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Using neutron diffraction to examine the onset of mechanical twinning in calcite rocks

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ABSTRACT

Experimental calibration of the calcite twin piezometer is complicated by the difficulty of establishing the stresses at which the twins observed in the final deformation microstructures actually formed. In principle, this difficulty may be circumvented if the deformation experiments are performed in a polychromatic neutron beam-line because this allows the elastic strain (and hence stress) in differently oriented grains to be simultaneously monitored from diffraction patterns collected as the experiment is proceeding. To test this idea small strain (<0.3%), uniaxial compression experiments have been performed on Carrara marble (grain size 150 μm) and Solnhofen limestone (5 μm) at temperatures of 20°–600 °C using the ENGIN-X instrument at the ISIS neutron facility, UK. At the lowest temperatures (25 °C Carrara; 200 °C Solnhofen) the deformation response was purely elastic up to the greatest stresses applied (60 MPa Carrara; 175 MPa Solnhofen). The sign of the calcite elastic stiffness component c_{14} is confirmed to be positive when the obverse setting of the calcite rhombohedral lattice in hexagonal axes is used. In the Carrara marble samples deformed at higher temperatures, elastic twinning was initiated at small stresses (<15 MPa) in grains oriented such that the Schmid factor for twinning was positive on more than one *e*-twin system. At greater stresses (65 MPa at 200 °C decreasing to 41 MPa at 500 °C) there was an abrupt onset of permanent twinning in grains with large Schmid factors for twinning on any one *e*-twin system. No twinning was observed in the Solnhofen limestone samples deformed at 200° or 400 °C at applied stresses of <180 MPa. These results highlight the potential of this approach for detecting the onset of twinning and provide, through experiments on samples with different microstructures, a strategy for systematically investigating the effects of microstructural variables on crystallographically-controlled inelastic processes.

1. Introduction

The orientation and abundance of mechanical *e*-twins in naturally deformed calcite-bearing rocks are widely used to evaluate palaeostresses and strains (for several case studies, see Lacombe, 2010). From a consideration of the crystallography of twinning on calcite *e*-planes (e.g. Handin and Griggs, 1951), techniques have been developed that allow calcite twinning microstructures to be used to infer the orientations of the principal stresses (Turner, 1953; Dietrich and Song, 1984; Shelley, 1992), to apply constraints on the relative magnitude of these stresses (Spang, 1972; Pfiffner and Burkhard, 1987), and to obtain the orientations and magnitudes of the principal strains (Groshong, 1972, 1974; Groshong et al., 1984). The magnitude of the difference between the greatest and the least principal stresses (the differential stress) may be evaluated from experimental calibrations of twin abundance as a function of differential stress (Rowe and Rutter, 1990). Alternatively, by assuming that mechanical twinning occurs once some critical shear stress resolved on the twin plane along the twin displacement vector has been attained, the magnitude of the differential stress experienced by a calcite rock with randomly oriented grains may be estimated from the relative number of twinned and untwinned grains (Jamison and Spang, 1976). When combined with twin orientation measurements, this last approach may be re-developed to evaluate the complete deviatoric stress tensor (e.g. Laurent et al., 1981, 2000; Lacombe and Laurent, 1992; Nemcock et al., 1999;

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Lacombe, 2007; Gagala, 2009; Yamaji, 2015a, 2015b).

Each of these techniques involves several approximations which restrict their applicability and introduce significant uncertainties into the results (for informative commentaries, see Burkhard, 1993; Ferrill, 1998; Rybacki et al., 2013). Among these we draw particular attention to the following. Firstly, the value of the critical resolved shear stress for twinning, and its dependence on deformation variables such as temperature, strain-rate and strain, is not tightly constrained by experiments. There is even doubt about whether or not a critical resolved shear stress for mechanical twinning exists at all (Christian and Mahajan, 1995; De Bresser and Spiers, 1997). Secondly, mechanical twinning is widely recognized to be sensitive to microstructural variables such as grain size and crystallographic preferred orientation (e.g. Rowe and Rutter, 1990; Meyers et al., 2001; Beyerlein et al., 2010). However, the nature of this effect remains poorly characterized. Thirdly, the application of several of these techniques to polycrystalline rocks and/or the interpretation of the findings usually involves an approximation that stress is homogeneous at the grain scale so that the magnitude and orientation of the grain scale stresses can be taken to be the same as those applied to the rock as a whole. Yet in a polycrystal comprising differently oriented grains of a mechanically anisotropic material, this cannot be precisely true. These matters all touch upon the strongly felt intuition that whether or not a grain twins is likely to be sensitive to factors that influence the local stress state within the microstructure, and is not simply determined by the macroscopically applied stress.

Nevertheless, despite these limitations, it is undeniable that analyses of calcite twinning have made a significant contribution to regional reconstructions of tectonic stresses and strains, and are able to place informative constraints on the kinematic evolution of sedimentary basins (e.g. the case studies described by Lacombe, 2010). This, together with the potential that exists for extending these twin-based techniques to other minerals such as dolomite, pyroxene and quartz (Carter and Raleigh, 1969; Tullis, 1980; Wenk et al., 2006), provides a strong motivation for seeking the kind of improved understanding of the factors influencing mechanical twinning that also increases confidence in twin-based palaeostress and strain estimates.

Attempts to use rock deformation experiments to reduce the uncertainty in twin-based palaeostress and strain techniques are hampered by the fact that it is difficult to monitor the onset and progress of mechanical twinning within polycrystalline samples during the experiments. The extent of twinning is instead obtained by examining the microstructures of the deformed samples after the experiment. This only provides information about the end-result of the deformation and does not indicate when during the deformation the twinning occurred. Nor does it provide information about precursory elastic twinning, a phenomenon that is well-documented in calcite single crystals (e.g. Klassen-Neklyudova, 1964; Kaga and Gilman, 1969; Boyko et al., 1994; Clayton and Knap, 2011).

The penetrating nature of neutrons offers a potential solution to these difficulties. By performing the deformation experiment within a polychromatic beam of neutrons and collecting diffraction data (effectively powder diffraction patterns) at different applied loads, changes in lattice spacing in several sub-sets of differently oriented grains may be simultaneously monitored as a function of the applied load. These changes in lattice spacing may be converted into the elastic strains. By comparing how these elastic strains change with increasing applied load in each of these sub-sets of grains, the onset and progress of crystallographically-controlled processes such as twinning in grains that are favourably oriented for them to occur may be tracked. In principle, polychromatic synchrotron X-rays may be used in the same way (e.g. Withers, 2013), but whereas the small gauge volumes accessed by synchrotron X-rays are ideal for high spatial resolution (sub-mm to micron) strain mapping (e.g. Chen et al., 2011), the larger gauge volumes accessed by neutrons provide a more convenient route to evaluating strain within a volume of sample that is large compared with the grain size.

In this study we report the results of some neutron diffraction experiments of this kind performed on two calcite rocks with different grain sizes. We show that the onset of mechanical twinning at small strains (<0.3%) in calcite can be monitored in this way.

2. Background to the adopted experimental strategy

Being able to monitor the onset and progress of mechanical twinning during a deformation experiment provides a method for establishing whether or not a macroscopic critical resolved shear stress can be defined, and if it does, then also the magnitude and sensitivity of that stress to deformation variables such as temperature and strain-rate. By performing experiments on samples with different microstructures, the influence of microstructural variables may be quantified.

2.1. Critical resolved shear stress for twinning

Experiments on a wide range of engineering materials show that mechanical twins nucleate at an externally applied stress that is much smaller in magnitude than theoretically predicted, and that when they do nucleate, they do so in regions of highly localized strain, that is, at localized stress concentrations on grain boundaries or other lattice defects (Christian and Mahajan, 1995). Two perspectives on twin nucleation then logically follow. Firstly, by analogy with dislocation glide, one might envisage that the magnitude of the local stress required to nucleate twins is approximately the same throughout the sample but also note that within the sample there is a wide range in the magnitude of local stress concentrations. Twinning occurs as the externally applied stress is increased and increasing numbers of the stress concentrators attain the critical activation stress. Alternatively, one might envisage that there is an array of potential twin nucleation sites, each with a different critical activation stress that reflects the specific local lattice structure at that site. In this case, as the externally applied stress is increased, more of these sites activate. In the former perspective it makes sense to refer to a critical resolved shear stress for twinning; in the latter perspective it does not. Most commentaries on twinning favour the latter perspective (e.g. Reed-Hill and Abbaschian, 1992; Christian and Mahajan, 1995; Tomé et al., 2011), and accordingly probabilistic models of twin nucleation have been developed that accommodate local lattice structure variability and a range of activation stresses (e.g. Beyerlein and Tomé, 2010; Beyerlein et al., 2011; Niezgoda et al., 2013). Nevertheless, if widespread twin nucleation is initiated over a small stress interval, it may be possible to define an apparent critical resolved shear stress that could be used in palaeostress analysis. A mechanism for rapid bursts of twin nucleation is provided by the observation that a twin propagating across one grain may nucleate a twin in the adjacent grain when the local stress concentration associated with its tip reaches the grain boundary, that is, twin nucleation may be autocatalytic (Ecob and Ralph, 1983).

In palaeostress analyses based on calcite twinning, a critical resolved shear stress of 10 MPa is widely used (Lacombe, 2010). This value is consistent with the stresses obtained from the twin microstructures of coarse-grained calcite marbles that have been experimentally deformed in axial compression to known differential stress (Lacombe and Laurent, 1996; Laurent et al., 2000). It is also consistent with the results of axial compression and axial