Laboratory and natural diffusivity calibrations for apatite (U-Th)/He thermochronometry

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A critical requirement for meaningful thermochronometry is reliable calibration of the temperature sensitivity of a particular dating method. Recent work suggests that (U-Th)/He dating of apatite provides unique information on cooling through very low temperatures (<100°C), but uncertainty remains about the general nature of He diffusion from apatite and any physico-chemical controls (Zeitler *et al.*, 1987, Lippolt *et al.*, 1994, Wolf *et al.*, 1996, Warnock *et al.*, 1997). Here we report new laboratory experiments and borehole ages supporting a He closure temperature t_c~ 75°C (cooling rate 10°C/Myr) that is applicable in nature and is apparently insensitive to grain size and composition.

We have investigated the He diffusion behaviour of gem-quality and more typical fine-grained apatites using a fully automated isotope dilution quadrupole mass spectrometry system. The new work extends our previous incremental outgassing results (Wolf *et al.*, 1996) by increasing the number of characterized apatites and the number of data points per sample, and through extensive use of temperature cycling. Fig. 1 is an Arrhenius plot showing our new results on two splits of Durango fluorapatite. The first split (open symbols) was subjected to monotonically increasing temperatures, yielding a linear Arrhenius relationship ($r^2 = 0.999$, n = 50) corresponding to t_c = 72°C. The second (filled symbols), subjected to a complex heating schedule involving multiple cycles from low to high temperatures, yields data that are indistinguishable from the first run (t_c = 70°C, r^2 = 0.997, n = 50). These experiments demonstrate good reproducibility with our current apparatus, confirm our previous t_c on Durango apatite, and argue for a single mechanism/site for He diffusion from this gem-quality sample.

Similar temperature-cycled experiments were performed on other igneous, metamorphic, and sedimentary apatites. Most of these apatites yield linear Arrhenius plots and closure temperatures in the range 70–80°C, similar to our results on Durango and apatites from the Peninsular Ranges Batholith (Wolf *et al.*, 1996). We can identify no compositional or grain-size control on He diffusivity. However, a subset of apatite samples display a more complex behaviour. Rather than defining a line on an Arrhenius plot, these samples exhibit a zig-zag pattern in which the apparent diffusivity at a given temperature increases as the experiment proceeds (Fig. 2; numbers and arrows indicate the sequence of points). We attribute this behaviour to failure of the common assumption that the initial concentration of diffusant within the sample is uniform. This assumption is required for transforming gas yield data into diffusivity, but is invalid in apatites recently at elevated temperatures; such apatites must have He profiles rounded on the edges by diffusion. We have simulated this effect numerically by computing the concentration profile in an apatite held at 55°C over geologic time. A synthetic incremental outgassing experiment of this 'sample', in which we again make the (erroneous) assumption of an initially uniform He profile, yields a zig-zag pattern nearly identical to Fig. 2. If this model is correct, the inherent diffusivity of a 'zig-zag' sample can only be obtained from the last linear segment of the Arrhenius profile.

There are two important conclusions from this observation. First, temperature cycling is critical for the accurate determination of apatite He diffusion parameters. For example, the first nearly linear segment of the run in Fig. 2a would yield t_c = 120°C, whereas the final linear segment corresponds to a more reasonable value of 78°C. Second, the diffusion behaviour revealed by stepped-heating is very sensitive to the shape of the initial He profile, which itself is very sensitive to the cooling history experienced by the sample. We are presently exploring the combination of apatite He ages and

stepped-heating behaviour to more fully constrain thermal histories. Interestingly, most of the samples showing the zig-zag pattern are indeed from areas that have experienced recent exhumation, such as the San Gabriel Mtns (southern California).

To confirm that laboratory diffusivity data can be extrapolated to conditions relevant in nature, we have analysed apatite He ages in several sedimentary (Otway Basin, SE Australia) and igneous (Cajon Pass, CA) boreholes. Plotted against downhole temperature, apatite He ages define a broad band from high values at the Earth's surface decreasing to zero by 85°C. The age-temperature profiles are generally consistent with extrapolations of laboratory diffusivity data, but a more quantitative treatment is possible. From the radiogenic production-diffusion equation it can be shown that isothermal apatites will approach a steady-state He age where ingrowth balances diffusion. This equilibrium age, $t_{eq} = (1/15)(D/a^2)^{-1}$, is achieved with a time constant $t^* \sim 1.5\ t_{eq}$. (Wolf *et al.*, 1998). For example, an equilibrium He age of 1 Ma corresponds to $D/a^2 = 2 \times 10^{-15}\ s^{-1}$ and will be achieved after a few Myrs. Note that this statement is independent of any laboratory calibration. Apatites with a He age of 1 Ma occur over the temperature range of about 72–82°C in our boreholes and are almost certainly at age equilibrium; thus we can associate this temperature range with a specific He diffusivity ($2 \times 10^{-15}\ s^{-1}$). On an Arrhenius plot, this point lies on the extension of the regression line through the in-vacuum diffusion measurements on Durango (Fig. 1). This observation provides strong evidence that the laboratory measurements are accurate and applicable in the natural setting.

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