ABSTRACT
Field observations and geophysical data reveal a causal link between brittle seismic failure and eclogitization of the lower continental crust. We present results from experimental deformation of plagioclase-rich samples at eclogite-facies conditions and quantify the link between rock rheology and the kinetics of the eclogitization reactions. The deformation was ductile both in the absence of reaction and when the progress of eclogitization was fast compared to the imposed strain rate. However, when the reaction rate was relatively slow, the breakdown of plagioclase into nanocrystalline reaction products triggered embrittlement, highlighted by a high acoustic emission activity. Fluid-induced plagioclase breakdown under eclogite-facies conditions is an exothermic reaction accompanied by a negative change in solid volume. This is similar to other mineral transformations that are known to trigger transformational faulting. We demonstrate that mineral reactions lead to brittle deformation in situations where reaction rates are slow compared to the deformation rate.

INTRODUCTION
The mechanisms leading to brittle seismic failure in Earth’s lower continental crust are still poorly understood. On the one hand, experimental studies reveal that plastic deformation should be dominant in plagioclase-rich rocks under granulite-facies conditions (<1 GPa and 973 K; Tullis and Yund, 1992) in the presence of water (Bürgmann and Dresen, 2008). On the other hand, field studies on lower-crustal rocks in Norway and geophysical data from the Himalaya–Tibetan Plateau reveal a strong coupling among seismicity, fluid infiltration, and eclogitization of the metastable lower continental crust (Austrheim and Boundy, 1994; Jackson et al., 2004; Lund et al., 2004; Hetényi et al., 2007; John et al., 2009; Jamtveit et al., 2018).

In this study, we used acoustic emissions to highlight the change in rheological behavior of granulite from dominantly plastic to mostly brittle while undergoing metamorphic transformation at eclogite-facies conditions in the presence of a hydrous fluid.

MATERIALS AND METHODS
A granulite from Holsnøy, southwestern Norway, was used as sample material. It mostly consisted of plagioclase (≈90 vol%) and minor amounts of epidote, sometimes intergrown with alkali feldspar, spinel, corundum, clinopyroxene, amphibole, biotite, kyanite, garnet, and magnetite, which together made up the remaining 10 vol% (Fig. DR1 in the GSA Data Repository). The sample was crushed and sieved to a grain size of <38 μm. This powder was washed using distilled water to remove the dust fraction, dried in an oven, and then left at ambient air humidity conditions. The same starting material was used for all experimental runs. The deformation experiments were conducted using the D-DIA apparatus (Wang et al., 2003), a deformation multi-anvil press, at the GeoSoilEnviro-CARS (GSECARS [Consortium for Advanced Radiation Sources]) beamline 13 BM-D of the Advanced Photon Source, Argonne National Laboratory, Illinois, USA. The D-DIA is equipped with an acoustic emission setup. The experimental setup has been explained in detail by Gasc et al. (2011) and Schubnel et al. (2013). Synchrotron X-ray radiographs as well as powder diffraction patterns were acquired every 5 min, respectively, to monitor the strain and the differential stress of the sample during the experimental run, at in situ pressure-temperature (P-T) conditions. Experimental conditions are summarized in the table in Figure 1. Details on the stress and strain calculation as well as on the analytical methods are given in the Data Repository.

RESULTS
Based on a comparison of the overall stress-strain evolution together with the records of acoustic emissions during the four experimental runs, three different mechanical behaviors can be distinguished. Differential stress continuously increased during the deformation of sample NG_2.5_1023 (constant T), which exhibited the highest strength and the least detected number of acoustic emissions (Figs. DR2A and DR2C). We refer to this behavior as strong and ductile. For sample NG_2.5_1173 (constant T),...
the differential stress first increased to a peak stress of ~1.8 GPa at ~15% axial strain and then remained approximately constant until the end of deformation. In total, 794 acoustic emissions were recorded during deformation, and so this sample was in the brittle regime. Samples NG_2.5_1225 and NG_3.1225 both exhibited stress increase to ~2.3 GPa, followed by a large stress drop at ~14% axial strain. Their stress drop correlated with an imposed increase in temperature and did not coincide with the onset of acoustic emissions (dashed vertical lines in Fig. DR2).

These samples showed a large variation in the total number of acoustic emissions: 82 for sample NG_2.5_1225 and only 9 for NG_3.1225. Sample NG_2.5_1225 clearly showed ample evidence for brittle deformation, while sample NG_3.1225 was weak and ductile (Figs. DR2A and DR2C). Figure 1 shows the calculated evolution in mean stress, \(\sigma_m = (\sigma_1 + 2\sigma_3)/3\), with temperature. All samples were deformed under eclogite-facies conditions outside the plagioclase stability field (Fig. 1; De Capitani and Petrakakis, 2010). The samples deformed furthest from the plagioclase stability field produced the fewest acoustic emissions (Fig. 1).

Two main microstructural features appeared in the samples after deformation. Backscattered electron (BSE) images show dark areas in the plagioclase matrix indicative of a local difference in chemical composition (Fig. 2A). Another feature is the appearance of bands that are brighter than the adjacent plagioclase matrix in BSE mode (Fig. 3G). Both features, the dark areas and the bright bands, are very small (often <1 µm) and seem to be composed of different phases (Fig. 2B; Fig. DR3D). This makes precise measurements very difficult. However, energy-dispersive X-ray spectroscopy (EDS) analyses and semiquantitative element distribution maps were acquired with the scanning electron microscope (SEM) to compare the chemical compositions between the plagioclase matrix and these features. The dark zones were enriched in Na and depleted in Ca relative to the plagioclase matrix, suggesting a chemical composition close to albite or jadeite (Fig. 2A). Those zones were prominent in sample NG_2.5_1173 and mostly contained nanocrystalline grains (Figs. DR3D and DR4B). The bright crystals, which were most frequent in NG_3.1225, were mainly composed of elongated zoisite needles together with kyanite, quartz, and albite/jadeite (Figs. 2B and 2C). The location and abundance of both features within each sample were spatially correlated, but their respective amounts differed between samples (Fig. 3; Fig. DR3). The dark zones were prominent in sample NG_2.5_1173 (Fig. 2A) and rare in sample NG_3.1225, which mostly showed bright band-forming crystals (Fig. 2B). Sample NG_2.5_1225 showed both features in equal amounts (Figs. 3E and 3F).

**DISCUSSION AND CONCLUSION**

Both the bright bands and the dark areas reflect the onset of eclogitization by plagioclase breakdown in the presence of a hydrous fluid according to the reaction: 

\[\text{plagioclase} + \text{fluid} \leftrightarrow \text{zoisite} + \text{kyanite} + \text{quartz} \pm (\text{NaSiCa}_x\text{Al}_y\text{plagioclase} \pm \text{jadeite})\]

modified after Wayte et al., 1989). First, zoisite nucleates and grows, followed by the other product phases, which start to nucleate and grow at a more advanced plagioclase-transformation stage of ~10 vol% (Wayte et al., 1989). Reaction progress was estimated based on image analysis of the area fraction of the reaction products. Since kyanite and quartz were only found in sample NG_3.1225, this sample showed the highest degree of eclogitization, i.e., the highest reacted plagioclase volume, estimated to be ~10 vol% (Wayte et al., 1989). With respect to NG_3.1225, the other samples showed lower reacted plagioclase volumes of ~5 vol% for NG_2.5_1225, ~2.5 vol% for NG_2.5_1173, and less than 1 vol% for NG_2.5_1023 (Fig. 3). It is likely that water from ambient air humidity was adsorbed onto the grain surfaces during

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**Figure 1.** Mean stress-temperature curves on pressure-temperature (P-T) phase diagram showing stability of plagioclase and experimental conditions (table), where \(P_c\) = confining pressure; \(T\) = temperature; \(\varepsilon\) = finite strain; \(\dot{\varepsilon}\) = strain rate; \(X\) = reacted plagioclase volume. AEs — acoustic emissions.

**Figure 2.** A: Backscattered electron (BSE) image with superimposed composite energy-dispersive X-ray spectroscopy (EDS) elemental map showing Ca and Na distribution. B: Composite EDS elemental map of area marked in Figure DR3H (see footnote 1). Bright bands in microstructure are composed of zoisite + kyanite + quartz ± albite ± jadeite. C: Transmission electron microscope (TEM) image, taken in bright field mode, showing cross-sectional view of selected bright band composed of elongated crystals with borders of isometrically shaped crystals. Location of this image is shown in Figure DR5 in the Data Repository. Maximum stress is vertical on images A and B.
sample preparation, thus providing a possible fluid source to produce the hydrous phase zoisite. Another possible fluid source could be the plagioclase itself, since it can contain minor amounts of water in its crystal structure. Although small amounts of fluid are available during deformation, the overall reaction kinetics are expected to be sluggish, because they involve two successive processes. First, plagioclase is highly metastable (Fig. 1), and its breakdown will occur very rapidly as soon as it comes into contact with even low amounts of water. Second, the nucleation and growth of the reaction products are expected to be slower, because they involve chemical transport over longer distances. It is therefore reasonable to assume that the net rate of eclogitization is controlled by the nucleation and growth of the reaction products.

In a reacting rock, strain can be accommodated by transformation-induced volume changes. An imposed strain rate, $\dot{\varepsilon}$, can be expressed as the sum of a transformational strain rate, $\dot{\varepsilon}_t$, and a residual strain rate, $\dot{\varepsilon}_r$, with $\dot{\varepsilon} = \dot{\varepsilon}_t + \dot{\varepsilon}_r$. In this study, three scenarios can be deduced (Fig. 3): (1) almost no reaction (NG_2.5_1023), (2) slow net eclogitization (NG_2.5_1225 and NG_2.5_1173), and (3) fast net eclogitization (NG_3_1225). If no or little deformation is accommodated by transformation reactions, the granulite is strong and ductile, and the microstructure of the sample shows the net eclogitization rate is slow, the granulite is strong and ductile (Austrheim, 1990). Although small amounts of fluid are available during deformation, the overall reaction kinetics depend on metastability (distance to equilibrium), plagioclase breakdown proceeded faster in sample NG_3_1225 than in sample NG_2.5_1225, deformed at a lower confining pressure. In conclusion, embrittlement is triggered by plagioclase breakdown, but as eclogite formation progresses, the overall rheological behavior of the sample becomes controlled by the soft eclogite. This has also been observed on Holsnøy, where unreacted and undeformed strong granulite blocks are embedded within a matrix of weak and deformed eclogite (Austrheim, 1990).

Eclogitization in nature, e.g., on Holsnøy, took place at $\dot{\varepsilon}$ and $T$ values significantly lower than in our experiments. Assuming that the observed embrittlement is controlled solely by the rate of plagioclase breakdown relative to the imposed $\dot{\varepsilon}$, we can scale our laboratory conditions to those expected for natural systems. As a first-order approximation, $\dot{\varepsilon}_e$ can be expressed as:

$$\dot{\varepsilon}_e = \frac{\Delta V/V \times (dV/ds)^3 \times k_e \times \exp \left( -\frac{E_a}{RT} \right)}.$$  (1)

This equation combines the volume change of the plagioclase breakdown reaction $\Delta V/V$ and the grain-size dependence on transformation progress, where $d_s$ is the initial grain size, and $d$ is the grain size after a certain amount of reaction with an Arrhenius-type $T$-dependence of the transformation rate ($k_e \times \exp (-E_a/RT)$). The parameters $k_e$, $E_a$, and $R$ are the reaction rate constant, the activation energy, and the gas constant, respectively (see the Data Repository for further details on the calculation of $E_a$). Assuming that the reaction extent required to cause embrittlement in the laboratory is similar to that in nature, the laboratory transformational strain rate, $\dot{\varepsilon}_{t,lab}$, and the natural transformational strain rates, $\dot{\varepsilon}_{t,nat}$, are related by:

$$\dot{\varepsilon}_{t,lab}/\dot{\varepsilon}_{t,nat} = (d_{s,nat}/d_{s,lab})^3 \times \exp \left( E_a/R \times (1/T_{nat} - 1/T_{lab}) \right).$$  (2)

where $T_{nat}$ is the eclogitization temperature on Holsnøy (~973 K; Raimbourg et al., 2005), and $T_{lab}$ is the temperature under which embrittlement is observed in our experiments (~1173 K). The initial natural grain size $d_{s,nat}$ is $\sim 1-3 \times 10^{-3} m$, and the initial laboratory grain size $d_{s,lab}$ is $\sim 1-38 \times 10^{-6} m$. Figure 4 shows how the transformational strain rate ratio ($\dot{\varepsilon}_{t,lab}/\dot{\varepsilon}_{t,nat}$) required to cause brittle behavior varies with $T$. The onset of embrittlement would be expected at imposed natural strain rates, $\dot{\varepsilon}_{t,nat}$, of $\sim 10^{-9} s^{-1}$ (horizontal solid red line in Fig. 4), if the grain-size ratio is minimal ($d_{s,nat}/d_{s,lab}$) (solid black line in Fig. 4). However, assuming a maximal initial grain-size ratio ($d_{s,nat}/d_{s,lab}$) (dashed black line in Fig. 4) yields even lower imposed natural strain rates of $\dot{\varepsilon}_{t,nat} \sim 10^{-11} s^{-1}$ (horizontal dashed red line in Fig. 4).

Previous experiments on the germanium - olivine-spinel transition (Burnley et al., 1991) showed a change in rheology similar to that presented in this study. Both transformations, plagioclase breakdown and olivine-spinel transition (Kirby et al., 1996), involve a negative volume change of $-14\% - 17\%$ and $3\% - 10\%$ and an exothermic latent heat release of $\sim 10 - 13$ kJ mol$^{-1}$ and $10 - 50$ kJ mol$^{-1}$, respectively. These thermodynamic properties probably play an important role in triggering brittle instabilities, as the negative volume change enables stress concentrations at crack tips (Kirby, 1987). Along with the heat released during the exothermic reactions, which would promote reactions to proceed even in the absence of larger amounts of fluid, these key ingredients may lead to the generation of self-localizing transformational faults. We show that, contrary to previous arguments (Kirby et al., 1996), transformational faulting is not limited to the case of polymorphic reactions.

Our results demonstrate that reaction-induced grain-size reduction together with a negative volume change trigger embrittlement in the laboratory. This is in accordance with findings of previous numerical and experimental studies on various lithologies (Burnley et al., 1991; Thielmann et al., 2015; Incel et al., 2017) and highlights embrittlement in the lower continental crust, at intermediate depth or deep in...
Earth’s mantle by the same underlying mechanism. It should be noted, however, that the reaction-driven embrittlement mechanism studied here requires a local fluid source. Otherwise, there will be no reaction. In our experiments, there was a small amount of fluid available for hydration reactions. For the Holsnøy case, fluids were introduced into an initially dry lower crust by lower-crustal earthquakes. Earthquakes in the lower crust prior to hydration may be triggered through stress pulses from earthquakes in the normal seismogenic regime (i.e., as aftershocks; Jamtveit et al., 2018). Incipient eclogitization reactions would then allow further brittle deformation in the wall rocks of earthquake-generated faults. In the case of the subducting Indian lower crust, deep crustal earthquakes have been interpreted to have been triggered by dehydration-produced fluids derived from the crust itself (Hetényi et al., 2007), in which case, reaction-driven embrittlement may even explain the initial earthquake activity.

ACKNOWLEDGMENTS

We thank Neil Mancktelow and the other anonymous reviewers who helped to improve the manuscript, as well as Damien Deldeix for the micro- and nanostructural analyses. The study received funding from the Alexander von Humboldt-Foundation (Feodor Lynen Research Fellowship to Incel). Further funding came from the People Program (Marie Curie Actions) of the European Union’s Seventh Framework Program FP7/2011-2013 and Horizon 2020 under REA grant agreements 604713 (to Schubnel) and 669972 (to Jamtveit), National Research Foundation Centre of Excellence grant 223272 (to Andersen), and National Science Foundation grant EAR-1661489 for the development of acoustic emission experiments (to Wang). This research used resources of the Advanced Photon Source, a U.S. Department of Energy Office of Science User Facility operated by Argonne National Laboratory (contract no. DE-AC02-06CH11357).

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Figure 4. Arrhenius plot showing log (εt,nat/εt,lab) (laboratory transformational strain rate and natural transformational strain rate) versus 10,000/K. Vertical lines represent estimated maximum (red) and minimum (blue) eclogitization temperature on Holsnøy, Norway. Depending on initial grain-size ratio, εt,nat/εt,lab required imposed natural strain rate (εn) for embrittlement is ~10–8 to 10–12 s–1 (solid and dashed red horizontal lines).

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