



Mechanisms of deformation-induced trace element migration in zircon resolved by atom probe and correlative microscopy

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Abstract

The widespread use of zircon in geochemical and geochronological studies of crustal rocks is underpinned by an understanding of the processes that may modify its composition. Deformation during tectonic and impact related strain is known to modify zircon trace element compositions, but the mechanisms by which this occurs remain unresolved. Here we combine electron backscatter diffraction, transmission Kikuchi diffraction and atom probe microscopy to investigate trace element migration associated with a ~20 nm wide, 2° low-angle subgrain boundary formed in zircon during a single, high-strain rate, deformation associated with a bolide impact. The low-angle boundary shows elevated concentrations of both substitutional (Y) and interstitial (Al, Mg and Be) ions. The observed compositional variations reflect a dynamic process associated with the recovery of shock-induced vacancies and dislocations into lower energy low-angle boundaries. Y segregation is linked to the migration and localisation of oxygen vacancies, whilst the interstitial ions migrate in association with dislocations. These data represent the direct nanoscale observation of geologically-instantaneous, trace element migration associated with crystal plasticity of zircon and provide a framework for further understanding mass transfer processes in zircon.

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1. INTRODUCTION

Zircon (ZrSiO₄) is a common accessory mineral that occurs in most crustal rocks. The low diffusivity of most trace elements through the zircon lattice, inferred from

trace element zonation (Vavra, 1990; Hoskin, 2000) and diffusion experiments (Cherniak et al., 1997; Cherniak and Watson, 2003, 2007), make zircon a robust geochemical repository. Hence, the trace and rare earth elements (REE) incorporated into the zircon are commonly used to place valuable constraints on petrogenetic processes (Hoskin and Schaltegger, 2003). For example, the trace element geochemistry of zircon yields source rock type and crystallization conditions of igneous rocks (Belousova

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et al., 2002; Ferry and Watson, 2007; Hanchar and van Westrenen, 2007; Grimes et al., 2009; Claiborne et al., 2010) and can place constraints on recrystallization mechanisms, hydrothermal alteration and the histories of metamorphic rocks (Hoskin and Black, 2000; Hoskin, 2005; Harley et al., 2007; Marsh and Stockli, 2015). The trace element composition of zircon also has economic importance, for example being used to assess the prospectivity of granites for mineralisation (Ballard et al., 2002; Dilles et al., 2015).

The incorporation of trace amounts of uranium, and its subsequent radioactive decay to lead, enables the U–Pb dating of zircon to place temporal constraints of numerous crustal processes (Harley and Kelly, 2007; Corfu, 2013). When combined with Lu–Hf and oxygen isotopic data, zircon can be used to constrain crustal evolution over a range of timescales (Hawkesworth and Kemp, 2006; Parman, 2015; Payne et al., 2016). In addition, the ability of zircon to withstand weathering, erosion, sedimentary transport and diagenesis, make zircon a common target for sedimentary provenance analysis (Fedó et al., 2003; Gehrels, 2014) and the geochemistry and geochronology of ancient detrital zircon grains is the principal means of understanding petrogenetic processes and environmental conditions in the earliest stages of Earth history (Maas et al., 1992; Wilde et al., 2001; Hoskin, 2005; Watson and Harrison, 2005; Harrison and Schmitt, 2007; Ushikubo et al., 2008; Harrison, 2009). Complementing the terrestrial studies of Hadean zircon are analyses from lunar and meteoritic zircon samples, which provide fundamental constraints on the early solar system and planetary evolution (Nemchin et al., 2010; Humayun et al., 2013; Iizuka et al., 2015). However, despite the broad application of zircon in geochemical and geochronological studies, it is widely recognised that a number of different processes may modify the trace element compositions of zircon.

Radiation damage within zircon can facilitate trace element redistribution and the incorporation of non-formula elements (Ewing et al., 2003; Palenik et al., 2003; Horie et al., 2006) even under low temperature hydrothermal conditions (Geisler et al., 2002; Pidgeon, 2014). Trace element modification associated with radiation damage reflects a complex interaction of the self-irradiation process, enhanced diffusion along radiation-induced defects, and reactions associated with fluid ingress by radiation-enhanced fractures and recrystallization (Geisler et al., 2007; Nasdala et al., 2010).

Detailed microstructural characterisation has demonstrated that crystal plastic deformation of zircon may take place in Earth's crust due to tectonic processes (Reddy et al., 2007, 2009; Piazzolo et al., 2012) and meteorite impact events (Moser et al., 2011; Cavosie et al., 2015). Geochemical analyses of deformed zircon indicate that trace element compositions may be modified in the vicinity of intracrystalline defects, particularly in the regions of low-angle boundaries (Reddy et al., 2006; Timms et al., 2006, 2011; Moser et al., 2009, 2011; Nemchin et al., 2009; Piazzolo et al., 2016). A number of models have been proposed to explain the observed relationship between microstructure and trace element migration including enhanced diffusion

along dislocation pipes and low-angle boundaries (Reddy et al., 2006; Moser et al., 2011; Timms et al., 2011; Piazzolo et al., 2016), incorporation of trace elements within migrating dislocations (Reddy et al., 2006, 2007; Piazzolo et al., 2016) and creep cavitation (Timms et al., 2012a). However, crystal defects may also trap trace elements; for example, Pb has been shown to segregate into dislocation loops during metamorphism (Peterman et al., 2016).

Constraining the processes that are responsible for deformation-related compositional modification of zircon has remained elusive because the volume of material typically needed to characterise compositional heterogeneities (100 s of μm^3) is considerably larger than the sub-micron scale microstructures in which these heterogeneities occur. Direct comparison with compositional data has required averaging of quantitative microstructural data over similar volumes to those measured by quantitative analytical techniques (Timms et al., 2006, 2011). Higher spatial resolution analytical methods, for example, hyperspectral cathodoluminescence (CL) data, indicate variations in the concentrations of trivalent REEs at the micrometre scale, but these are not quantitative (Reddy et al., 2006; Timms and Reddy, 2009; Timms et al., 2011). As a result, the spatial relationships between deformation microstructures and compositional variations, as well as the processes responsible for trace element mobility in deformed or defect-enriched zircon, have proved difficult to resolve.

The recent applications of atom probe microscopy to zircon have highlighted the potential for this analytical technique to quantify nanoscale compositional variations and establish the controls and processes associated with trace element modification (Valley et al., 2014, 2015; Peterman et al., 2016; Piazzolo et al., 2016). Here we combine electron backscatter diffraction (EBSD), transmission Kikuchi diffraction (TKD) and atom probe microscopy to investigate the nanoscale relationships between microstructure and trace element composition in a zircon grain that records a single, shock deformation event associated with a meteorite impact.

2. SAMPLE AND ANALYTICAL PROCEDURES

2.1. Sample description

The Stac Fada Member of the Stoer Group of sedimentary rocks in NW Scotland represents an ejecta deposit associated with a meteorite impact ~ 1.18 billion years ago (Amor et al., 2008; Parnell et al., 2011; Reddy et al., 2015). The unit extends some 50 km along strike and has a variable thickness that in places exceeds 20 m (Fig. 1). It comprises three main facies types attributed to deposition from a single decelerating granular density current (Branney and Brown, 2011). The analysed sample (14-SF-01) was collected from the basal layer of the Stac Fada Member (UK Grid Reference NC 03348 28515 equivalent to Latitude 58.2014, Longitude -5.3482 in WGS84) (Fig. 1) and is a matrix-supported, poorly-sorted breccia comprising centimetre size clasts of lithic and devitrified melt fragments. The sample shows no evidence of

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