Mineral-water interactions and stress: Pressure solution of halite aggregates

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ABSTRACT: The deformation of synthetic halite aggregates in the presence of circulating fluids was investigated in order to better understand the mechanisms of pressure solution reactions. Halite samples were subjected to differential stresses ranging from 1-5 MPa over periods of time from several weeks to several months. The samples were deformed in the presence of various fluids at 25°C: NaCl-saturated salt brine, H_2O, and ethanol. The axial deformation rates of the samples varied as a function of the differential stress, the initial state of the samples (dry vs. wet), the chemical nature of the circulating fluids, and the total amount of deformation. The relationship between axial strain rate and differential stress was essentially linear. The chemistry of the fluids was of critical importance to the deformation process; in the presence of ethanol, in which halite is only sparingly soluble, deformation rates and total deformation were significantly less than was the case in aqueous fluids.

1 INTRODUCTION

When considering cataclasis, plastic deformation, and pressure solution, the three major mechanisms of rock deformation in the upper crust, only pressure solution is based on a chemical reaction mechanism between a solution and the constituent grains of a rock. Rock deformation at pressures and temperatures ranging up to greenschist facies conditions is thought to take place primarily by solution mass transport, in which pressure solution takes a prominent role (Evans & Kohlstedt, 1995). Pressure solution reactions play an important role in diagenesis reactions associated with sediment burial, compaction and lithification, the development of pressure solution cleavage, and mineral segregation during low grade metamorphism.

The overall pressure solution process is based on dissolution reactions at grain-to-grain boundaries containing an intergranular fluid, aqueous transport of dissolved species, and reprecipitation in intergranular void spaces. The dissolution rate of a given chemical species at a grain-solution interface is a function of the difference in chemical potentials between the surface and the fluid. Such factors as the applied effective stress and the surface free energy (with contributions from elastic, plastic, and interfacial energies) determine the chemical potential of the solid (see details in Paterson, 1973). Measured strain rates, which are dependent on dissolution rates at grain-fluid boundaries under differential stress, are not only dependent on chemical driving forces, but also on intergranular diffusion rates and grain dimensions (for a general review, see Evans & Kohlstedt, 1995; and references therein).

The study of pressure solution reactions applied to the deformation of salt aggregates is of particular interest in understanding mass transfer and flow associated with the geological evolution of salt deposits and diapirs. Salt pressure solution studies also have certain industrial applications in the petroleum and mining fields. In addition, there is also interest in the use of salt mines as potential nuclear and chemical waste repository sites. Applications such as these require precise knowledge of salt deformation behavior in the presence of fluids.

There have been numerous studies published on the deformation of salt by pressure solution (e.g., Urai et al., 1986; Spiers et al., 1990; Spiers & Brzesowsky, 1993; and references therein). Other studies have concentrated on grain indentation and the precise nature of grain-to-grain contacts to better define the operative mechanisms associated with pressure solution (e.g. Tada & Siever, 1986; Hickman & Evans, 1991; Gratier, 1993). The present study emphasizes the deformation behavior of halite in the presence of various fluids over extended
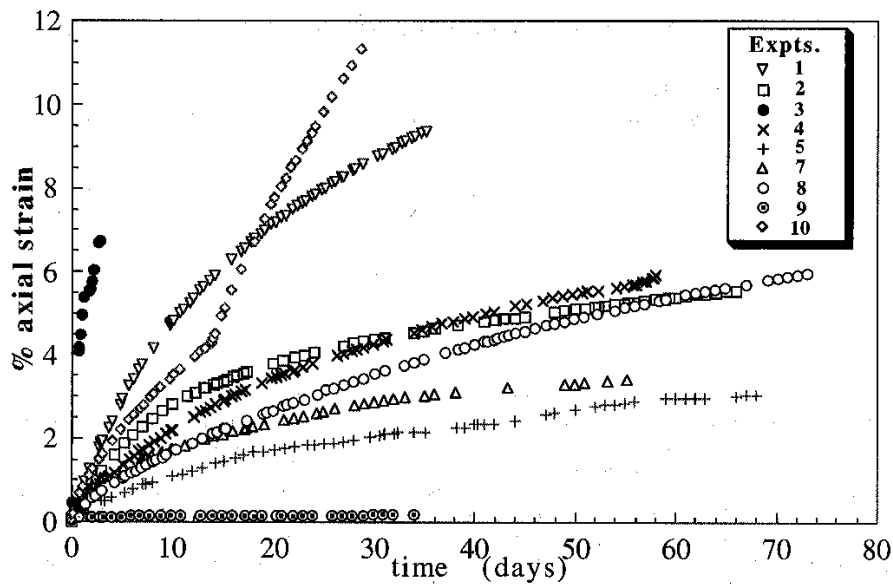
Fig. 1   Measured axial strain as a function of time.   All of the experiments were conducted in NaCl - saturated solutions, with the exception of expts. 8 (water), 9 (ethanol), and 10 (salt brine). The differential stress (σ_1 - σ_3) was on the order of 3 MPa, except for expts. 1 (5 MPa), 2 (1 MPa), and 10 (1, 3 MPa).   Steady-state strain rates were generally reached within 1 month.   The break in the slope of data from expt. 10 is due to an increase in the axial stress during the experiment.
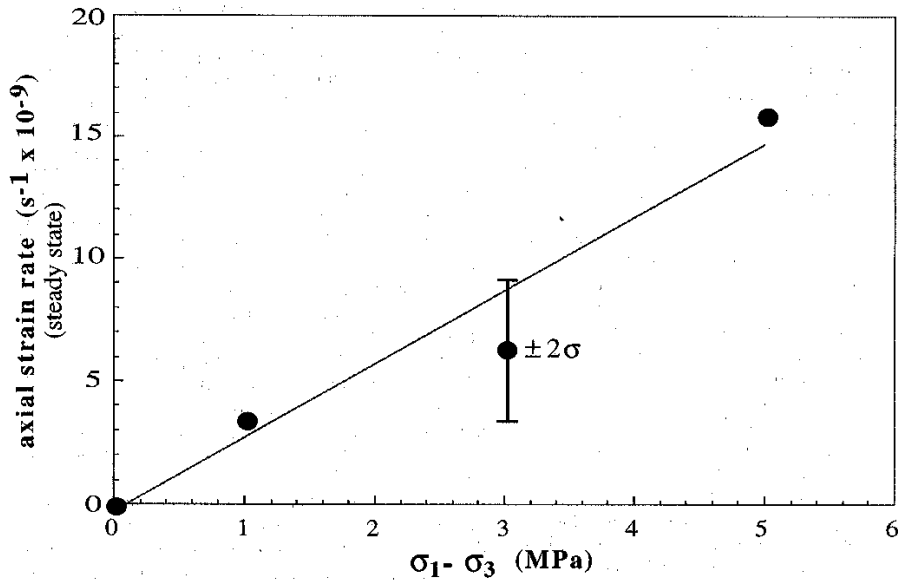

Fig. 2   Axial strain rate as a function of the applied differential stress.   The data are from one set of experiments with approximately the same grain size ($\approx$ 50-60 μm).   The relative linearity of the data is indicative of Newtonian behavior.

778

periods of time, as well as a complete characterization of the time-dependent evolution of permeability and porosity using various techniques.

2 EXPERIMENTAL METHODS

The deformation behavior of salt aggregates in the presence of fluids was studied over periods of time ranging up to 70 days. This ensured that true steady-state deformational flow of salt was achieved, free from the effects of purely mechanical deformation associated with grain diminution, grain sliding, and partial pore collapse. This study used a triaxial compaction cell, where the axial stress (σ_1), the confining stress (σ_3), and the fluid pressure (P_f) were independent. We used an open compaction cell; this enabled us to measure fluid flow and permeability as a function of time during the experiments. The main experimental parameters were the applied differential stress ($\sigma_1 - \sigma_3$) and the chemical composition of the fluids. The chemical nature of the fluids was varied (NaCl-saturated, salt brine, water, ethanol) to test the importance of dissolution rates of halite with respect to the overall rates of measured deformation. Acoustic velocities and x-ray tomographic images were used to characterize the density of the samples before, during, and after deformation in order to monitor the changing permeability and to understand the nature of fluid flow within the samples.

3 RESULTS AND DISCUSSION

3.1. *Axial strain rates and effective stress*

Measured axial strains as a function of time for all of the experiments are shown in Fig. 1. Steady-state axial strain rates ranged up to about 1.6×10^{-8} s^{-1}. A plot of steady-state axial strain rates vs. effective axial stress (i.e. axial stress (σ_1) - confining stress (σ_3)) reveals an approximate linear relationship (Fig. 2). This is indicative of a Newtonian rheology for these particular stress conditions. The definition for the effective axial stress is normally ($\sigma_1 - P_f$). However, based on SEM images, we have assumed that the intergranular fluid films have the following approximate pressures:

$P_{f1} \approx \sigma_1$ (fluid film $\perp \sigma_1$); $P_{f3} \approx \sigma_3$ (fluid film $\perp \sigma_3$). The effect of fluids in microchannels, where $P \approx P_f$, is probably of minor importance to the overall, steady-state deformation process.

3.2. *Chemistry of the fluids*

The importance of dissolution reactions with respect to the overall deformation process is well illustrated in these experiments. The use of fluids that were good solvents (polar solvents with elevated dielectric constants: NaCl-saturated fluids, salt brine, and H_2O) vs. ethanol, which is a non-solvent (non-polar, low dielectric constant), showed that the chemical nature of the fluid plays a critical role in determining the rate of deformation. The recorded total deformation, as well as the deformation rate, of the salt in the presence of ethanol was only a fraction of that recorded for samples deformed in the presence of aqueous solutions (Fig. 2). This can be explained by the fact that the salt-ethanol reaction has a smaller chemical affinity than the corresponding affinities of salt-aqueous solution reactions. At conditions close to equilibrium, a smaller chemical affinity translates to a slower rate of dissolution. This is proof that chemical reactions play a dominant role in wet-salt deformation under these differential stress conditions.

3.3 *Permeability*

The permeability of the samples continually evolved during the course of the experiments. This is evidence that the permeability of a rock or an aggregate is not an intrinsic, fixed property, but rather continually evolves during deformation due to reactions between permeating fluids and mineral grains. Plots of cumulative mass of fluid collected as a function of time revealed, in almost all cases, sigmoidal behavior. This type of behavior is the result of samples that were initially impermeable to fluid flow, followed by a rapid increase in permeability. Fluid flow was apparently channelized in the samples, as shown by x-ray tomography images of samples before and after deformation. Eventually, however, most of the samples became impermeable to further fluid flow. Porosimetry measurements before and after each experiment revealed that pore dimensions decreased, due mostly to a destruction of pores with diameters between 1-10μm. The decrease in pore volumes is in accord with the samples becoming impermeable to fluid flow. The net transfer of material from grain-to-grain contacts subject to stress to pore voids is the principal reason for the decrease in permeability. Acoustic measurements of samples during the course of experiments showed a significant increase in velocities, this being evidence for an increase in the overall density of the samples, and concomitantly, a decrease in void spaces.

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