Plastic behavior of halite single-crystals at different temperatures and strain rates: New insights from in-situ experiments and full field measures

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A B S T R A C T

Rock salt, or halite, is a rock forming mineral with numerous applications. In the laboratory, natural or synthetized, it is an analog material for understanding of the physical properties of rocks and ductile deformation mechanisms of other crystalline classes of materials. It is thus surprising that very few studies have been devoted to the plasticity of NaCl single crystals with well documented experimental conditions. In the present study, we deform at several temperatures NaCl single crystals of selected initial orientations by uniaxial compression. The new contribution of this work resides in the multi-scale investigations, with the combination of mechanical tests and observations by optical microscopy and scanning electron microscopy. The use of digital image correlation (DIC) at these different scales allows the measure of displacements and the computation of global or full-field strains as well as very local ones during the loading history. The optical set-up provides images of two orthogonal surfaces of the samples, so that the slip lines and their orientations can be observed on two surfaces resulting in a better identification of the active slip systems. We show that for NaCl, temperature is a key parameter for the activation of slip systems. At high temperature ($0.6T_f$), we present the variation of the yield stress with strain rates, in the range 2×10^{-6} to $2 \times 10^{-5}\,\text{s}^{-1}$. The initial critical shear stress values and the hardening behavior that we have experimentally determined are essential for the implementation of crystal plasticity finite element models that will simulate the plastic behavior of polycrystalline halite.

1. Introduction

Rock salt caverns, whether natural or man-made, have attracted a lot of attention for a long time as geological storage of waste or hydrocarbons. More recently, the Compressed Air Energy Storage (CAES) projects [15] consider salt caverns for short term or cyclic underground storage of energy in the form of compressed air, which motivates a renewed interest in the plastic behavior of salt. Rock salt is essentially constituted of halite single crystals (NaCl) with very little remaining porosity or residual water, and a strong cohesion. It may thus be idealized as a polycrystalline solid. As such, the rheological behavior of natural and synthetic halite has been largely investigated during the last century ([8,37,10,21,33,30]). It has been shown that rock salt deforms by several mechanisms such as intragranular dislocation glide ([17,9,4,3,8,21]), pressure-solution ([43,35,42,18]) and grain boundary sliding ([6]).

Among these mechanisms, the first one, crystal plasticity, is a key mechanism for the ranges of strain rates and temperatures that we shall consider here, respectively from 10^{-6} to 10^{-3}s^{-1} and from room temperature to 400 °C which corresponds to about $0.6T_f$ where T_f is the

melting temperature of halite, 1078 K. In the case of the ionic cubic crystal NaCl, dislocation glide is known to occur along the glide directions <011> on three families of slip planes, respectively dodecahedral {011}, octahedral {111} and cubic {001}. The conditions for the activation of the three families of slip systems in halite, i.e. the knowledge of the initial critical resolved shear stresses (CRSS) and their evolutions with strain, are poorly documented. They have mostly been studied in extension and under high confining pressure ([9]), but not in uniaxial compression. Moreover, the identification of the effectively activated slip systems has always been indirectly inferred from post-mortem investigations of relict microstructures either by etching or by birefringence, which meant that the history of activation along the deformation path remained unknown. The motivation of the experimental study which we present in the following is to revisit the evaluation of CRSS on the different families of glide systems since these data are essential to better understand the mechanical behavior of rock salt and to provide reliable estimates for parameters which are needed by numerical models such as CPFEM (Crystal Plasticity Finite Elements Models).

Mechanical testing is paired with observations and full field

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recording, so that the combined analyses of the mechanical data (force and displacements), the direct observations (slip lines) and the digital image correlations (displacements and strain) allow the identification of the local mechanisms of deformation.

To better constrain the history and conditions of activation of the different slip systems, we investigate the crystal plasticity of pure NaCl single crystals deformed in uniaxial compression at different temperatures and strain rates with in-situ optical microscopy (OM) and scanning electron microscopy (SEM) observations. Four crystal orientations have been selected to potentially activate glide on each family of slip systems. Crystal slip related microstructures have been analyzed during the deformation process by full strain field measurements based on digital image correlation (DIC) techniques ([2]). This micro mechanical approach allows the precise determination of the active deformation mechanisms, their history and the heterogeneity of the strain field in relation with sample and loading geometry and microstructure. The computation of strain by DIC applied on the whole sample allows retrieving actual loading curves without the effects of compliance and adjustments of the loading frame and column. When several equivalent slip systems are available, the actual number of activated ones is directly imaged, which hence provides the opportunity of investigating system interactions. Our results show and discuss the effects of temperature, strain rate and crystal orientation on the mechanisms of plastic deformation and CRSS magnitudes.

2. Material and experimental procedure

2.1. Plasticity of NaCl

Salt is a cubic ionic crystal, its space group is $Fm\bar{3}m$. It has been investigated for its plastic anisotropy with other rocksalt-type or halide crystals ([23,39,4]). Some studies are phenomenological, but most are at the dislocation level ([40,7,3]). Dislocations in ionic crystals have been extensively studied: See the textbook by Hirth and Lothe [27], a review by Castaing [11] and more specifically for NaCl, Haasen [24], Hesse [25] or Hesse and Matucha [26].

The most comprehensive study on the plastic deformation of halite single crystals accounting for temperature and strain rate effects is due to Carter and Heard [9]. The authors emphasize that very few investigations on the plasticity of NaCl single crystals have been reported and we can still make the same comment today. It has been established that the main slip planes for NaCl are the {110} planes, also called dodecahedral planes, and the slip directions are the <110> directions. There are six such planes, each containing a glide direction (see Table 1). One may show (see for instance [13]) that these provide only two independent systems. The dodecahedral systems are thus not sufficient to accommodate an arbitrary plastic strain (see for instance Chin [12]), since five independent slip systems are theoretically required to accommodate any arbitrary plastic strain at the local (i.e. grain) scale and this rule should apply for single crystals. However for crystals lacking sufficient independent systems, such as HCP crystals, Kocks and Westlake [29], later quoted by Hutchinson [28] argue that four systems are sufficient, "since a missing DOF in each grain may be accommodated by inelastic deformation in the neighboring grains". This is also supported by later investigations on olivine reporting that " self-consistent estimations and full field computations have shown that four independent systems are sufficient to ensure that the polycrystal deforms as a whole" ([19]). These comments are made for polycrystals and do not readily apply to single crystals. The need to accommodate an imposed stress state with too few systems may lead to the development of subgrains or brittle behavior. Slip on non-compact planes can provide the additional plastic DOF. It has been reported for fcc metals at high temperatures [14,31]. More specifically for halite, Carter and Heard [9] have observed slip on {100} and {111} planes (along <110> directions) but at higher stresses, with a strength highly temperature and rate dependent. By including the six {100} < 011> systems (the

Table 1
Naming convention for the slip systems of NaCl. The six slip directions are numbered 1–6. The four octahedral planes are labeled A to D, following the convention used for fcc materials.

Systems	Plane	Direction
Dodecahedral		
I2	(011)	[0$\bar{1}$1]
II1	(0$\bar{1}$1)	[011]
III4	(101)	[$\bar{1}$01]
IV3	($\bar{1}$01)	[101]
V 6	(110)	[$\bar{1}$10]
VI5	($\bar{1}$10)	[110]
Octahedral		
A2	($\bar{1}$11)	[0$\bar{1}$1]
A3	($\bar{1}$11)	[101]
A6	($\bar{1}$11)	[$\bar{1}$10]
B2	(111)	[0$\bar{1}$1]
B4	(111)	[$\bar{1}$10]
B5	(111)	[110]
C1	($\bar{1}\bar{1}$1)	[011]
C3	($\bar{1}\bar{1}$1)	[101]
C5	($\bar{1}\bar{1}$1)	[110]
D1	(1$\bar{1}$1)	[011]
D4	(1$\bar{1}$1)	[$\bar{1}$01]
D6	(1$\bar{1}$1)	[$\bar{1}$10]
Cubic		
X1	(100)	[011]
X2	(100)	[0$\bar{1}$1]
Y3	(010)	[101]
Y4	(010)	[$\bar{1}$01]
Z5	(001)	[110]
Z6	(001)	[$\bar{1}$10]

cubic systems), one adds three independent systems. The theoretical number of five independent systems is then reached. The twelve octahedral systems, {111} < $\bar{1}$10>, have also been identified as potential systems, but Carter and Heard [9] have not found a crystal orientation which deforms only on these systems. They have evidenced only combined glide on {111} < $\bar{1}$10> and {110} < $\bar{1}$10>.

It is generally assumed that the potential activation of a given slip system s depends only on the magnitude of the projection of the applied stress onto the system plane and in the direction of glide. This effective component is called the resolved shear stress (RSS) or Schmid stress τ^s. In the case of uniaxial tension (or compression) τ^s is related to the applied stress σ by:

$$\tau^s = \sigma \cos\chi \cos\theta \tag{1}$$

where χ and θ are the angles between the uniaxial load direction and the normal and the glide direction of the slip system, respectively. The geometric factor, $\cos\chi \cos\theta$, also called Schmid factor (Sf) ([36] or [27], pp. 269–275), ranges from 0 for a non-activatable slip system ($\tau^s = 0$) to 0.5 for a slip system best oriented for its activation. This notion is easily extended to any stress state characterized by a stress tensor σ. For each slip system s defined by its unit slip plane normal $\mathbf{n}^s$ and unit vector $\mathbf{m}^s$ along the slip direction, one forms a symmetric Schmid tensor

$$\mathbf{S^s} = \frac{1}{2}(\mathbf{m^s} \otimes \mathbf{n^s} + \mathbf{n^s} \otimes \mathbf{m^s}). \tag{2}$$

The Schmid stress then becomes, $\tau^s = S_{ij}^s \sigma_{ij}$ and the activation of the considered slip system occurs when τ^s reaches a critical value (CRSS) τ_c^s, the latter being temperature, history and strain rate dependent.

For geometrical reasons, the values of Sf are the same for each pair of dodecahedral systems which exchange slip direction and slip plane. For instance, the Schmid factors on the two systems (110)[$\bar{1}$10] and