Percolation of radiation damage and its implications for diffusivity in zircon

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Computational analysis of the geometry of elongate radiation damage zones from alpha recoil and fission provides new estimates of the doses required to reach percolation and full connectivity in zircon. Alpha recoil track damage percolates at doses from 2.5-3.1 x $10^{16} \alpha/g$, about two orders of magnitude lower than previous estimates, with the difference partially due to elongation, which previous modeling neglected, and partially due to decay chains creating pre-made networks of connected tracks. This dose level is far below that required for metamictization, and suggests that alpha recoil track percolation has no effect on macroscopic or unit cell properties, at least as measured to date. However, fission tracks percolate at a dose of approximately 1.9 x $10^{18} \alpha/g$, similar to the level formerly ascribed to alpha recoil damage percolation and correlating with various transitions in material properties, such as an inflection in the relationship between dose and macroscopic swelling. Consideration of the undamaged regions between damage zones indicates that c-axis-parallel channels are frequently interrupted by alpha recoil damage, at the μ m scale at very low doses and 10's of nm at usual doses in natural zircon, with the probable effect of decreasing diffusivity anisotropy. The percolation and further interconnectivity of alpha recoil damage corresponds with a general minimum in diffusivity and maximum in (U-Th)/He closure temperature in zircon, indicating that alpha recoil damage percolation does not make a grain "leaky." Instead, the onset of poor He retentivity at high damage levels correlates with fission-track percolation. These results, combined with the observation that measurements thus far indicate little diffusivity increase over more than an order of magnitude of damage accumulation, are non-intuitive with respect to the trapping mechanism for He diffusivity reduction. Increasing tortuosity of diffusional pathways is an alternative the mechanism for explaining the decrease in diffusivity at low doses. Increasing diffusivity at high damage levels may be partly explained as a change in the length scale of diffusion on the subgrain scale, due to the decrease in the mean distance from any point within the grain to the percolating fission-track damage network.