Quantifying the effects of radiation damage on helium diffusion in damaged zircon

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Zircon (U-Th)/He is an increasingly popular method of interpreting low-temperature thermal histories, based on the production and diffusive loss of radiogenic ⁴He from individual zircon grains. Both the α -decay of U and Th, and the spontaneous fission of ²³⁸U, cause atomic dislocations in the zircon crystal lattice. This damage accumulates over geologic timescales and can undermine standard assumptions regarding helium diffusion in zircon, especially at doses exceeding $\sim 2 \times 10^{18} \alpha$ -decays/g. A quantitative understanding of the relationship between radiation dose and helium diffusion kinetics is not only critical for accurate modeling of thermal histories, but could recover information from a range of temperatures and/or multiple thermal events. The newly published model by Guenthner et al [1] provides an excellent framework for these relationships; however, it has only been tested in a limited number of natural settings. In order to develop a model to account for these relationships, we analyze zircons from slowly-cooled crystalline rocks from the Hall Peninsula of Baffin Island, Canada and assorted crystalline rocks from the Sinai Peninsula, Egypt. Samples in this study are analyzed by: Raman spectroscopy, to estimate radiation LA-ICP-MS depth profiling, to dose: account for heterogeneous U and Th distribution; and step-heated, fractional-loss diffusion experiments to measure diffusion kinetics, followed by standard (U-Th)/He analysis. We observe a clear negative log-linear relationship between apparent zircon (U-Th)/He age and effective uranium concentration (eU = [U])+ 0.23[Th], as a proxy for radiation dose), with dose estimates one order of magnitude lower than previously observed for significant diffusivity increases. Preliminary interpretation of step-heating experiments have yielded closure temperatures in the range of 140 - 186 °C (dT/dt = 10 °C/m.y.). Furthermore, we note significant changes in diffusion kinetics occurring at 540 - 560 °C for highly damaged zircon, including decreases in D_0/a^2 over two orders of magnitude, resulting in significant changes in nominal closure temperature. These data will be instrumental in the development of an improved model capable of extracting complex thermal histories from metamict zircon.

[1] Guenthner et al (2013) American Journal of Science.