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Empirical constraints on the effects of radiation damage on helium diffusion in zircon

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Abstract

In this study, we empirically evaluate the impact of radiation damage on zircon (U-Th)/He closure temperatures for a suite of zircon crystals from the slowly cooled McClure Mountain syenite of south-central Colorado, USA. We present new zircon, titanite, and apatite conventional (U-Th)/He dates, zircon laser ablation (U-Th)/He and U-Pb dates, and zircon Raman spectra for crystals from the syenite. Titanite and apatite (U-Th)/He dates range from 447 to 523 Ma and 88.0 to 138.9 Ma, respectively, and display no clear correlation between (U-Th)/He date and effective uranium concentration. Conventional zircon (U-Th)/He dates range from 230.3 to 474 Ma, while laser ablation zircon (U-Th)/He dates show even greater dispersion, ranging from 5.31 to 520 Ma. Dates from both zircon (U-Th)/He datasets decrease with increasing alpha dose, indicating that most of the dispersion can be attributed to radiation damage. Alpha dose values for the dated zircon crystals range from effectively zero to 2.15×10^{19} α/g, spanning the complete damage spectrum.

We use an independently constrained thermal model to empirically assign a closure temperature to each dated zircon grain. If we assume that this thermal model is robust, the zircon radiation damage accumulation and annealing model of Guenthner et al. (2013) does not accurately predict closure temperatures for many of the analyzed zircon crystals. Raman maps of the zircons dated by laser ablation document complex radiation damage zoning, sometimes revealing crystalline zones in grains with alpha dose values suggestive of amorphous material. Such zoning likely resulted in heterogeneous intra-crystalline helium diffusion and may help explain some of the discrepancies between our empirical findings and the Guenthner et al. (2013) model predictions. Because U-Th zoning is a common feature in zircon, radiation damage zoning is likely to be a concern for most ancient, slowly cooled zircon (U-Th)/He datasets. Whenever possible, multiple mineral-isotopic systems should be employed to add additional, independent constraints to a sample's thermal history.

Keywords: Zircon (U-Th)/He thermochronology; Radiation damage

1. INTRODUCTION

Helium diffusion in U- and Th-bearing accessory minerals is sufficiently rapid at low temperatures that (U-Th)/He dating has evolved into a powerful tool for reconstructing cooling histories in the upper crust, informing our understanding of surface and near-surface processes (e.g. Farley, 2002; Ehlers and Farley, 2003; Reiners and Brandon, 2006). Although most commonly applied to geologic problems in active orogenic systems, (U-Th)/He thermochronology also has proven useful for studies of the development and long-term stability of continental interiors (Flowers et al., 2006; Flowers, 2009; Enkelmann and Garver, 2016).

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Applications of low temperature thermochronology to slowly cooled terrains however, present unique challenges. (U-Th)/He datasets from Precambrian and Paleozoic settings are often dispersed well beyond what is expected based on analytical uncertainty alone, making datasets difficult to interpret unambiguously (Flowers et al., 2006). Zircon (U-Th)/He (ZrnHe) datasets in particular are often problematic in this regard. Dispersion has been attributed to: (1) the effect of U-Th zonation on alpha ejection corrections, (Hourigan et al., 2005; Bargnesi et al., 2016); (2) anisotropic diffusion (Cherniak et al., 2009); (3) multidomain diffusion, (Reiners et al., 2004); and (4) radiation damage (Guenthner et al., 2013). Of these, radiation damage predominantly attributed to alpha decay of U and Th may be especially significant for slowly cooled zircons (Guenthner et al., 2013).

During an alpha decay event, kinetic energy is lost through the emission of an alpha particle and recoil of the heavy daughter nuclide (Weber, 1990; Ewing et al., 2003). Alpha recoil results in a cascade of atomic-scale collisions, damaging the crystal structure. Damage accumulates over time, affecting material properties. Importantly, however, radiation damage can also anneal over time at elevated temperatures (Murakami et al., 1991; Zhang et al., 2000), such that the amount of damage to a crystal's structure is a function of U and Th concentration, age, and thermal history (Nasdala et al., 2001; Guenthner et al., 2013).

For years, researchers have noted correlations between U and Th content and zircon (U-Th)/He dates (e.g. Hurley, 1952; Hurley and Fairbairn, 1953; Guenthner et al., 2013; Guenthner et al., 2014; Guenthner et al., 2015; Orme et al., 2016; Powell et al., 2016; Johnson et al., 2017). Guenthner et al. (2013) recently attempted to quantify the effects of radiation damage on helium diffusion kinetics through a series of step-heating experiments on crystallographically oriented zircon slabs with varying degrees of structural damage. Results indicated that He retentivity increases at low damage doses from $\sim 1 \times 10^{16}$ to 1.5×10^{17} α/g. At greater damage doses, however, their data suggested that He diffusivity dramatically increases. The authors proposed that the low-dosage increases in He retentivity reflect progressive obstruction of fast pathways for helium diffusion parallel to the *c*-axis. They posited that, as radiation damage levels increase, the damage zones interconnect to create a series of new fast paths, increasing He diffusivity. Their findings demonstrated that radiation damage has important implications for ZrnHe thermochronology, namely, that heavily damaged zircon grains may be sensitive to temperatures far lower than the previously accepted 180–200 °C closure temperature for helium in zircon (Guenthner et al., 2013; Reiners et al., 2004; Reiners et al., 2002).

From these data, Guenthner et al. (2013) developed a zircon radiation damage accumulation and annealing model (ZRDAAM) designed to calculate helium diffusivity as a function of temperature history based on each zircon crystal's measured U-Th content. ZRDAAM has been applied to a number of bedrock and detrital ZrnHe datasets, yielding valuable information about the thermal history of the regions studied (e.g. Guenthner et al., 2014; Guenthner et al., 2015; Orme et al., 2016). A few recent studies however, have acknowledged that applications of the model are not uniformly successful, especially for zircons with higher damage doses (Powell et al., 2016; Johnson et al., 2017). For example, Powell et al. (2016) has suggested that the damage threshold for increased helium diffusivity may be too high. Johnson et al. (2017), in turn, reported a dataset in which heavily damaged zircons appeared more helium retentive than predicted by ZRDAAM.

The McClure Mountain syenite of south-central Colorado, USA affords an opportunity to empirically test ZRDAAM on a suite of variably damaged zircons from a single sample with a simple, independently constrained thermal history. In this study, we present new zircon, titanite, and apatite conventional (U-Th)/He dates, zircon laser ablation (U-Th)/He and U-Pb dates, and zircon Raman spectra for crystals separated from a sample of the syenite. We modeled the cooling history of the syenite by combining our TtnHe and ApHe dataset with previously published geochronologic and thermochronologic data. We then used this information to derive empirical constraints on the relationship between radiation damage and ZrnHe closure temperature for comparison with the Guenthner et al. (2013) model.

2. THE MCCLURE MOUNTAIN SYENITE

Located in the northern Wet Mountains of south-central Colorado, the McClure Mountain Complex spans ~74 km^2 from McClure Mountain and Deer Mountain in the southwest to Iron Mountain in the northeast (Parker and Hildebrande, 1963; Olson et al., 1977). Originally discovered and named by the U.S. Geological Survey in the 1960s, this concentrically zoned, alkalic complex intrudes Precambrian granitic gneisses and metamorphic rocks. The complex consists of pyroxene-olivine-plagioclase cumulates intruded by the syenite (Parker and Hildebrande, 1963; Olson et al., 1977; Alexander et al., 1978). Major-rock forming minerals in the syenite include K-feldspar, plagioclase, hornblende, biotite, and clinopyroxenes, but significant amounts of titanite and apatite occur in most samples as well (Samson and Alexander, 1987; Schoene and Bowring, 2006). Zircon, baddeleyite, zirconolite, ilmenite, magnetite, nepheline, calcite, iron-sulfides, and alteration products are minor constituents (Schoene and Bowring, 2006).

The syenite has been the subject of numerous geochronologic studies, mostly aimed at establishing its intrusive age. Zircon isotope dilution, thermal ionization mass spectrometry (ID-TIMS) $^{207}Pb/^{235}U$ dates are interpreted to indicate a crystallization age of 523.98 ± 0.12 Ma (Schoene and Bowring, 2006). (Throughout this paper, all uncertainties are presented at the 2σ level.) ID-TIMS $^{207}Pb/^{235}U$ data for titanite and apatite yield dates of 523.26 ± 0.65 Ma and 523.5 ± 1.5 Ma respectively, implying rapid cooling of the intrusive body following crystallization (Schoene and Bowring, 2006). Laser ablation inductively coupled plasma mass spectrometry (LA-ICPMS) analyses of McClure Mountain apatite yield