American Mineralogist, Volume 99, pages 2095–2104, 2014

ACTINIDES IN GEOLOGY, ENERGY, AND THE ENVIRONMENT

Quantification of α-particle radiation damage in zircon†

KATIE M. SMYE[1,*], CLIVE BRIGDEN[1], ERIC R. VANCE[2] AND IAN FARNAN[1]

[1]Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge CB2 3EQ, U.K.
[2]Institute of Materials Engineering, Australian Nuclear Science and Technology Organisation, Lucas Heights, Menai, New South Wales 2234, Australia

ABSTRACT

Analysis of radiation damage in natural mineral analogs such as zircon is important for the evaluation of the long-term behavior of nuclear waste forms and for geochronology. Here we present results of experiments to determine the partitioning of radiation damage due to the heavy nuclear recoil of uranium and thorium daughters and the α-particles ejected in an α-decay event in zircon. Synthetic polycrystalline zircon ceramics were doped with ^{10}B and irradiated in a slow neutron flux for 1, 10, and 28 days to achieve the reaction $^{10}B + n \rightarrow {}^{7}Li + \alpha$ (+2.79 MeV), creating an α event without a heavy nuclear recoil. The ^{7}Li atoms produced in the nuclear reaction were directly detected by NMR "spin-counting", providing a precise measurement of the α-dose applied to each sample. The amount of damage (number fraction and volume fraction) created by each α-event (one α-event being a ^{7}Li + α-particle) has been quantified using radiological nuclear magnetic resonance and X-ray diffraction data. The number of permanently displaced atoms in the amorphous fraction was determined by ^{29}Si NMR to be 252 ± 24 atoms for the $^{10}B(n,\alpha)$ event when the heavy recoil is absent, which is broadly in agreement with ballistic Monte Carlo calculations. The unit-cell swelling of the crystalline fraction, determined by X-ray diffraction, is small and anisotropic. The anisotropy is similar to that observed in ancient natural samples and implies an initial anisotropic swelling mechanism rather than an anisotropic recovery mechanism occurring over geological timescales. The small unit-cell volume swelling is only ~6% of the expansion frequently attributed to α-particles associated with an actinide α-decay event. The lattice parameters indicate a volume increase as a function of α dose of 0.21 $A^3/10^{18}$ α-events/g, which is significantly less than the increase of 3.55 $A^3/10^{18}$ α-events/g seen in Pu-doped zircon and 2.18 $A^3/10^{18}$ α-events/g seen in natural zircon. It is concluded that the heavy recoil plays a more important role in unit-cell swelling than previously predicted. The likely mechanism for such an effect is the rapid, and thus defect-rich, recrystallization of material initially displaced by the heavy recoil.

Keywords: Zircon, NMR spectroscopy, radiation damage, α-particle