Highly retentive core domains in K-feldspar and their implications for $^{40}$Ar/$^{39}$Ar thermochronology illustrated by determining the cooling curve for the Capoas Granite, Palawan, The Philippines

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K-feldspar from the late Miocene Capoas Granite on Palawan in The Philippines appears to contain highly retentive diffusion domains that are closed to argon diffusion at near-solidus temperatures during cooling of this ~7 km-diameter pluton. This is an important result, for K-feldspar is commonly considered not retentive in terms of its ability to retain argon. Closure temperatures for argon diffusion in K-feldspars are routinely claimed to be in the range ~150–400°C but the release of $^{39}$Ar from irradiated K-feldspar during furnace step-heating experiments in vacuo yields Arrhenius data that imply the existence of highly retentive core domains, with inferred closure temperatures that can exceed ~500–700°C. These high closure temperatures from the Capoas Granite K-feldspar are consistent with the coincidence of $^{40}$Ar/$^{39}$Ar ages with U–Pb zircon ages at ca 13.5 ± 0.2 Ma. The cooling rate then accelerated, but the rate of change had considerably slowed by ca 12 Ma. Low-temperature (U–Th)/He thermochronology shows that the cooling rate once again accelerated at ca 11 Ma, perhaps owing to renewed tectonic activity.

KEY WORDS: dating emplacement, $^{40}$Ar/$^{39}$Ar geochronology, U–Pb SHRIMP geochronology, (U–Th)/He thermochronology, K-feldspar geochronology, closure temperature, South China Sea, hydrocarbon maturity, age of unconformity.

INTRODUCTION

Forster & Lister (2010) demonstrated that, at least in theory, K-feldspar might be considerably more retentive in terms of argon diffusion than present estimates would allow. This is an intriguing result because for two decades, in reference works (e.g. McDougall & Harrison 1999), and in research papers, potassium feldspar is generally considered to have variable but relatively low closure temperatures. Estimates range from: (i) 150 to 300°C (e.g. McLaren et al. 2002; Streepey et al. 2002; Cassata & Renne 2013); (ii) 150 to 350°C (Lovera et al. 1989; Karlstrom et al. 2010); or even (iii) 200 to 400°C (Harrison & Lovera 2013). Lovera et al. (1997) report activation energies in the range 30–77 kcal/mol, with frequency factor a function of a correspondence equation determined by Lovera et al. (1997) for slab-shaped diffusion domains. The least retentive domains in the average K-feldspar ($E$ ~46 kcal/mol; Lovera et al. 1997) are closed to argon diffusion at ~250°C, for a cooling rate of 20°C/Ma. Naturally the size of a diffusion domain is critical in determining retentivity, and the smaller the domain, the less its retentivity. Even for the most retentive diffusion domains, considered by McDougall & Harrison (1999) to be ~100 µm in dimension, closure temperature estimates are 150–320°C for cooling at 20°C/Ma at zero pressure, assuming a slab-shaped diffusion domain.

In part, these estimates vary because individual researchers analyse Arrhenius data differently. Forster & Lister (2010) showed that traditional ‘slope-averaging’ methods (e.g. using least-squares analysis) invariably underestimate the actual values of activation energy and frequency factor used in simulations. Many published values for the diffusion parameters of K-feldspar could thereby have inadvertently underestimated its retentivity.
To test this hypothesis we present a case study demonstrating the analysis of $^{40}$Ar/$^{39}$Ar ages from K-feldspar in Miocene granite, based on geochronology using furnace step-heating experiments. The results imply the existence of highly retentive core domains, with closure temperatures high enough to allow the retention of argon at temperatures above, or just below, the granite solidus. Within uncertainty the ages obtained from these domains coincide with the results of U–Pb geochronology using zircon analysed with SHRIMP. Thermochronology is still possible, because less retentive diffusion domains exist, but different methods need to be applied to interpret the data.

We also simulate the response of a collection of diffusion domains with varying activation energy, and to couple the results with those from the low-temperature thermochronometers provided by ($U$–$Th$)/$He$ geochronology. Of particular interest is the way we can infer a previously unrecognised two-stage cooling history based on K-feldspar alone, and how this could be confirmed by ($U$–$Th$)/$He$ geochronology. We back these conclusions by a modelling study that shows that the fundamental asymmetry principle (FAP) developed by Forster & Lister (2010) can be extended to the analysis of Arrhenius plots for a material with two activation energies, as long as these are very different.

This theoretical model is used to show that minimum estimates for diffusion parameters can be directly inferred from internal sequences in the Arrhenius plot. Previously, it had been thought that estimates for activation energy could be obtained only from the first few data points in the Arrhenius plots, and the value of the activation energy inferred from these points was then routinely applied to a set of diffusion domains calculated for the K-feldspar in question (Lovera et al. 1989, 1993, 1997, 2002). In these previous studies monotonic cooling curves were imposed, constrained using multi-domain diffusion (MDD) models. MDD models published prior to 2012 have extrapolated the activation energy inferred for the least retentive diffusion domains to the entire MDD model.

We first discuss the data, and thereafter the theoretical considerations that allow analysis of Arrhenius plots using the FAP when widely varying activation energies apply. The paper applies these methods by using the eArgon program for iterative forward modelling to construct model crystals with specified parameters for different diffusion domains. $^{39}$Ar release from these theoretical constructs produce results that closely mimic Arrhenius plots derived from the measured data. These analyses yield complex MDD models that are used by the MacArgon program for iterative forward modelling. In this way we were able to constrain the shape of temperature–time ($T$–$t$) histories that allow observed $^{40}$Ar/$^{39}$Ar age spectra to be approximated, assuming release by diffusional processes alone.

**GEOCHRONOLOGY**

The Capoas Granite (Figure 1) is a fresh equigranular to porphyritic biotite granite well exposed around the base of Mt Capoas on Palawan in The Philippines. It displays a textural continuum between K-feldspar phenocrystal-rich (Figure 2) and phenocryst-poor varieties, as well as enclaves of biotite-rich fine-grained granite and K-feldspar phenocrysts that show magmatic flow alignment (Encarnación & Mukasa 1997). Thin-sections of the K-feldspar mineral separate illustrate a variety of microstructures typical of fresh unaltered granite (Figure 3); there is no evidence of deformation. The bulk of the separate (>~60% of the volume) consists of relatively unstructured crystalline material, with occasional subgrains demonstrating <2–3° misorientation. Microcline, as evidenced by twinning, is a minor component. Occasional inclusions of white mica are very small, and volumetrically insignificant. Maps of atomic density variation obtained using Scanning Electron Microscopy (Figure 4) show minor variation in the Na/K ratio from one microcline twin to another, and the late (very minor) introduction of albite in narrow veins and cracks.

**SHRIMP U–Pb geochronology**

A SHRIMP U–Pb zircon study of four samples (PAL33, 34, 35 and 37) of the Capoas Granite as reported in Sug-gate et al. (2014) yielded dates all within the uncertainty of each other, and a pooled U–Pb weighted mean $^{206}$Pb/$^{238}$U date of 13.5 ± 0.2 Ma (95% confidence limit). One of these samples (PAL33, 13.4 ± 0.1 Ma), was also used for a parallel $^{40}$Ar/$^{39}$Ar study, as described below. The zircons from these samples are generally euhedral to subhedral, clear and lightly coloured with cathodoluminescence imaging showing magmatic zoning (see Sug-gate et al. 2014) consistent with crystallisation within felsic to intermediate igneous rocks. The range of individual measurements is plotted in Figure 5.

**$^{40}$Ar/$^{39}$Ar geochronology**

PAL33 was utilised for $^{40}$Ar/$^{39}$Ar geochronology. The sample was dissected so K-feldspar from the groundmass could be analysed separately to K-feldspar derived from crushed phenocrysts. The two K-feldspar aliquots were analysed using the step-heating method in a resistance furnace attached to a VG1200 mass spectrometer. Isothermal heating steps (cf. Lovera et al. 1997; Forster et al. 2014) were included in the step-heating sequence because these appear to have some capacity to drive out extraneous or ‘excess’ argon (i.e. argon that had not been accumulated in the crystal lattice as the result of radiogenic decay, but was present because it had been incorporated in fluid inclusions, cracks and other high diffusivity pathways). Since the release of $^{40}$Ar and $^{39}$Ar is not concordant in these steps, one can safely assume such ‘excess’ argon to have been present. The age spectra for the first 30% of gas released (Figure 6a, b) show the greatest effect of these isothermal steps, with considerable oscillation in age. The first (generally older) step appears to be the most contaminated so this step is referred to as the cleaning step. Age spectra based on the relatively clean second isothermal step rise relatively smoothly (Figure 6c, d), so the method of asymptotes and limits (Forster & Lister 2004) can be applied to provide the age estimates shown. All age data obtained during cleaning steps were therefore systematically eliminated.

Methods and data are further outlined in the supplementary material (Appendices 1, 2). Note that (following existing conventions) all steps on the age spectra are
shown with an error band that is one standard deviation on either side of the mean age obtained. When an average of the age of a group of steps is taken, the age reported is the mean ± two standard deviations of the scatter of the mean age for each step included. No volume weighting of the results is undertaken. This allows an approximation of the 95% confidence limit, in order to compare with the SHRIMP ages.

The youngest ages recorded in the initial steps are in the range ca 11.5–11.2 Ma. Once data from the cleaning steps are discarded, the age spectra can be seen to rise smoothly (and asymptotically) towards limits defined by sub-plateaux at ca 13.0 and ca 12.7 Ma (Figure 6c, d). These sub-plateaux are attained before ~20% gas release has taken place, and they imply a distinct stage in the cooling history of the Capoas Granite.

The age spectra, from the phenocrysts, and from the groundmass, then rise from these sub-plateaux to an average of ca 13.5 ± 0.2 Ma (95% confidence limit, or CL) for the phenocrysts, and ca 13.6 ± 0.4 Ma (95% CL) for the groundmass. There is more scatter in the data from the groundmass, whereas the data from the phenocrysts can be taken to asymptotically rise to a poorly defined plateau at ca 13.5 ± 0.2 Ma (95% CL) and thus provides (following the logic outlined by Forster & Lister 2004) a minimum estimate for the age of the Capoas Granite.

Within uncertainty this estimate is the same as provided using U–Pb zircon geochronology.

(U–Th)/He analysis

Low-temperature thermochronology data on sample PAL33 were obtained using fission track and (U–Th)/He methods (e.g. Farley 2002; Gleadow et al. 2002). Apatite fission track analysis was unsuccessful, as most crystals contained abundant crystallographic dislocations that could not be confidently distinguished from genuine fission tracks (see also Cottam et al. 2013).

Apatite and zircon (U–Th)/He analyses were then carried out using the methodologies detailed by Gleadow...
Six single grain zircon (U-Th)/He analyses (Table 1) yielded a weighted average age of 10.3 $\pm$ 1.1 Ma (95% CL), and five single grain apatite (U-Th-Sm)/He analyses yielded a weighted average age of 8.5 $\pm$ 1.5 Ma (Table 2). Note this latter age does not include apatite analysis no. 8772, which is considered to be an excess He age that is close to the crystallisation age of the granite and older than coexisting zircon (U-Th)/He ages, which record a higher closure temperature than apatite (see below). Such excess He ages and (U-Th)/He age dispersion between grains from the same sample are commonly observed in (U-Th)/He thermochronometry and have been attributed to a number of possible factors such as heterogeneous distribution of U and Th, radiation damage, or grain breakage (e.g. Fitzgerald et al. 2006; Shuster et al. 2006).

The zircon (U-Th)/He age is interpreted as recording cooling of the granite through a closure temperature of $\sim$170–190°C that is estimated on the basis of the relatively young crystallisation age of the granite and its relatively low radiation damage (accumulated alpha-dose of $\sim$2–6 $\times$ 10$^{16}$ a/g; Guenthner et al. 2013). The apatite (U-Th-Sm)/He age is considered to record cooling through a closure temperature of $\sim$70°C (Ehlers & Farley 2003).

**ANALYSIS OF ARRHENIUS DATA**

Forster & Lister (2010) investigated Arrhenius plots produced using $^{39}$Ar release data from simulations of step-heating experiments applied to theoretical materials comprising a multitude of crystalline diffusion domains. These simulations were limited to domains that all had the same activation energy, and the same frequency factor, although the domain size varied. In some cases investigated the multi-diffusion domain (MDD) model had a volume-size variation that was fractal, with different types of fractal also investigated. The aim of this investigation was to establish procedures that should be followed when attempting to infer diffusion parameters from an Arrhenius plot, taking advantage of the fact that the parameters used to construct the MDD simulation were determined beforehand. Because the input parameters were controlled the answer was predetermined, and we were able to confirm mathematical ‘rules’ established by Forster & Lister (2010) that allowed objective analysis of the data and the determination of quantitative (minimum) constraints for the values for two of the diffusion parameters (namely, the activation energy and the frequency factor).

This led to the formulation of the FAP, which states simply that any line drawn to estimate diffusion parameters on an Arrhenius plot must divide the population of points by rank order. A corollary of the FAP is that any valid estimate for the value of the diffusion parameter in question is less than or equal to the actual value. This means that, when comparing different estimates obtained graphically from the Arrhenius plot, the highest value should always be taken.

These two simple rules allow for expedient analysis, and objective determination of minimum constraints for the diffusion parameters. In practice, by using the program eArgon, the analysis can be quite rapid: (i) using the graphic interface to eliminate any estimate that does not divide the population by rank order; and (ii) since any estimate mathematically can be shown to be a minimum estimate, adopting the highest estimates for activation energy and normalised frequency factor.

In review of this paper, it became evident that more explanation was necessary, in particular the nature of decisions that need to be made when dividing the population by rank order. In the simplest terms, dividing by rank order means a line on the Arrhenius plot where points measured before are on or to the right of the line, and those measured after are on the line or to its left. For isothermal sequences, before the line, the sequences are entirely to the right, and after, entirely to the left.
Practical issues in application of the FAP

Conventional MDD analysis considers each diffusion domain to have the same activation energy, with each domain differing only in size and thus varying only in its normalised frequency factor, $D_0/r^2$. In such cases, Forster & Lister (2010) showed that it is a mathematical requirement that analysis of an Arrhenius plot should proceed according to the FAP and that the traditional ‘slope-averaging’ method produces false results, although this is a well-established practice for the analysis of Arrhenius data. In many cases slope ‘averaging’ will considerably underestimate the activation energy inferred using Arrhenius data derived from furnace step-heating experiments. Forster & Lister (2010) also showed that a fractal distribution of diffusion domain size and volume blurred the contribution of individual domains in the Arrhenius plot so that an end-member

![Figure 3 Optical microstructures from a thin-section of an aliquot of the K-feldspar mineral separate used in this analysis: (a–l) from the groundmass; (g–l) from the phenocrysts.](image)
fractal distribution with domains having an activation energy of 75 kcal/mol yields an Arrhenius plot with an ‘average’ slope estimated as 46 kcal/mol, which is the average value reported by Lovera et al. (1997).

Although to apply the FAP certain ‘rules’ need to be followed, practical requirements mean that some latitude needs to be given because estimates that are sensu stricto compliant with the FAP can be based on two points alone resulting in estimates made using the FAP limited by measurement uncertainty. A pragmatic solution is to define an estimate by a ‘least-squares best fit’ for a FAP-compliant line on the Arrhenius plot through three or more FAP-compliant points, to adopt only those estimates that do not include significant outliers. What constitutes a significant outlier is left to the judgment of the individual, noting that including too many points will lead to traditional ‘slope-averaging’ and significant underestimates of the diffusion parameters.

The question then arises as to how to proceed when estimates suggest the existence of domains with widely different diffusion parameters. Is it possible to apply the FAP in a MDD model with ‘core’ domains that are considerably more retentive than inferred ‘rim’ domains, such as is the case in the samples from the Capoas Granite on Palawan? To answer this question further analysis was undertaken, using the step-heating schedule adopted for the Palawan samples, extending the type of simulations reported in Forster & Lister (2010) by modelling the effect of MDD materials with widely differing diffusion parameters (Figure 7). This work demonstrates that it is possible to apply the FAP when distinct regions in the Arrhenius plot can be recognised. The same ‘rules’ are applicable, but note that these apply for step-heating experiments that utilise monotonically increasing sequences of heating steps. However, this exclusion still allows the FAP to be applied to the analysis of Arrhenius data obtained from an experiment using sequences of two or more isothermal heating steps.

The FAP states that an estimate for the diffusion parameters can be obtained by extrapolation of a line defined by joining points on an Arrhenius plot, if and only if, that line defines the population of Arrhenius points by rank order. In other words, the line must be such that measurements made earlier in the step-heating sequence plot on or to its right, whereas measurements made later in the step-heating sequence must plot on the line, or to its left. This statement of the FAP applies at any point in the collection of Arrhenius points used to define the dividing line. Such estimates for the diffusion parameters yield minimum values, that is the estimates made are less than or equal to the actual values of the activation energy and/or the (normalised)
Figure 6  Apparent $^{40}$Ar/$^{39}$Ar age spectra for the K-feldspar phenocrysts (a) and the groundmass (b). In (c, d) the same spectra are shown, but excluding data from the first of the two isothermal steps that appear to act as cleaning steps. The method of asymptotes and limits (Forster & Lister 2004) is then applied to provide the age estimates shown (with the steps used in obtaining the estimates coloured dark green). In (e, f), the FAP is used to define diffusion parameters for the highly retentive core domains (red points) as well as for the least retentive domains (turquoise points). The normalised frequency factor shown is calculated assuming 100% of the volume is occupied by the domain in question, and a correction must be applied when using these data for forward modelling.
frequency factor (i.e. \(D_o/\alpha^2\)) used in the simulation, where \(\alpha\) is the diffusion domain radius, and \(D_o\) is the frequency factor (or maximum diffusivity that could ever be measured).

The first step in the application of FAP (Figure 7a) involves two points on the Arrhenius plot chosen to obey the rule that they locally divide the measurement sequence by rank order. What can then be seen is a sequence of lines that asymptotically increase (or decrease) in slope as we progress across the plot. Each of the lines drawn locally divides the population by rank order, either by joining a step (e.g. 25) at the end of one isothermal sequence (e.g. 23–25) to the step (e.g. 26) at the start of the very next isothermal sequence (e.g. 26–28), or by joining steps (e.g. 9 and 11) at the start of a succession of isothermal sequences (e.g. 9–10 and 11–12), or by joining steps at the end of a succession of isothermal sequences (e.g. 10 and 12). As it turns out, the first three steps (1–3) accurately define the activation energy for the 20\% volume of the least retentive domains, and the last three steps (29–31) accurately define the activation energy for the 80\% volume of the highly retentive 'core' domains. However, these divisors by rank order are so far of local import only. To determine whether they comply with the FAP they must be extrapolated (Figure 7b) and because the two domains utilised in the simulations have such widely differing diffusion parameters, this allows the application of the FAP within specified regions, as shown. The Arrhenius plot can then be divided into a region affected dominantly by retentive core domains, and a region dominantly affected by the unretentive rim domains (Figure 7b).

Considering estimates for the unretentive rim domains (Figure 7b), it is evident that linking points 16–17 and 14–15 causes clear FAP violations, so these can be eliminated leaving other divisors to produce numerically more acceptable estimates (Figure 7c). This leads to a least-squares best fit using points 1, 2, 3, 5, 7, 9 and 11 to estimate the diffusion parameters for the least retentive domains. Again the values obtained (e.g. 45.5 kcal/mol for the activation energy) underestimate the values utilised in the actual simulations, in this case, an activation energy of 46 kcal/mol for the least retentive domains. Figure 7c also shows that the join 20–21 produces a noticeable FAP violation, whereas the join 22–23 (Figure 7d) and the join 25–26 (Figure 7e) produce numerically 'acceptable' divisions by rank order. Since these estimates are minimum estimates, the more retentive values of the two estimates should be the one used. This leads to a least-squares best fit using points 25, 26, 29, 30 and 31 for the highly retentive core domains, noting that the data obtained (e.g. 149 kcal/mol for the activation energy; Figure 7f) still underestimates the actual values utilised in the simulations (150 kcal/mol for the activation energy of the highly retentive core domains).

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### Table 1 Single grain zircon (U–Th)/He data.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Lab. no.</th>
<th>He no.</th>
<th>(^4)He (ncc)</th>
<th>Mass (mg)</th>
<th>Mean (F_T)^a</th>
<th>U ppm</th>
<th>Th ppm</th>
<th>Th/U</th>
<th>[eU]^b ppm</th>
<th>Age (Ma)</th>
<th>Error (±1σ)</th>
<th>Grain radius (µm)</th>
<th>Grain length (µm)</th>
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<td>27043</td>
<td>7.075</td>
<td>0.0103</td>
<td>0.79</td>
<td>585.6</td>
<td>68.9</td>
<td>0.12</td>
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<td>9.4</td>
<td>0.6</td>
<td>44.6</td>
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<td>0.7</td>
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<td>0.7</td>
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<td>0.31</td>
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Weighted average age = \(10.3 \pm 1.1\) Ma (at the 95\% confidence level)

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### Table 2 Single grain apatite (U–Th–Sm)/He data.

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<tr>
<th>Sample</th>
<th>Lab. no.</th>
<th>He#</th>
<th>(^4)He (ncc)</th>
<th>Mass (mg)</th>
<th>Mean (F_T)^a</th>
<th>U ppm</th>
<th>Th ppm</th>
<th>Sm ppm</th>
<th>Th/U</th>
<th>[eU]^b ppm</th>
<th>Age (Ma)</th>
<th>Error (±1σ)</th>
<th>Grain radius (µm)</th>
<th>Grain length (µm)</th>
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<td>0.0078</td>
<td>0.78</td>
<td>13.8</td>
<td>9.9</td>
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<td>0.72</td>
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<td>19.3</td>
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<td>0.83</td>
<td>16.3</td>
<td>8.9</td>
<td>213.3</td>
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<td>16.8</td>
<td>382.0</td>
<td>0.60</td>
<td>32.2</td>
<td>7.3</td>
<td>0.5</td>
<td>33.7</td>
<td>203.2</td>
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Weighted average age = \(8.5 \pm 1.5\) Ma (at the 95\% confidence level)

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^a\(F_T\) is the \(\alpha\)-ejection correction after Farley \etal\ (1996).

^bEffective uranium content = \((U\ \text{ppm} + 0.235\ \text{Th\ ppm})\).

^cAnalysis not included in weighted average age calculation.
Application of the FAP is not constrained to using the few data points at the beginning of a step-heating sequence. The FAP allows Arrhenius data to be divided by rank order at any point in the data sequence, and estimates of activation energy may be made with FAP-compliant estimates that rely on three or more data points.

**Figure 7** Illustration of the application of the FAP with Program eArgon used to simulate an Arrhenius plot, using a model crystal with known diffusion parameters and volume ratios as shown (a). Two regions are delineated on the Arrhenius plot. Many local divisors by rank order (a) fail when extrapolated (b–c). Other extrapolations are numerically acceptable within the defined regions (c–e). When application of the FAP leads to two divisors by rank order (d, e), the maximum slope should be chosen.

**Diffusion parameters based on fractional loss of $^{39}\text{Ar}$**

Diffusion parameters can be inferred from the percentage volume of $^{39}\text{Ar}$ released during step-heating experiments, using methods pioneered by Lovera et al. (1989, 1993, 1997, 2002). Note that although the age data from...
the first isothermal cleaning step are discarded, diffusion data obtained using $^{39}\text{Ar}$ are internally consistent throughout the entire step-heating sequence and the release of $^{39}\text{Ar}$ from all heating steps is included. Its release depends only on the kinetics of the diffusion that allow its migration.

Neutron irradiation is required to produce $^{39}\text{Ar}$ so this isotope is sourced only in consequence of its diffusion and escape from the lattice. In contrast, released $^{40}\text{Ar}$ can include extraneous argon, for example, derived from fluid inclusions, or $^{40}\text{Ar}$ caught in relative fast diffusion pathways as it attempted to diffuse back into the crystal lattice from the ambient pore fluid. Consequently, the release of $^{40}\text{Ar}$ can include both argon from the lattice as well as from these extraneous sources, and age data based on the $^{40}\text{Ar}/^{39}\text{Ar}$ ratio can show both concordant as well as discordant trends.

Note that plots of the Arrhenius data have been prepared assuming a spherical geometry (Figure 6c, f) to allow comparisons between different datasets. Parameters inferred using spherical geometry should not be exchanged with parameters inferred using other geometries (see Forster et al. 2014).

The measured Arrhenius data allow classification of three distinct domains (Figure 8). The initial steps involve relatively low activation energies, estimated as $\sim 54–55$ kcal/mol. Thereafter there is a zone of transition that may involve a range of diffusion radii. Finally, in the highest temperature steps, slopes estimated by locally applying the FAP, activation energies are in the range 92–110 kcal/mol, about twice as large as those that apply in the initial steps. Figure 6(e, f) shows estimated normalised frequency factors for these domains, with closure temperatures calculated for zero pressure and a $20^\circ\text{C}/\text{Ma}$ cooling rate. The least retentive domains close at $304–309^\circ\text{C}$ according to this simple calculation, while the retentive core domains close at $547–588^\circ\text{C}$. However, there is a problem with estimating closure temperatures.
in this way since the calculation assumes that 100% of the volume is occupied by the domain in question. For the least retentive domains, to correct for the volumetric effect, the normalised frequency factor must be increased, and less retentive values assumed (Table 3) reducing the closure temperature for the least retentive domains to 273–278°C.

Figure 8 provides a useful classification. The least retentive domains can be seen to be responsible for the steps with the greatest oscillation in age data. The sub-plateaux ages at ca 13.0–12.7 Ma are recorded by the median retentivity domains. The age data from the highly retentive core domains is more scattered in the groundmass K-feldspar but the average age for both phenocrysts and groundmass is the same. These retentive core domains account for >70–75% of the volume of gas released and the points (corresponding the age steps coloured dark green in Figure 8) define the alignments on the Arrhenius plot from which estimates for extraordinarily high activation energy can be obtained.

Simulation and modelling

The final step in this process was to undertake a modelling and simulation study. The Arrhenius data were first used to produce r/r0 plots (Lovera et al. 1989) that allow a ready correspondence between parts of the Arrhenius plot and parts of the apparent age spectrum (Figure 9). This correspondence suggests that the bulk of the gas measured was derived from the highly retentive core domains. Using these steps, the timing of the onset of rapid cooling after granite emplacement can be estimated.

With the volumetric ratios established we used equations for fractional loss based on specific geometries to forward model the pattern of 39Ar gas release, applying the same step-heating schedule used during the in vacuo experiments that produced the measured age spectra (Figure 9). These methods (including code, and equations used in the modified version of the eArgon program utilised) are summarised by Forster et al. (2014).

The eArgon program was used to forward model the Arrhenius plot and the r/r0 plot that would result based on model crystals comprising collections of diffusion domains with a specified activation energy and a normalised frequency factor (Figure 9). Fractal size-volume relations were imposed for the two domains with an activation energy of 55 kcal/mol. In terms of numerical analysis, this assumption implies these less retentive domains are not perfectly spherical, but quite rough in their geometry, with fractal outlines. In terms of the highly retentive core domains, surprisingly, the fit was markedly improved by considering at least two domains, with approximately the same retentivity and similar volumetric ratios. The data used in the final model illustrated in Figure 10 are shown in Table 4; the fit is greatly improved by the addition of this complexity.

Manually iterating volumetric ratios is tedious so the eArgon program was improved by incorporating iterative forward modelling based on Monte Carlo simulations. Interestingly, there were two valid applications of the FAP (Figure 9a, c), both of which divide the population of Arrhenius points by rank order, one yielding an activation energy of 92 kcal/mol and the other yielding an activation energy of 134 kcal/mol. Both choices of activation energy allowed accurate approximation of the Arrhenius data, but it was not possible to replicate the r/r0 plot with the less retentive value for the activation energy of 92 kcal/mol (Figure 9b). The higher value of the activation energy (134 kcal/mol) did allow an accurate replication of the r/r0 plot, however (Figure 9d). This value was therefore assumed in constructing the model crystal used in the MacArgon program to iteratively forward model the effects of arbitrary pressure–temperature (P–T) histories.

The MacArgon program (modified from code described by Lister & Baldwin 1996) was further developed to allow interactive modelling of the effect of parametric variation of an arbitrary P–T history. The variation of T–t curves is shown in Figure 10. An initial ambient pressure of 3 kbar was assumed, consistent with estimates of the depth of emplacement of the Palawan granitoids (cf. Zulauf et al. 1997). The calculation is not particularly sensitive to the assumed ambient pressure, but note that the activation volume used was 15 cm3, and this accentuates the increase in retentivity with increasing pressure (cf. Forster & Lister 2013). The interactive use of MacArgon is particularly efficient in terms of effort, for the effect on the age spectrum of any change in the T–t history is interactively provided. One can first model the effect of the earlier part of the temperature history, obtain the best fit (Figure 10a, b) and then move to lower temperature segments of the history to repeat the exercise (Figure 10c, d). In this way an estimate for the complete cooling history is obtained.
T–t points have been added that show the estimated closure temperature for helium diffusion in conjunction with the ages obtained using (U/Th)/He thermochronometry for zircon and apatite. A two-stage cooling curve is therefore confirmed (Figure 10e). Note that this two-stage cooling curve is not dependent on the assumption of highly retentive core domains but is required by the variation in the K-feldspar data itself. To demonstrate that this is correct we carried out a traditional analysis of the Arrhenius plot based on ‘averaging’ the slope of the least retentive part of the dataset (Figure 11a), and then extrapolated the activation energy obtained to all other domains. This analysis yielded a simple model crystal based on two diffusion domains, but (see small dots in Figure 11a) is incapable of accurately modelling diffusional release at the high-temperature end of the Arrhenius sequence. This model for inverse modelling yielded reasonable fits at the start and end of the age spectrum (Figure 11b) but it did not allow a fit to its overall form, particularly in respect to intermediate segments of the age spectrum. The T–t variation implied by this traditional analysis (Figure 11c) again requires a two-stage cooling history, with the detail of the second stage constrained by the (U–Th)/He thermochronological data.

The two-stage cooling history is required by the interdependence of retentivity estimates for the different diffusion domains in K-feldspar. This can be further demonstrated using a simple model based on estimates for three diffusion domains (Figure 12). This method requires closure temperatures to be iteratively estimated based on the assumed form of the cooling curve, and since these depend on cooling rate, several iterations may be required. Again, once points are added that show the estimated closure temperature for helium diffusion in conjunction with the ages obtained using (U–Th)/He thermochronometry for zircon and apatite, a two-stage cooling curve is required.
Figure 10 Forward modelling and simulation using diffusion parameters inferred using the eArgon program, as listed in Table 4. The MacArgon program was used to forward model the effect of cooling curves (a) considering the range shown. The optimal fit to the higher temperature part of the age spectrum (b) is the thicker path. The cooling curve (c) was then adjusted until an optimal fit was obtained for the lower temperature part of the age spectrum (d). The optimal fit to the temperature–time path (e) produces a reasonable match to the age spectrum (f) from the groundmass K-feldspar.
Although this method approximates the cooling curve by considering ‘representative’ domains, it requires the form of the cooling curve to be known. Newtonian cooling of the granite body was assumed, i.e. an exponential decrease with time in respect to the difference between the temperature in the body in comparison with that determined by an ambient geotherm.

**DISCUSSION**

The cooling curve presented here is based on two assumptions, namely that: (i) microstructure remains constant during the evolution of the sample in the natural environment; and (ii) diffusion is the only mechanism releasing argon into the mass spectrometer.

Each of these two assumptions has significant drawbacks, and caveats related to the limitations posed by these assumptions need to be considered when attempting to draw geological inferences based on the results, as discussed below.

**Effect of microstructural change**

It is unlikely that any microstructure remains constant throughout the history of a rock, and this is true even in granite that has cooled rapidly without deformation (e.g. Villa 2006; Parsons & Lee 2009; Putnis 2009; Putnis &

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<th>55</th>
<th>55</th>
<th>134</th>
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**Figure 11** Forward modelling and simulation using diffusion parameters inferred using traditional ‘slope-averaging’ methods, extrapolating the low activation energy that can be inferred using the first few points on the Arrhenius plot to all other domains (a). The MacArgon program was used to forward model the effect of cooling curves (b) until a fit was obtained. Even though the fit was poor, the inferred cooling curve (c) still required two stages. Note that, for the activation energy used here, increasing the cooling rate by a factor of 10 will lead to elevation of the calculated closure temperature by merely ~40°C. However, this model cannot explain the coincidence of the U-Pb and the 40Ar/39Ar ages.
John 2010). This has led to controversy, noting contrasting views expressed in Villa (2013) and Harrison & Lovera (2013). Nevertheless, we must conclude that MDD modelling limits itself by the assumption that microstructure remains constant, at least in MDD models as they are currently applied.

Does microstructure matter? It is not at all certain that microstructural change prevents systematic analysis of an age spectrum, since in the final microstructure it is typically the less retentive parts that have been created and/or modified on the cooling path by processes such as (fluid-assisted) recrystallisation and grain boundary migration. Therefore, the modification of a microstructure need make relatively little difference in terms of the $P-T-t$ history that can be inferred.

Consider the effect of argon release as the result of grain boundary migration in the natural environment. This will lead to an abrupt age transition in the age spectrum that cannot be effectively simulated. Yet the relict microstructure continued to be subject to the same $P-T-t$ history, and thus coexisted with the newly created/modified microstructure. For this reason, in constraining the range of possible $P-T-t$ histories, the most important part of the age spectrum to match is the early part of the release from the least- and median-retentive domains. Ensuring coincidence with the earlier released parts of younger age sub-spectra ensures that the model reflects the effect of diffusional release from these modified or newly created aspects of the microstructure.

Thus, we arrive at an important qualification of the statement above. While MDD models need not exactly replicate the abrupt transition to the oldest part of the age spectra, they must not neglect the match with the earlier part of the age spectrum, i.e. it is important to match that part that results from release of argon from less retentive domains. We thus disagree with the reviewer who asserted that an ‘intermediate solution between A and B (in Figure 10a) ... produces a model age spectra that is apparently as good as any in (Figure 10c).’ This is not correct. None of the single-stage cooling paths offers a good match to the early part of the age spectrum.

Significantly, the lower $T$ parts of the age spectrum could be matched only once the option of a two-stage cooling history had been introduced (Figure 10d, e). In other words, the K-feldspar data alone require a two-stage cooling history.

The $^{39}\text{Ar}/^{39}\text{Ar}$ age spectra provide no information on the precise form of the cooling curve past $ca$ 12 Ma. To further constrain the two-stage cooling path ($U$–$Th$)/He data are then necessary. Consistent with the above observations, these data require the onset of renewed cooling at $ca$ 11 Ma, and an acceleration of cooling rates as would be expected in the footwall of large normal faults that are inferred to exist along the coast of Palawan (Franke et al. 2011; Walla et al. 2012; Hall 2013). Cooling of the Capoas Granite appears to have considerably slowed by $ca$ 12 Ma, suggesting that the rim of the granite plug had reached temperatures expected in consequence of the ambient geotherm by that time. An ambient geotherm in the range 25–30°C/km implies a depth of emplacement at $7-9$ km.

We note that this two-stage history makes sense in the context of independent geological data. Renewed tectonism readily explains the Late Miocene unconformity in the adjacent basin, and the influx of sediment that deposited on the unconformity from $ca$ 10 Ma (personal communication, Prof. Robert Hall). Steuer et al. (2013) note the oldest sediments that lie on this unconformity deposited at 9.2 Ma. The initial asymptotic decrease in temperature towards $\sim 220$°C allows an inference of ambient conditions, implying (as above) a depth of emplacement of $7-9$ km, and ample latitude in terms of the requirement for large volumes of material to have subsequently been made available for erosional denudation.

Break-down reactions in the mass spectrometer

A reviewer was concerned that the minerals were breaking down (or melting). Indeed, it is true that the Arrhenius data reported in this paper have been calculated by applying fractional loss equations on the basis of the percentage release of $^{39}\text{Ar}$ from temperature controlled step-heating experiments in vacuo by diffusion alone.

These data have been taken as constraining the diffusion of $^{39}\text{Ar}$ from the crystal lattice but only if this assumption were correct can we infer the existence of highly retentive core domains in K-feldspar. Perhaps we should also consider the option that some other process is mimicking (or complicating the kinetics of) diffusional release, e.g., the advance of sub-microscopic melting fronts in the K-feldspar at high temperatures during the step-heating experiment, or the effects of non-Fickian diffusion. The advance of sub-microscopic melting fronts does not prevent diffusive loss, although it may...
modify the release path. It is likely that argon would be lost by diffusion before melting occurred.

Non-Fickian diffusion says nothing more than that simple equations do not apply, to which claim we can offer no comment except that Fickian diffusion does not need to be limited to a single species model (cf. Villa 2013). All that numerical models need is reasonable numerical accuracy, and from this viewpoint it is interesting that we seem to be able to reasonably approximate these experiments using only simple diffusion equations. Since we can predict the percentage of argon released during a heating step of known duration, then perhaps it is reasonable to assume that diffusion also occurs in nature.

We note that argon diffusion inferred in nature appears to coincide with the onset of dislocation climb, as implied by microstructures that suggest dynamic recovery (Forster & Lister 2013). However, exceptionally high values for the frequency factor can be inferred for the retentive core domains and Forster & Lister (2013) demonstrate compression of the temperature interval during which such materials degas. Argon release from the retentive core domains is thus likely to precede the final disintegration of the lattice that occurs during melting. We emphasise that the advance of sub-microscopic melting fronts during heating in an ultra-high vacuum will thus not prevent diffusional release of argon from within the (almost ready to disintegrate) lattice.

Comparing data from K-feldspar with zircon

In the natural environment the same argument as above holds, in reverse. The highly retentive diffusion parameters mean that K-feldspar should be expected to retain some argon even in a magmatic environment, after K-feldspar mineral grains crystallise. In this case not only will K-feldspar core domain ages record the earliest stages of the cooling history; but occasionally (depending on the relative roles of cooling vs depressurisation in triggering zircon growth) K-feldspar ages may predate ages obtained using U–Pb measurements. There are thus limitations on the conclusions that can be drawn based on coincidence with ages obtained using U–Pb zircon geochronology, as well as limitations on the conclusions that can be drawn based on any lack of coincidence.

It can be difficult to interpret zircon data as requiring a single magma emplacement age, e.g. Schoene et al. (2012) showed that crystallisation in a ballooning pluton (in Adamello, Italy) endured for ca 0.3 Ma. There is a natural spread of U–Pb zircon ages that reflects the time taken to inflate and construct a magmatic edifice.

Another difficulty is caused when fluid-catalysed recrystallisation leads to grain boundary migration within the newly formed K-feldspar crystal, leading to in situ replacement and regrowth. This means that, independent of any coeval diffusion process, regrowth of K-feldspar core domains can reset ages to be younger than the time at which the original K-feldspar crystals grew. Sánchez-Muñoz et al. (2012) described microstructures in granitic pegmatite that formed during cooling. Such microstructural evolution may destroy or modify retentive core domains. The median-retentive domains might thus record the time of such resetting, and thus become useful indicators of progress.

Other difficulties arise when excess argon is present (i.e. 40Ar incorporated into the lattice from the pore fluid in the natural environment, and released into the mass spectrometer during the highest temperature part of the step-heating experiment) (Foster et al. 1990). Such effects are routinely evident when calcic plagioclase is measured; potassium concentrations in such minerals are low and the accuracy of an age determination can be overwhelmed. Calcium produces both 39Ar and 36Ar in the reactor during fast neutron irradiation. Caution is needed in respect to ages with high Ca/K ratios, since these may reflect the influence of gas released from minor plagioclase at the end of the step-heating experiment.

A need for further comparative analysis

The inferred cooling rate for the 7 km-diameter stock that defines the Capoas Granite was initially very much faster than expected. The outcrop of the Capoas Granite is similar to the granite body exposed in Mt Kinabalu, ~600 km to the southwest. There is a rapid increase in surface elevation, rising from sea-level to ~960 m within less than ~4 km distance. The cooling rate during exhumation and cooling of the Kinabalu Granite is estimated at 120°C/Ma (Cottam et al. 2013). More rapid rates were estimated for initial cooling of the Shivling leucogranite in the High Himalaya immediately beneath the South Tibetan Detachment (175–350°C/Ma; Searle et al. 1999). This 1–2 km-thick laccolith had cooled below 400°C in about one million years from the time of its initial crystallisation. In comparison, for a 9 km-thick sill-like body, the Falkenberg Granite (Oberpfalz, Germany) required ca 6 Ma before it cooled below 400°C (Zulauf et al. 1997).

Cooling rates can be very rapid for samples near the rim of a pluton. Nabelek et al. (2012) showed that the solidification time can be prolonged because ‘wall rocks become more insulating as temperature rises’ but these authors also showed that temperatures near the rim of the batholith drop below 400°C within 0.1–0.2 Ma of the time when the pluton crystallised. Modelling the cooling of a high-level granite body (5 km-diameter, 2 km thick, with its top level 3 km from the surface) produced cooling rates initially as high as ~1500°C/Ma, but larger granite bodies deeper in the crust would cool more slowly. The Capoas Granite has a typical granitic texture, and does not exhibit characteristics of a hypabyssal intrusion such as that above. Yet cooling rates inferred from the K-feldspar data are high, and perhaps are higher than should have been expected. This raises questions as to the effect of mechanical forcing of the stock upwards immediately after emplacement, since this process would explain the odd nature of the start of the cooling path implied by the simulation illustrated in Figure 10e. Nabelek et al. (2012) found that the thermal conductivity of rock decreased at temperatures around the solidus, so at those temperatures the wall rock of the Capoas Granite will have hindered cooling. Granite emplacement thus appears to be self-insulating.
Continual episodes of intrusion as noted by Schoene et al. (2012) would also slow overall cooling for the first ca 0.5 Ma there after cooling would accelerate towards rates as inferred in this study if the stock were then ‘punched’ upward through the adjacent cooler country rock.

The rapid cooling rates inferred for the Capoa Granites suggest further experiments are needed, along a traverse from its core to its rim. Only if initial cooling rates were slower will it be possible to readily discriminate when in the cooling curve K-feldspar core domains have individually closed, and to provide independent proof as to the inferred high retentivity of the core domains. The uncertainty in the age determinations based on the U–Pb zircon data mean that there is overlap with the uncertainty in the age determinations based on the high-temperature $^{40}\text{Ar}/^{39}\text{Ar}$ data, and this preclude use of these data to independently choose between: (i) coincidence with the SHRIMP ages because the K-feldspar core domains are highly retentive (see inverted Arrhenius model in Figure 9c, d and forward modelling in Figure 10); or (ii) coincidence with the SHRIMP ages because of the effect of very rapid cooling (see inverted Arrhenius model in Figure 11a and the forward modelling in Figure 11b, c).

Figure 11 was constructed using a ‘classic’ averaging approach to the analysis of the Arrhenius data, extrapolating the relatively unretentive diffusion parameters inferred from the initial data points. The inferred closure temperature for the retentive core domains then becomes a mere $\sim 370 ^\circ \text{C}$ for a cooling rate of 700°C/Ma allowing closure 0.2 Ma after the time of initial emplacement. Taking the initial cooling rate as 1400°C/Ma implies crystallisation at 13.9 Ma, which is outside the limit of uncertainty of the estimated age of 13.5 ± 0.2 Ma. While the uncertainty mentioned above, means that this alternative scenario cannot be completely dismissed, we further note the relatively poor fit between the model simulation and the observed age spectrum. As demonstrated, even with a two-stage cooling history, it was not possible to match the detail in the observed age spectrum. The observed age spectrum cannot be more exactly matched unless highly retentive core domains are assumed (Figure 10).

**K-feldspar can retain argon above the solidus**

K-feldspar has been shown to be able to retain argon while immersed in a magma. Gillespie et al. (1983, 1984) noted this phenomenon, while Renne et al. (2012) used barium diffusion fronts to quantify the period of immerssion. This paper in turn demonstrates the highly retentive diffusion parameters that make this phenomenon possible.

The implication is that sanidine such as that in the Fish Canyon Tuff (Renne et al. 2010; Phillips & Matchen 2013) would be capable of recording ages relict from the time of K-feldspar growth, as well timing the rapid cooling that occurs after eruption (Renne et al. 2012). A similar comment can be made in respect to sanidine from the Younger Toba Tephra (Mark et al. 2013).

The diffusion parameters for K-feldspar recorded in this paper mean that highly retentive core domains also impact on the question of inheritance, e.g. the age of 20 ± 1 Ma (Copeland et al. 1990) for the Manaslu Granite while other work (Deniel et al. 1987; Guillot et al. 1994; Harrison et al. 1999) suggest it intruded as sills in magmatic pulses starting at 25–22 Ma and continuing for 6–9 million years thereafter. K-feldspar grown during earlier magmatic pulses may well be able to retain vestiges of its original age, a phenomenon which is well possible if core domains are as retentive as some of the diffusion data reported here can be taken to imply. This is not excess argon _sensu stricto_, although it has been referred to as such. It is argon relict from decay in the minerals grown at an earlier stage of the rock’s history.

**Practical implications of this work for argon geochronology**

This paper demonstrates that estimates of activation energy can be made internally within an Arrhenius data sequence, and that such an analysis can yield estimated values for the activation energy and frequency factor that are well beyond the range documented by previous workers (e.g. Lovera et al. 1997, 2002). This is important for many previously published furnace step-heating $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology experiments using K-feldspar that also contain internal sequences in Arrhenius data, which can similarly be taken to imply the existence of highly retentive ‘core’ domains, e.g. Knapp & Heizler (1990) in K-feldspar from the Maria Fold and Thrust Belt, Lovera et al. (1993) for the classic MH10 K-feldspar; or Warnock & Zeitler (1998) for K-feldspar from the German KTB deep drill hole. Such internal analysis of an Arrhenius sequence should therefore become routine.

On the practical side, this paper demonstrates K-feldspar can be sufficiently retentive so as to allow direct dating of processes that reduce the dimensions of diffusion domains, e.g., cataclasised and/or recrystallised K-feldspar in fault rock and/or mylonite. Forster & Lister (2009) were thereby able to date the formation of crushed feldspar in 25 Ma north-sense shear zones that overprinted the south-sense 35–30 Ma South Cyclades Shear Zone (Ios, Cyclades, Greece). Forster et al. (2014) duplicated this success, dating the timing of crushing of K-feldspar in greenschist facies ductile shear zones exposed in the Wyangala Batholith, NSW, Australia. Analysis of the latter dataset showed that deformation of K-feldspar in a shear zone is capable of pervasively modifying the diffusion parameters, and that it is this process that makes it possible to directly date the timing of movement.

These are important developments in the methodology of $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology, but we need to clarify the nature of these highly retentive core domains. In particular, we need to better understand how microstructural processes modify the core domains during deformation and metamorphism. In our future research it is now key that we determine the role of fluid ingress in triggering grain boundary migration, since we know cases that involved _in situ_ recrystallisation, and when this occurs, this process appears to be able to eliminate and regrow the core domains, leading to relatively youthful ages.
CONCLUSIONS

The Capoas Granite on the northwest coast of the island of Palawan in the Philippines, was dated with U–Pb zircon using SHRIMP and by 40Ar/39Ar K-feldspar geochronology using temperature controlled step-heating experiments involving isothermal cleaning steps. The 40Ar/39Ar age spectra show considerable scatter when all steps are included, but if the data for the (contaminated) cleaning steps were omitted, the age spectrum simplifies, and allows application of the method of asymptotes and limits as described by Forster & Lister (2004). The ages obtained are within uncertainty of those obtained using U–Pb dating of the zircon crystallisation. This is consistent with the interpretation of the Arrhenius data that K-feldspar from the Capoas Granite contains diffusion domains that close at temperatures 500–700°C for rapid cooling rates, allowing direct dating of mineral growth and/or the timing of magma emplacement.

Our analysis of the Arrhenius data confirms that K-feldspar can be considerably more retentive of argon than previous estimates would allow, and in consequence that there need be little difference between U–Pb ages measured from zircon, and 40Ar/39Ar ages measured from highly retentive ‘core’ diffusion domains in K-feldspar. In these circumstances, direct estimates of granite emplacement ages are possible, especially if rapid cooling then follows. The remaining part of the cooling curve can also be determined using data from other less retentive domains in the same mineral grains.

Forward modelling based on MDD inferred from systematic and self-consistent analysis of Arrhenius data derived from release of 39Ar from K-feldspar during step-heating require a two-stage curve for cooling of the Capoas Granite. After some time, very rapid cooling took place, but with an asymptotic decrease in cooling rate implying the depth of emplacement was 7–9 km. The granite had thus cooled towards ambient temperatures by ca 12 Ma, and we can infer a period of tectonic quiescence thereafter. The (U–Th)/He data allow inference that exhumation then once again commenced, leading to an acceleration of the cooling rate. Cooling continued at an averaged rate of ~70–75°C/Ma for another 4.2 million years until 8.5 ± 0.4 Ma, by which time ambient temperatures had declined to ~70 ± 5°C.

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SUPPLEMENTARY PAPERS

Appendix 1 40Ar/39Ar Geochronology analytical procedures (Capoas Granite).
Appendix 2 40Ar/39Ar geochronology data: Sample 11, PAL30a, K-feldspar.
Appendix 3 SEM data: on the Hitachi 4300 FESEM.

DISCLOSURE STATEMENT

No potential conflict of interest was reported by the authors.

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