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$^{40}Ar/^{39}Ar$ dating and thermal modeling of adularia to constrain the timing of hydrothermal activity in magmatic settings

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ABSTRACT

Hydrothermal systems develop via interspersed thermal events over tens to hundreds of thousands of years. The timing of how these systems evolve is commonly established via application of geochronology to a variety of phases and/or the indirect correlation of dated stratigraphy. Here we report $^{40}Ar/^{39}Ar$ results from adularia extracted from a single mineralized fracture in the late Quaternary Tauhara geothermal system of New Zealand. By utilizing both the age and Ar diffusion properties, we demonstrate how adularia can provide reliable temporal and thermal constraints on the evolution of geologically youthful and active geothermal systems. Our results indicate that adularia formation occurred after 30 ka (mean age 15 ± 10 ka), possibly resulting from subsurface fracturing induced by a 25.4 ka hydrothermal eruption. Simulation of transient heat effects upon Ar retention in adularia with respect to the thermal history of the Tauhara geothermal system confirm that this age is consistent with the time of adularia crystallization. Overall, $^{40}Ar/^{39}Ar$ dating on geologically young hydrothermal adularia with respect to its thermal history may be used to assess the timing and potential events (e.g., eruptions and fracturing) related to hydrothermal system evolution.

INTRODUCTION

Geothermal fields are geographically stable features that exhibit thermal variability throughout their geological lifetimes (Bibby et al., 1995; Arehart et al., 2002; Milicich et al., 2014). The details of transient subsurface temperature variation over the lifetime of a hydrothermal system are difficult to constrain, especially in young and active systems, and remain a central subject of debate at many active hydrothermal sites. Similarly, application of radiometric dating approaches to hydrothermal-related mineralization is a challenging task that typically requires assumptions that are controversial. Previous studies of fossil geothermal systems (e.g., Mauk and Hall, 2004; Mauk et al., 2011) and active geothermal systems (e.g., Grimes et al., 1998; Arehart et al., 2002) have provided a foundation for dating the evolution of these systems; however, continued advancement, whether in methodology and/or mineral studies, is needed to assess the complexity of geologically young and active hydrothermal systems.

The Taupo Volcanic Zone (TVZ; Bibby et al., 1995) of New Zealand hosts 23 active geothermal systems and 2 fossil systems. The rifted continental arc forms the southernmost expression of the Tonga-Kermadec subduction system and has been a locus for highly productive silicic volcanism over 2 m.y. (Wilson et al., 1995). In TVZ geothermal systems, life cycles of individual hydrothermal cells have been considered to span several hundreds to thousands of years with elevated temperatures maintained by repetitive heat pulses of tens to thousands of years from local magmatic intrusions (Grindley, 1965; Browne, 1979; Chambefort et al., 2014; Milicich et al., 2014). However, the specific timing and extent of thermal events in the context of these short life cycles and young systems creates challenges, especially with respect to their noble gas isotope geochemistry and related dating methods.

Vein adularia previously showed promise as a geochronologic target to elucidate the thermal histories of youthful geothermal systems (Hulen et al., 1997). Drill cores recently recovered from the Tauhara sector of the joint Wairakei and Tauhara geothermal fields (Wairakei-Tauhara) of the TVZ (Fig. DR1 in the GSA Data Repository[1]) intersected a mineralized fracture sealed by large hydrothermal adularia crystals ($KAlSi_3O_8$) that present an ideal opportunity to directly assess mineralization related to its Ar isotope geochemistry as well as events that may have aided adularia formation in the geothermal system. Multicollector ion-counting capabilities in modern $^{40}Ar/^{39}Ar$ mass spectrometers combined with development of reference gases for detector intercalibration have reduced some of the challenges of dating young materials (Coble et al., 2011; Jourdan et al., 2014). Previous investigations directly dating TVZ geothermal systems using U-Th disequilibrium and $^{40}Ar/^{39}Ar$ methods support continual thermal replenishment (Grimes et al., 1998; Arehart et al., 2002), but lack the age resolution to accurately constrain the most recent (i.e., younger than 100 ka) stages of evolution. Here we conduct $^{40}Ar/^{39}Ar$ dating and assess the effect of thermally activated Ar diffusion on adularia sampled from a single fracture to help constrain the youthful timing, magnitude of heating, and possible events within an active geothermal system.

MATERIALS AND METHODS

Sample Collection and Preparation

Four core fragments containing hydrothermal adularia were collected between 774 and 786 m depth along a mineralized fracture in well THM16 from the Tauhara geothermal field (Fig. DR1; samples 16_774.3, 16_775.4, 16_778.73, and 16_785.85). The fracture was intersected between 760 m and 790 m depth, was nearly vertical with an aperture of 0.5–1 cm, and was normally offset by 1–2 cm (Fig. DR2A; see the Data Repository). Lining the fracture is a hydrothermal mineral assemblage of quartz, adularia (Figs. DR2A and DR2B), accessory pyrite, calcite, wairakite ± epidote, and hematite. The absence of mineral layering or crustiform-type banding indicates coprecipitation of the phase assemblage following fracture formation. The fracture cuts a ca. 310 ka light brown pumice and lithic lapilli-tuff ignimbrite of the lowermost Waiora Formation drilled between 730 m and 800 m depth in THM16 (Rosenberg et al., 2009a, 2009b). The ignimbrite is intensely silicified and occurs in a propylitic alteration zone. Overlying the ignimbrite are lacustrine-deposited tuffs and at least

[1]GSA Data Repository item 2017011, background of adularia collection and characterization $^{40}Ar/^{39}Ar$ measurements, including Figure DR1 (New Zealand field location maps), Figure DR2 (adularia samples), Figure DR3 (conceptual image of adularia formation), Figure DR4 (diode laser heated adularia packet), Figure DR5 (adularia heating over time), and Figure DR6 (calibration measurements), is available online at http://www.geosociety.org/pubs/ft2017.htm or on request from editing@geosociety.org.

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eight local hydrothermal eruption breccias of the upper Waiora Formation (210–730 m) followed by at least four further hydrothermal eruption deposits of the Crown Breccia (44–210 m; Rosenberg et al., 2009b).

Adularia crystals were hand-picked under a binocular microscope from the vein surface exposed within the recovered core fragments (Fig. DR2C). The selected grains were cloudy white, rhombic, 400–600 μm, and free from intergrowths. Petrological microscopy and compositions measured using an Oxford Instruments X-ray energy-dispersive spectroscopy (EDS) detector mounted on a JEOL 6400 scanning electron microscope at the University of Canterbury (Christchurch, New Zealand) confirmed that the selected crystals had the crystallographic and compositional properties of adularia.

$^{40}Ar/^{39}Ar$ Measurements

CO_2 laser total fusion $^{40}Ar/^{39}Ar$ analyses were obtained from 87 adularia single crystals from the 4 hydrothermal adularia samples at Stanford University (California, USA) using a Noblesse multicollector noble gas mass spectrometer (Coble et al., 2011). Taylor Creek sanidine (28.34 Ma; Renne et al., 1998) was used as the flux monitor. Zero age kalsilite glass yielded a $(^{40}Ar/^{39}Ar)_K$ value of $4.9 \pm 1.6 \times 10^{-3}$. An additional 1 mg (4 grain) sample was wrapped in a 3 mm Ta foil disk and incrementally heated using a near infrared (908 μm) diode laser to determine the diffusion properties of the adularia using standard methods developed for basement K-feldspar (Lovera et al., 1997). (For complete experimental details and data tables, see the Data Repository.)

RESULTS

Adularia Mineralogy

Petrography of the adularia crystals identified patchy tile domains and polysynthetic twinning within the crystal interiors. Chemical EDS results confirmed that the crystals are adularia, similar to results by Steiner (1970). No fluid inclusions or impurities were apparent at 40× magnification for all samples investigated. In the vein host rock, the dominant matrix and void spaces were intensely silicified by secondary quartz and primary feldspar was replaced by patchy adularia pseudomorphs (Fig. DR2D).

$^{40}Ar/^{39}Ar$ Inverse Isochron Results

Figure 1 displays the pooled laser fusion $^{40}Ar/^{39}Ar$ results from the four hydrothermal adularia samples (n = 87). Inverse isochron analyses yielded an intercept age of 15 ± 10 ka and a trapped $^{40}Ar/^{39}Ar$ atmospheric intercept of 290.6 ± 1.5 (2σ standard deviation; mean square of weighted deviates, MSWD = 1.17). As indicated in Figure 1, the sample gas was dominantly atmospheric (~99%) with only a trace of radiogenic ^{40}Ar ($^{40}Ar^*$) detected. This resulted in a wide extrapolated data spread at 2σ along the radiogenic axis. Ages calculated for individual analyses are highly imprecise but clustered about the inverse isochron age.

Simulation of Transient Heating Effects Upon Ar Retention

Incremental heating results from the 1 mg adularia sample yielded an activation energy of 45.1 ± 2.5 kcal/mol and a preexponential frequency factor $(D_0/r^2) = 3.1 \pm 0.5\ s^{-1}$ (Fig. 2A). To simulate the $^{40}Ar^*$ retentivity of adularia in nature, we applied the multidiffusion domain (MDD) approach (e.g., Lovera et al., 1997) to model the ^{39}Ar release below the onset of melting at ~1200 °C (see the Data Repository). Figure 2B contours the $^{40}Ar^*$ loss in adularia in percent as a function of temperature (°C) and heating duration (years) based upon the calculated MDD properties. No cooling occurs in these calculations and radiogenic ingrowth of $^{40}Ar^*$ is considered. As indicated, ^{40}Ar is quantitatively retained when a temperature of 200 °C is maintained for any interval between 1 yr and 10^6 yr. For short-lived (i.e., 100 yr) thermal events at higher temperatures, adularia undergoes $^{40}Ar^*$ fractional loss of 0.3%, 5.8%, and 31.6% for 300, 400, and 500 °C, respectively (Fig. 2B). Longer duration heating (e.g., 10 k.y. intervals) yields progressively higher $^{40}Ar^*$ fractional loss values of 3.3%, 35.2%, and 96.7% for 300, 400, and 500 °C, respectively.

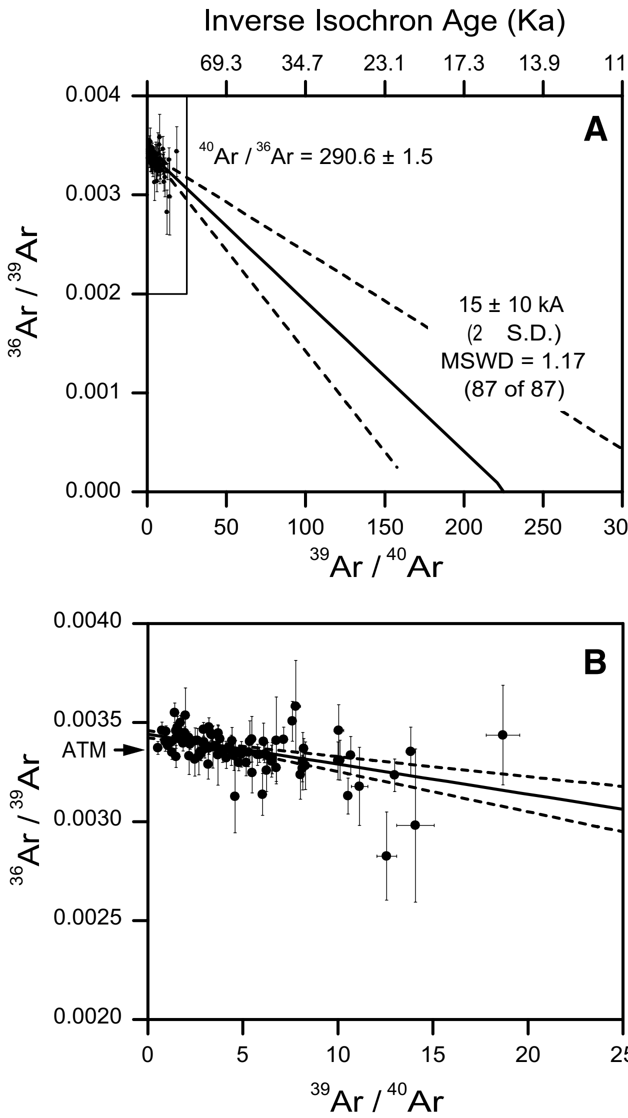

Figure 1. A: Inverse isochron plot showing all results (4 samples, n = 87 measurements). No data have been omitted from the overall data set. The upper and lower projected lines are 95% confidence bands. The central line is the extrapolated best fit that corresponds with a mean age of 15 ± 10 ka (2σ standard deviation, S.D.; mean square of weighted deviates, MSWD = 1.17). Note that the best-fit y-intercept ($^{40}Ar/^{36}Ar$ = 290.6) is different from atmospheric composition (ATM; $^{40}Ar/^{36}Ar$ = 295.5). B: Detail of the data close to the y-axis is shown.

DISCUSSION

The $^{40}Ar/^{39}Ar$ inverse isochron age of 15 ± 10 ka (2σ) suggests that the fracturing and mineralization event is geologically young relative to the overall age span of the system (older than 200 ka; Grindley, 1965; Browne, 1979; Arehart et al., 2002). To confirm the validity and usefulness of this young age, we must critically consider what it may mean. While significant extrapolation of the clustered results near the atmospheric ($^{36}Ar/^{40}Ar$) axis to the radiogenic ($^{39}Ar/^{40}Ar$) axis degrades the precision of the intercept age, the ±10 k.y. (2σ) error envelope is sufficiently precise to yield a geologically useful result (Fig. 1). For example, no line of best fit can be extrapolated to older than ca. 30 ka based on the examination of all of the data points identified. At face value, our adularia $^{40}Ar/^{39}Ar$ results document youthful hydrothermal activity within the long-lived Tauhara

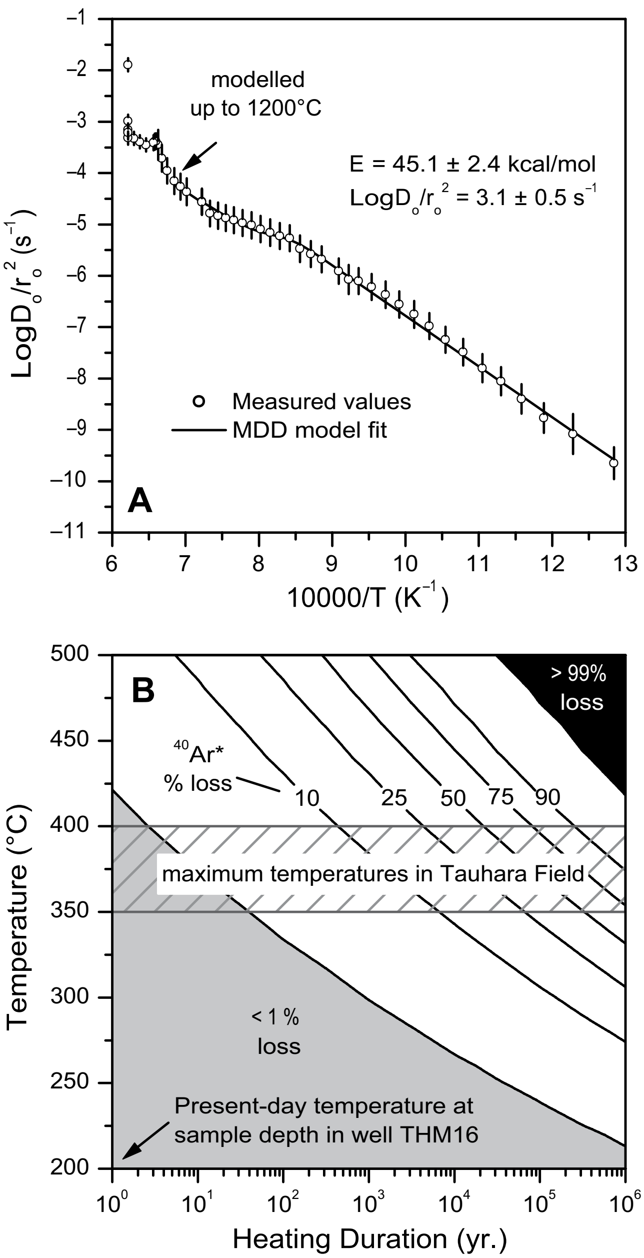

Figure 2. A: Arrhenius plot calculated from ^{39}Ar release from adularia during incremental heating. The indicated activation energy (E) and frequency factor (D_o/r^2) represent an average fit to ^{39}Ar diffusivities below the onset of melting (~1200 °C). MDD—multidiffusion domain. B: Contours of predicted $^{40}Ar^*$ fractional loss from adularia as a function of temperature and heating duration. Contours are the percent of Ar lost at the given heating duration in years and temperature. The maximum temperature (hachured field) within the Tauhara field and present-day temperature measured in well THM16 at the sample depth are indicated.

geothermal system and thus support the principal concept of continual system evolution in the TVZ consisting of thermal waxing and waning periods (e.g., Milicich et al., 2014; Chambefort et al., 2014). Given the compositional limitations of the samples, we confidently suggest that ca. 30 ka represents a maximum bound for the time that the adularia vein formed. However, it is possible that uptake of excess ^{40}Ar ($^{40}Ar_E$) during crystallization and/or thermally induced ^{40}Ar loss by protracted heating could invalidate the geologic significance of the age.

Existence of $^{40}Ar_E$ is common in hydrothermal fluids associated with plutonic heat sources (Kelley et al., 1986). Uptake of $^{40}Ar_E$ into the mineral lattice or in trapped fluid inclusions would erroneously increase the model $^{40}Ar/^{39}Ar$ age of the adularia. Based upon likely fluid/crystal partition coefficients for Ar, Kelley et al. (1986) and Kelley (2002) calculated that the $^{40}Ar/^{39}Ar$ ages of K-feldspar that interacted with hydrothermal fluids containing 0.1–1 ppm of $^{40}Ar_E$ would be 1–10 k.y. too old. The observed high degree of atmospheric contamination detected in all four of the adularia samples (Fig. 1) is likely due to interaction with meteoric and hydrothermal fluids. In hydrothermal fluids from surface geothermal fumaroles at Wairakei-Tauhara, low nonatmospheric (≤5.2 × 10^{-4} ppm) and atmospheric ^{40}Ar (2 × 10^{-4} to 2 × 10^{-3} ppm) concentrations have been measured (Mazor et al., 1990). Given fluid/crystal partition coefficients <10^{-3}, such low concentration levels of Ar are insufficient to affect our $^{40}Ar/^{39}Ar$ model age results beyond stated 2σ uncertainties. This conclusion is supported by the well-constrained, lower than atmosphere, $^{40}Ar/^{36}Ar$ axis intercept (290.6 ± 1.5, 2σ) indicated by our York regression results.

Loss of $^{40}Ar^*$ by thermally activated diffusion could have reduced the $^{40}Ar/^{39}Ar$ age below the crystallization age of the vein if past temperatures were appreciably higher than currently observed in the borehole (Fig. 2). The hydrothermal mineral assemblage suggests that crystallization occurred at >230 °C (White and Hedenquist, 1990; Rosenberg et al., 2009b). The temperature in well THM16 at the sample depth was measured as 200 °C. Maximum measured temperatures in the Tauhara field are slightly >300 °C at depth (1500 m; Contact Energy, 2010). Thermal conditions beyond deep drilling at Wairakei-Tauhara (maximum 3 km) are not well understood, but extrapolation of known thermal gradients (20 °C/km) from deeper geothermal wells (McNabb, 1992) to 8 km depth suggests that temperatures between 350 and 400 °C may occur at great depth (Kissling and Weir, 2005). We regard a temperature range of 350–400 °C as a maximum bound upon the magnitude of a thermal pulse that could have occurred subsequent to vein formation. The diffusion calculations (Fig. 2) show that transient heating at 400 °C could have caused $^{40}Ar^*$ loss between 5.8% and 35.2% for heating intervals between 0.1 and 10 k.y.

An existing U-Th age by Grimes et al. (1998) from Tauhara well THM1 (Fig. DR1C) returned a hydrothermal calcite age of 99 ± 44 ka. Had adularia crystallization occurred at 100 ka, and a 1-k.y.-long, 400 °C transient heating event followed ca. 10 ka, diffusive ^{40}Ar loss would only have reduced the $^{40}Ar/^{39}Ar$ age to ca. 87 ka. Our diffusion calculations indicate that geologically unrealistic protracted heating at higher temperature (e.g., heating at 500 °C for >10 k.y.) would have been required to completely reset the $^{40}Ar/^{39}Ar$ age to 15 ka. Overall, the calculations demonstrate that adularia requires either much smaller diffusive transport distances (Fig. 2A) or more protracted heating at very high temperature (Fig. 2B) to induce significant $^{40}Ar^*$ loss. Because such conditions exceed plausible limits at the comparatively shallow 780 m depths we have sampled in the Tauhara geothermal system, we regard the measured $^{40}Ar/^{39}Ar$ age of 15 ± 10 ka as being consistent with the time of adularia crystallization.

By confirming the validity of this age, we now discuss its usefulness. The fracturing mechanism that permitted adularia crystallization is most consistent with the stress induced by a volcanic or hydrothermal eruption event (Nelson and Giles, 1985; Henneberger and Browne, 1988). In the overlying Crown Breccia hydrothermal eruption deposit, lithic clasts from the 25.4 ka Oruanui Formation (Rosenberg et al., 2009b; Vandergoes et al., 2013) support an eruption age consistent with the $^{40}Ar/^{39}Ar$ age. The eruption focal depth could be reflected by the fracture depth at 550 m below the Crown Breccia depositional surface, a depth consistent with other TVZ hydrothermal eruptions (~450 m deep; Browne and Lawless, 2001). We infer, therefore, that fracturing at the sample depth (760–790 m) is due to hydraulic fracturing associated with Crown Breccia hydrothermal eruptions (Fig. DR3). Such overpressurization fracturing and brecciation can initially enhance reservoir rocks permeability, but promote hydrothermal mineralization, sealing fractures and reducing permeability (Nelson and Giles, 1985; Henneberger and Browne, 1988; Browne and Lawless, 2001).

Enhanced permeability by fracturing and fault movement in part regulates the longevity of geothermal systems in the TVZ (Browne and Lawless, 2001; Kissling et al., 2015). Together with fluid recharge and repeated heating intrusions (Milicich et al., 2014), the presented age is consistent with active TVZ geothermal surface features identifying hydrothermal systems to be continually evolving features with a level of natural self-sustainability. By developing the $^{40}Ar/^{39}Ar$ dating of adularia, temporal constraints with regard to crystallization, fracturing, permeability, and thermal histories may begin to be assessed at higher resolution in hydrothermal systems on a time scale much less than hundreds of thousands of years.

CONCLUSIONS

We have demonstrated that $^{40}Ar/^{39}Ar$ dating of adularia is suitable for directly assessing the age of young hydrothermal adularia in order to refine the timing of activity in complex and relatively youthful geothermal environments. Argon diffusion models and the thermal history of the Tauhara geothermal system demonstrate that our age, although highly imprecise, is geologically meaningful. Specifically, adularia formation occurred after 30 ka and is likely related to a hydrothermal eruption (25.4 ka) that increased permeability and provided favorable conditions for adularia crystallization in the hydrothermal system. Overall, this method may be used to constrain the age of hydrothermal activity and to interpret or further clarify the timing of thermal events, reservoir fracturing, and/or faulting in other major geothermal fields.

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