}$ and $10^{-13}$ s$^{-1}$. The data provide a view of very slow deformation of the Hengelo Halite, up to strains of about 10%, with a significant role played by solution-precipitation processes in controlling rheology.

2.3 Salt glaciers

Microstructural processes in mylonitic shear zones from extrusions of Eocene-Oligocene rocksalts from the Garmser hills and Eyvanekey plateau (central Iran) were recently studied by Schleder & Urai (in press), using transmitted light microscopy of gamma-irradiated thin sections, subgrain size palaeopiezometry of polished and chemically etched samples and texture measurements by EBSD.

The less deformed “protomylonites” found in the stem region of the emerging diapirs? comprise 2-6 mm sized grains, occasionally rich in primary fluid inclusions indicative of their primary non-recrystallized state. Abundant, well-developed subgrains suggest that the protomylonite deformed mainly by dislocation processes. Elongated subgrains at grain edges point to recrystallization by fluid-assisted grain boundary migration. Recrystallized, strain-free grains are common. The material in the highly deformed mylonitic zones found in the salt glaciers is extremely fine-grained (~0.6 mm). Microstructures such as oriented fibrous overgrowths and growth banding (observed in gamma-irradiated sections, Fig. 11) suggest that the principal deformation mechanism was solution-precipitation creep accompanied by grain boundary migration and grain boundary sliding.

Crystal fabrics measured by EBSD show only a weak crystallographic preferred orientation consistent with solution-precipitation accommodated grain boundary sliding. Using published flow laws for this mechanism (Spiers et al., 1990), plus slope-based estimates of the gravitational shear stress driving glacier flow, the strain rate in the fine-grained mylonites was estimated to be about $10^{10}$ s$^{-1}$.

3 DISCUSSION AND CONCLUSIONS

Through integration of all available data, it is now possible to provide a rather complete model of the deformation mechanisms and microstructural evolution of rock salt under a wide range of conditions, including strain rates well below those reached in laboratory experiments (equations 1-3).

At these low rates, microstructural studies of natural salts and extrapolation, long term in-situ and subsidence measurements, and extrapolation of experimental data for fine grained samples indicate that solution-precipitation creep and fluid assisted dynamic recrystallization are important processes, significantly contributing to the total strain rate.

The available data provide a reasonable basis for modeling the mechanical behaviour of salt under geotechnical and natural conditions. However, in many current studies the effects of water-activated grain boundary processes are often neglected, and this omission must lead to errors in prediction of displacement rates, especially over long periods.

Geo-mechanical modeling efforts can thus be significantly improved by making full use of the data available on the effects of water, and some of the discrepancies seen in experimental data on dif-
different salts can probably also be explained in terms of these effects.

Additional improvements can be made by obtaining a more detailed understanding of the mechanism of both transient and steady state dislocation creep, and by improving microphysical models for the effects of solution-precipitation creep, recrystallization and surface energy driven grain boundary and crack healing on flow and transport properties. Further work is also needed on deformation mechanisms in naturally deformed rocksalt from a wider range of geological settings, and in comparing constitutive equations obtained in the laboratory with those obtained from inverting surface displacement data obtained above salt mining sites and salt extrusions.

4 REFERENCES


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