Radiation damage control on apatite (U-Th)/He dates from the Grand Canyon region, Colorado Plateau

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ABSTRACT

Individual detrital apatite grains from the Esplanade, Coconino, and Moenkopi Formations in the Grand Canyon region of the Colorado Plateau yield (U-Th)/He dates from 104 to 5 Ma. The range of dates within each unit far exceeds analytical uncertainty, but correlates with both He concentration [He] and effective U concentration [eU]. These dates are all significantly younger than the sandstone units, indicating partial to complete He loss following deposition. Recently published laboratory diffusion data suggest that He retentivity in apatite increases with radiation damage. Forward models predict that the consequences of this effect will be manifested most clearly as a correlation between (U-Th)/He dates and the [He] and [eU] in suites of apatites that (1) are characterized by a large span of [eU], and (2) had thermal histories in which sufficient time elapsed for the apatite He diffusion kinetics to diverge prior to reheating and partial resetting. Apatites in the sedimentary units investigated fit these criteria. Using geologically reasonable deposition, burial, and unroofing histories, simulations that include the effect of radiation damage on apatite He retentivity can reproduce the observed distributions of apatite dates and correlations with parent and daughter concentrations. These results suggest that a span of (U-Th)/He dates positively correlated with [eU] may provide important information regarding a sample's thermal history.

Keywords: (U-Th)/He, apatite, radiation damage, Colorado Plateau, He diffusion kinetics, thermochronometry, Grand Canyon.

INTRODUCTION

Understanding He diffusion kinetics is critical for accurate interpretation of apatite (U-Th)/He thermochronometry. Based almost exclusively on laboratory diffusion measurements, (U-Th)/He apatite (AHe) dates are inferred to record cooling from ~70 to ~30 °C, making them uniquely suited to quantifying the cooling of rocks as they are exhumed through the upper 1–3 km of crust (Wolf et al., 1998; Farley, 2000). However, a recent study of He diffusion kinetics in apatite (Shuster et al., 2006) and previous results for Durango apatite (Farley, 2000) suggest that the accumulation of He and associated radiation damage may increase the He retentivity of apatite grains. The geological relevance of this phenomenon has not yet been clearly documented in an AHe data set, although the possibility has been raised in prior studies (Crowley et al., 2002; Lorencak et al., 2004; Green and Duddy, 2006). Here we present new single-grain AHe data from the Grand Canyon region of the Colorado Plateau that display variations in dates that can be explained by the effects of radiation damage on He mobility.

DETRITAL AHe DATA

The Grand Canyon region, on the southwestern portion of the Colorado Plateau, is characterized by Proterozoic basement and a nonconformably overlying cratonic section of Paleozoic and Mesozoic strata (Fig. 1). Limestone of the Permian Kaibab Formation forms the modern plateau surface, discontinuously overlain by remnants of fluvial sandstone of the Lower Triassic Moenkopi Formation. Stratigraphic and apatite fission-track data suggest that, following deposition, these upper Paleozoic and lower Mesozoic units were buried by 2.7–4.5 km of Mesozoic and Cenozoic strata before erosional unroofing (e.g., Hunt, 1956; Naeser et al., 1989, 2001; Dumitru et al., 1994; Kelley et al., 2001).

We acquired AHe dates on individual detrital apatite grains from four sandstone samples north of the Grand Canyon (Fig. 1). We analyzed Permian Esplanade and Coconino sandstone samples from a single stratigraphic section to test for geological consistency in the results. Two Moenkopi sandstone samples were analyzed to establish the reproducibility of data from nearby locations. Single crystals of apatite were selected based on morphology, clarity, and lack of inclusions using a binocular microscope with crossed polars. Backscatter, cathodoluminescence, and electron microprobe studies of representative detrital grains were used to characterize the apatites from each sandstone unit. (See the GSA Data Repository[1] for additional sample details and analytical methods.)

Each of the four samples yielded a broad span of AHe dates that are significantly younger than the sandstone units, indicating postdepositional He loss (Table DR1; see footnote 1). The Esplanade and Coconino samples yielded dates from 69 to 19 Ma (n = 7), and 63–35 Ma (n = 7), respectively (Figs. 2A, 2B). The two Moenkopi
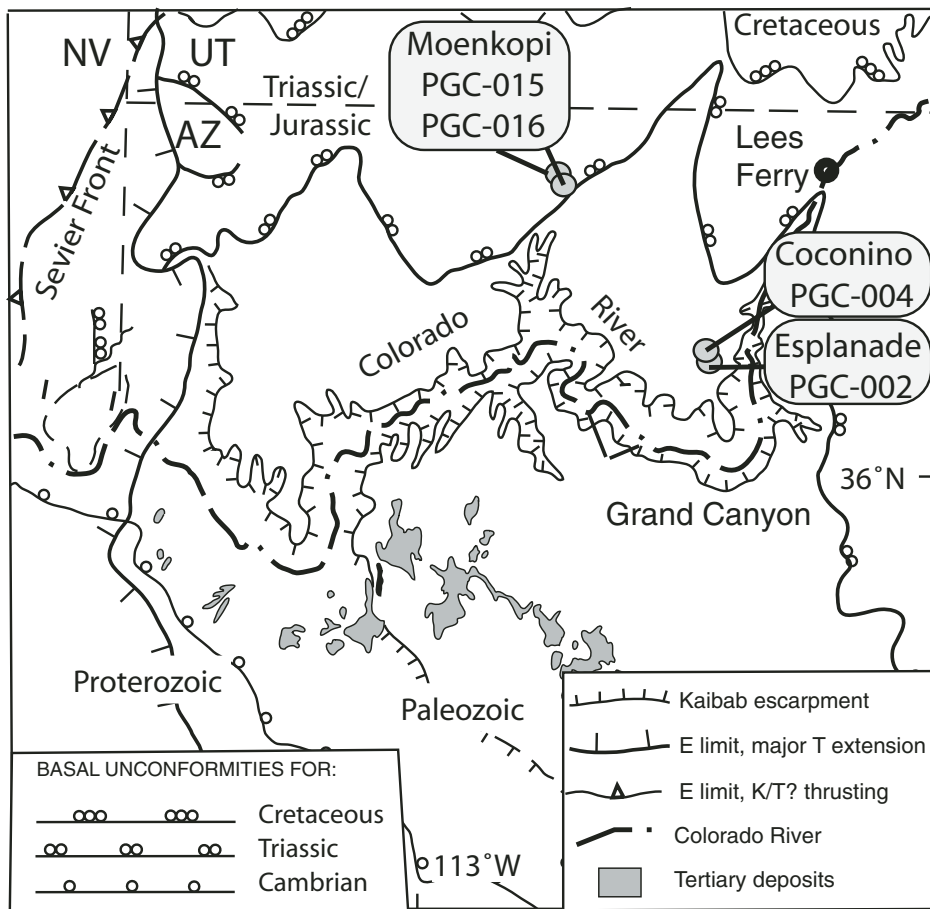
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Figure 1. Simplified geological map of Grand Canyon region of Colorado Plateau with sample locations.

samples yielded dates from 77 to 5 Ma (n = 20) and 104–13 Ma (n = 13) (Figs. 2C, 2D). The variation in dates is far greater than can be accounted for by analytical uncertainty (~8%, 2σ), and the dates are not correlated with crystal size. However, data from all four samples show an obvious visual correlation between AHe date and both effective U concentration [eU] and He concentration [He], where [eU] weights the decay of the two parents for their α productivity and is computed as U + (0.235 · Th) (Figs. 2A–2D).

Correlation between [eU] and [He] is anticipated for a suite of uniform apatites with variable [eU], because higher [eU] apatites have a higher He production rate. However, the AHe dates of grains with uniform He diffusion kinetics and thermal history should be invariant with [eU]. Thus, commonly assumed apatite He diffusion kinetics (Farley, 2000) cannot explain the distributions of dates in our samples regardless of the thermal history. Although the F/Cl ratio has been proposed to influence apatite helium diffusivity (Warnock et al., 1997), the F/Cl ratio does not correlate with [eU] in our detrital apatites (Table DR2; Fig. DR1 [see footnote 1]) or with apatite He diffusion parameters and AHe

dates in a broader study of apatite He diffusivities (Shuster et al., 2006). These patterns lead us to consider whether radiation damage–enhanced He retentivity suggested by laboratory diffusion measurements (Shuster et al., 2006; Farley, 2000) can explain our results.

RADIATION DAMAGE CONTROL ON AHe DATES

The radiation damage trapping model incorporates He diffusion kinetics that evolve with temperature and [He] (Shuster et al. 2006). We simulated distributions of dates expected for detrital grains of uniform diameter (100 µm) characterized by an arbitrary but reasonable range of [eU] (2–100 ppm) and (U-Th)/He provenance dates (AHe date of the apatite when deposited, AHe_{dep}) from 5 to 100 Ma. We first consider as a simple example that may be broadly relevant to our samples, monotonic heating from 0 °C at 250 Ma to peak temperature (T_{pk}) at 125 Ma during burial, with subsequent monotonic cooling during exhumation to the surface by 0 Ma (Fig. 3A). In these simulations, the key parameters that control the predicted date are [eU], AHe_{dep}, and T_{pk}. We display the results as

plots of AHe date versus [eU] and [He] (Figs. 3B, 3C), and for comparison show the results for a model that assumes conventional Durango apatite He diffusion kinetics (Fig. 3D).

The distribution of AHe_{dep} must be considered, because the apatites enter the system (in this example, at 250 Ma) with different dates and thus different He diffusion kinetics. An apatite characterized by a short period of He accumulation prior to burial (younger AHe_{dep}) will be less radiation damaged and have a lower effective closure temperature (T_{ec}) than an apatite characterized by the same [eU] but longer initial He accumulation duration (older AHe_{dep}). Thus, during an episode of partial resetting, the date of the former apatite will be more reset than the latter. For this reason we model a suite of apatites with a span of AHe_{dep} (5–100 Ma) and plot the results for each thermal history as a fan (Figs. 3B–3D). The lower bound represents AHe_{dep} of 5 Ma, and the upper bound AHe_{dep} of 100 Ma.

A T_{pk} of 20 °C is insufficient to cause He loss from any of the apatites, such that the final distribution of dates mimics the provenance distribution, and is independent of [eU] (Fig. 2B). For T_{pk} of 60 °C, apatites with the lowest [eU] (least radiation damaged and lowest T_{ec}) undergo complete He loss and thus yield the youngest dates, while apatites with higher [eU] (more radiation damaged and higher T_{ec}) are incompletely reset under the same conditions and so yield older dates. Increasing T_{pk} to 70 °C induces greater resetting of apatites with higher [eU]. A T_{pk} of 80 °C causes nearly complete He loss in all apatites, thereby generating a fairly uniform population of dates. Thus, using the radiation damage trapping model, burial and unroofing simulations that induce no apatite resetting show no correlation between date and [eU] and those that induce complete apatite resetting yield dates that cluster fairly tightly. Only those simulations that include an episode of partial He loss can generate broad distributions of AHe dates that correlate with [eU] and [He], due to divergence of He retentivities in the apatite suite prior to partial resetting.

APPLICATION TO (U-Th)/He DATA FROM THE GRAND CANYON REGION

We now apply this model to slightly more complex thermal histories to determine whether we can reasonably reproduce our detrital AHe dates. Given the geographic proximity of the four samples (Fig. 1), it is reasonable to suspect that they had similar thermal histories. We assume monotonic heating, which is justified by the more or less steady accumulation of overlying Mesozoic and Cenozoic strata (e.g., Hunt, 1956; Hintze, 1988), from the time of deposition until the attainment of peak temperature at 80 Ma, coincident with the end of Sierran arc magmatism and the onset of Laramide tec-
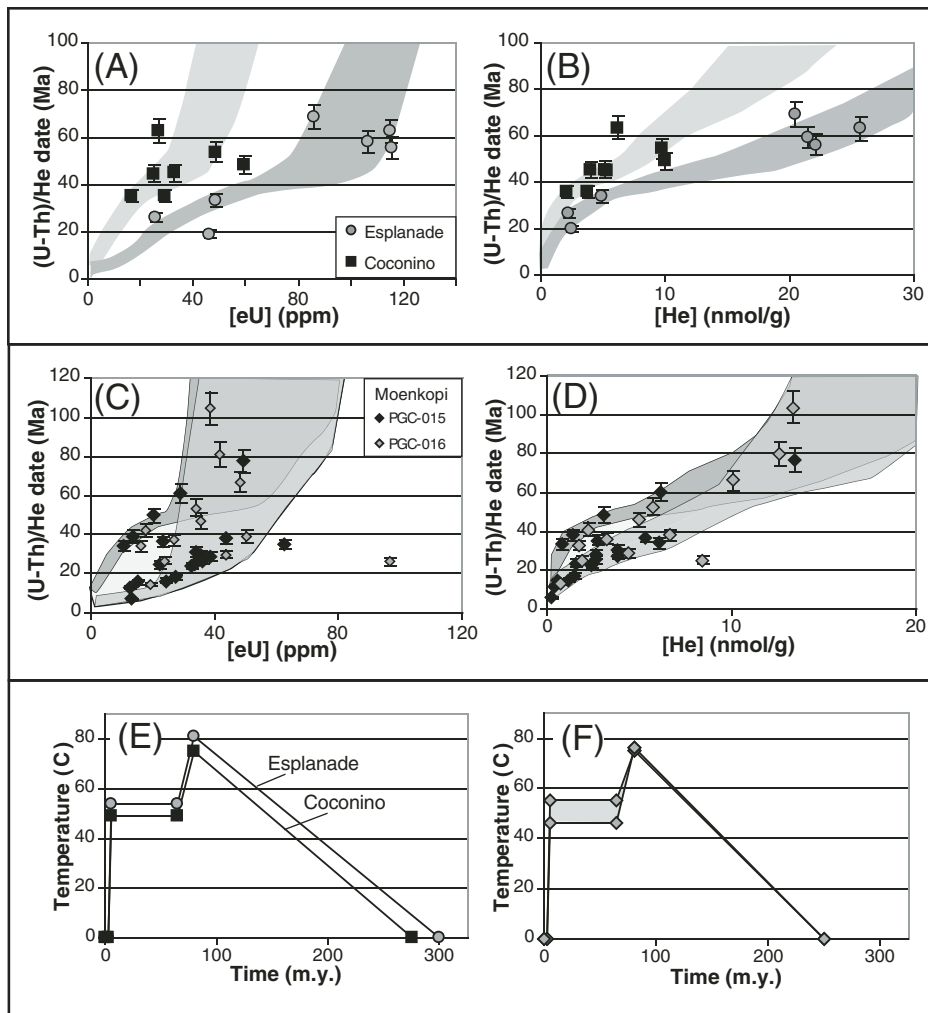
Figure 2. Individual (U-Th)/He apatite (AHe) dates (symbols) as function of effective U concentration [eU] and He concentration [He] for Esplanade and Coconino samples (A and B) and Moenkopi samples (C and D). Analytical errors are plotted as 8% (2σ). E: Simulated thermal histories used to reproduce Esplanade and Coconino data distributions using radiation damage trapping model, with simulated distributions of dates depicted as shaded fields in A and B. These simulations use AHe$_{dep}$ (AHe date of apatite when deposited) of 5–100 Ma. F: Simulated thermal histories used to reproduce Moenkopi data distributions using radiation damage trapping model, using AHe$_{dep}$ of 5–200 Ma. Single thermal history does not generate entire data spread of samples, so we depict end-member thermal histories that together encompass fan of data. Two end-member simulated distributions are depicted as separate, but overlapping, shaded fields in C and D.
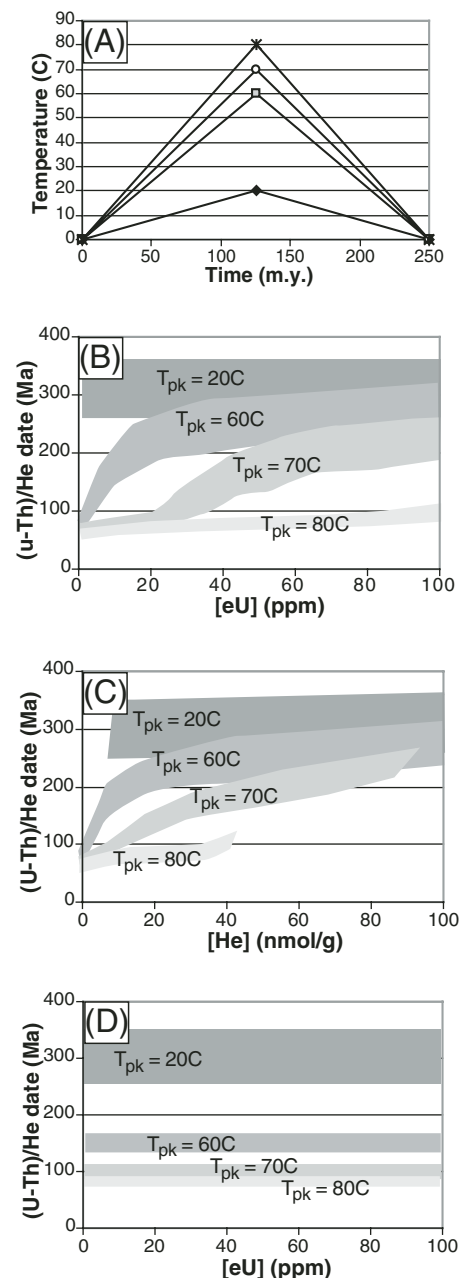

Figure 3. Predictions of radiation damage trapping model for distributions of dates resulting from simple burial and unroofing histories. A: Simulated temperature (T)-time paths. B, C: Simulated (U-Th)/He apatite (AHe) distributions of dates as function of effective U concentration [eU] and He concentration [He] for thermal histories in A. These simulations use AHe$_{dep}$ (AHe date of apatite when deposited) from 5 to 100 Ma, where lower bound in each fan of data represents simulated results for AHe$_{dep}$ of 5 Ma, and upper bound for AHe$_{dep}$ of 100 Ma. D: Simulated AHe distributions of dates as function of [eU] predicted by conventional Durango He diffusion kinetics and not including effect of radiation damage on He retentivity for thermal histories in A.

tonism (Burchfiel et al., 1992). Unroofing to the surface occurs by 0 Ma. We varied the thermal history from 80 Ma to 0 Ma to approximate the history consistent with the observed spectrum of dates. We use depositional ages of 300 Ma for the Esplanade, 275 Ma for the Coconino, and 250 Ma for the Moenkopi (e.g., Hintze, 1988), and [eU] values that encompass the range for apatites analyzed in each sample.

A two-pulse exhumation model reproduces the distributions of AHe dates in our four samples (Fig. 2). The first pulse coincides with Laramide time. A second mid-late Miocene pulse is necessary to explain the youngest dates. This thermal history is consistent with apatite fission-track

results in the region (Dumitru et al., 1994; Kelley et al., 2001). The Esplanade sample was collected ~200 m deeper in the same stratigraphic section as the Coconino sample, implying higher peak temperatures during burial. Using AHe$_{dep}$ of 5–100 Ma, and applying the same unroofing model but maintaining a temperature difference of 6 °C, predicts apatite suites consistent with the data from both the Esplanade and Coconino samples (Figs. 2A, 2B, 2E). The Moenkopi samples are characterized by a broader fan of dates (Figs. 2C, 2D). We use AHe$_{dep}$ of 5–200 Ma to better reproduce this wider distribution, and apply the same two-pulse unroofing history used for the Esplanade and Coconino samples but different temper-

ature conditions. Individual model simulations reproduce much, but not all, of the data spread. Therefore, we display a range of temperature-time paths that predict apatite suites that together encompass the Moenkopi data (Fig. 2F); the temperature differences between these histories are small. Cathodoluminescence imaging revealed internal zoning in some Moenkopi apatites that was not observed in the other samples (Fig. DR2; see footnote 1). This may account for some additional scatter in the Moenkopi data, because apatite dates are corrected for ejection of [4]He from the crystal assuming a homogeneous distribution of U and Th, and zonation can contribute to errors in this correction (Farley et al., 1996).

These simulations highlight the sensitivity of the correlations between date and [eU] and [He] to small variations in the thermal history. Only a restricted set of temperature-time paths can reproduce the distributions of dates in our samples, although the particular paths depicted in Figure 2 are not unique. Two key results of these simulations are that all four samples require a major mid-late Miocene pulse of cooling to generate the youngest apatite dates, and that extended residence in the apatite He partial retention zone after attainment of peak temperature is necessary to generate the large spread of dates. The tectonic implications of these results will be discussed in a subsequent publication.

IMPLICATIONS FOR (U-Th)/He THERMOCHRONOMETRY

Our results suggest that the increase in apatite He retentivity due to radiation damage, implied by laboratory diffusion data, is important for the interpretation of data in certain geological settings. However, we emphasize that for moderate to rapid monotonic cooling rates common for bedrock exhumation, the radiation damage trapping model does not predict a broad span of dates even for a large range in [eU] (Shuster et al., 2006). This is because monotonic cooling generally provides insufficient time for the apatite He retentivity characteristics of individual grains to diverge during cooling, and is consistent with the uniform AHe dates of many rocks inferred to be monotonically cooled. Additional complicating factors may contribute to dispersion of AHe dates not positively correlated with [eU] and unrelated to radiation damage control on He diffusion kinetics. These include micro-inclusions with high [eU], fluid inclusions, He implantation from external phases, and zonation (e.g., Fitzgerald et al., 2006).

Forward models predict that the effect of radiation damage on He retentivity will be manifested in suites of apatites with a range of [eU] that had a history in which the apatite He diffusion kinetics had sufficient time to diverge prior to an episode of partial resetting. A common geological history that satisfies these requirements is one like that in the Grand Canyon region, involving (1) deposition of compositionally diverse apatites with variable provenance dates in sedimentary units, and (2) burial, partial He loss, and subsequent exhumation. Models incorporating the effect of radiation damage can explain both the spans of AHe dates and their positive correlations with [eU] and [He] in the four samples investigated from this region. A similar data pattern attributed to enhanced He retention in higher U apatites has been reported in AHe data from the Otway Basin, where high [eU] apatites in a granitic cobble are significantly older than lower [eU] apatites within the host volcanogenic sediment (Green et al., 2006).

Thus, our data suggest that in some situations, a span of AHe dates positively correlated with [eU] is geologically meaningful. Analysis of individual detrital apatites is essential for successful data interpretation, because the averaging effect introduced by analysis of multigrain fractions would obscure the relationships observed in this study. Our simulations predict that the correlations between AHe date, [He], and [eU] can be very sensitive to the thermal history. Thus, it may be possible to extract additional information regarding the details of the temperature-time path from these relationships that would not be possible in a sample characterized by a uniform distribution of apatite dates.

REFERENCES CITED
Burchfiel, B.C., Cowan, D.S., and Davis, G.A., 1992, Tectonic overview of the Cordilleran orogen in the western United States, *in* Burchfiel, B.C., et al., eds., The Cordilleran orogen: Coterminous U.S.: Boulder, Colorado, Geological Society of America, Geology of North America, v. G-3, p. 407–479.

Crowley, P.D., Reiners, P.W., Reuter, J.M., and Kaye, G.D., 2002, Laramide exhumation of the Bighorn Mountains, Wyoming: An apatite (U-Th)/He thermochronology study: Geology, v. 30, p. 27–30, doi: 10.1130/0091–7613 (2002)030<0027:LEOTBM>2.0.CO;2.

Dumitru, T.A., Duddy, I.R., and Green, P.F., 1994, Mesozoic–Cenozoic burial, uplift and erosion history of the west-central Colorado Plateau: Geology, v. 22, p. 499–502, doi: 10.1130/0091–7613(1994)022<0499:MCBUAE>2.3.CO;2.

Farley, K.A., 2000, Helium diffusion from apatite: General behavior as illustrated by Durango fluorapatite: Journal of Geophysical Research, v. 105, p. 2903–2914, doi: 10.1029/1999JB900348.

Farley, K.A., Wolf, R.A., and Silver, L.T., 1996, The effects of long alpha-stopping distances on (U-Th)/He ages: Geochimica et Cosmochimica Acta, v. 60, p. 4223–4229.

Fitzgerald, P.G., Baldwin, S.L., Webb, L.E., and O'Sullivan, P.B., 2006, Interpretation of (U-Th)/He single grain ages from slowly cooled crustal terranes: A case study from the Transantarctic Mountains of southern Victoria Land: Chemical Geology, v. 225, p. 91–120, doi: 10.1016/j.chemgeo.2005.09.001.

Green, P.F., and Duddy, I.R., 2006, Interpretation of apatite (U-Th)/He ages and fission track ages from cratons: Earth and Planetary Science Letters, v. 244, p. 541–547, doi: 10.1016/j.epsl.2006.02.024.

Green, P.F., Crowhurst, P.V., Duddy, I.R., Japsen, P., and Holford, S.P., 2006, Conflicting (U-Th)/He and fission track ages in apatite: Enhanced He retention, not anomalous annealing behavior: Earth and Planetary Science Letters, v. 250, p. 407–427.

Hintze, L.F., 1988, Geologic history of Utah: Brigham Young University Geology Studies Special Publication 7, 202 p.

Hunt, C.B., 1956, Cenozoic geology of the Colorado Plateau: U.S. Geological Survey Professional Paper 279, 99 p.

Kelley, S.A., Chapin, C.E., and Karlstrom, K.E., 2001, Laramide cooling histories of Grand Canyon, Arizona, and the Front Range, Colorado, determined from apatite fission-track thermochronology, *in* Young, R.A., and Spamer, E.E., eds., Colorado River origin and evolution: Grand Canyon National Park, Grand Canyon, Arizona, Grand Canyon Association, p. 37–42.

Lorencak, M., Kohn, B.P., Osadetz, K.G., and Gleadow, A.J.W., 2004, Combined apatite fission track and (U-Th)/He thermochronometry in a slowly cooled terrane: Results from the 3440 m deep drill hole in the Canadian Shield: Earth and Planetary Science Letters, v. 227, p. 87–104.

Naeser, C.W., Duddy, I.R., Elston, D.P., Dumitru, T.A., and Green, P.F., 1989, Fission-track dating: Ages for Cambrian strata and Laramide and post-middle Eocene cooling events from the Grand Canyon, Arizona, *in* Elston, D.P., et al., eds., Geology of the Grand Canyon, northern Arizona: Washington, D.C., American Geophysical Union, p. 139–144.

Naeser, C.W., Duddy, I.R., Elston, D.P., Dumitru, T.A., and Green, P.F., 2001, Fission-track analysis of apatite and zircon from Grand Canyon, Arizona, *in* Young, R.A., and Spamer, E.E., eds., Colorado River origin and evolution: Grand Canyon National Park, Grand Canyon, Arizona, Grand Canyon Association, p. 31–36.

Shuster, D.L., Flowers, R.M., and Farley, K.A., 2006, The influence of natural radiation damage on helium diffusion kinetics in apatite: Earth and Planetary Science Letters, v. 249, p. 148–161.

Warnock, A.C., Zeitler, P.K., Wolf, R.A., and Bergman, S.C., 1997, An evaluation of low-temperature apatite U-Th/He thermochronometry: Geochimica et Cosmochimica Acta, v. 61, p. 5371–5377.

Wolf, R.A., Farley, K.A., and Kass, D.M., 1998, Modeling of the temperature sensitivity of the apatite (U-Th)/He thermochronometer: Chemical Geology, v. 148, p. 105–114.