Radiation damage in zircon

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ABSTRACT

A single, zoned, Sri Lankan zircon exhibits a range of microstructures from crystalline to nearly amorphous that are the result of radiation damage over a dose range of $2.1 \times 10^{15}$ to $1.0 \times 10^{17}$ α-decay events/mg (0.16–0.47 dpa). The zones in the crystal were examined at a variety of length scales using optical microscopy, micro-Raman spectroscopy, electron microprobe analysis, and transmission electron microscopy. Birefringence varies linearly with dose: 

$$birefringence = -4.71 \times 10^{-18} / g \cdot D_a + 4.86 \times 10^{-2}.$$ 

Full width at half maximum (FWHM) measurements of the $B_{1g}(v_1)$ peak, as determined by micro-Raman spectroscopy, were used to estimate the extent of radiation damage in each zone. The radiation dose (calculated on the basis of U and Th concentrations and sample age) vs. damage (from the FWHM measurements of the $B_{1g}(v_1)$ peak) relationship among zones was consistent with results for a suite of Sri Lankan single crystals, suggesting that all of these zircon crystals have undergone a similar thermal history. Based on a comparison with zircon crystals that are considered to have undergone minimal annealing (e.g., lunar zircon), the Sri Lankan zircon crystals have accumulated less damage than is expected based on their calculated dose, consistent with previous evidence for annealing. To estimate the extent of annealing in Sri Lankan samples, an equivalent damage dose (i.e., the dose required to produce the same amount of damage in an unannealed sample) was calculated for a given U and Th concentration by determining the time of damage accumulation required to create an equivalent amount of damage in an unannealed zircon. The dose vs. damage relationship in Sri Lankan zircon crystals (560 Ma) is equivalent to that of a ~375 Ma unannealed zircon, suggesting that approximately one-third of the damage has been lost due to annealing. The dose vs. damage relationship is consistent with the direct impact model of damage accumulation. Transmission electron microscopy revealed zoning on a scale finer than could be identified optically (<0.5 μm) and the presence of abundant ZrO2 nano-particles at zone boundaries.

INTRODUCTION

Natural zircon (ZrSiO4) (I4/amd, Z = 4) commonly contains up to 5000 ppm UO2 + ThO2, but has been found with total U and Th concentrations up to 7 wt% (Speer, 1982; Deer et al. 1997). Mineralogical analyses of the “Chernobyl Lavas” from the destroyed nuclear power plant in the former Soviet Union have shown that crystalline zircon (6.1–12.9 wt% U) formed as a result of melted nuclear fuel (U), fuel rod material (Zr) and sand (Si) (Anderson et al. 1993). The capacity of zircon to incorporate U has lead to the proposal that zircon be considered as a potential waste form for actinides, particularly for Pu from dismantled nuclear weapons (Ewing et al. 1995; Ewing, 1999, 2001). Zircon containing up to 10 wt% Pu has been synthesized (Weber 1990; Burakov et al. 2002). In applications to geologic dating and the use of zircon as a nuclear waste form, radiation damage may affect isotopic ratios or decrease chemical durability due to the radiation-induced crystalline-to-aperiodic transition.

In natural zircon, the dose (alpha-events/mg) can be calculated on the basis of the U and Th content and the age of the sample. The crystalline-to-aperiodic transition results from cascade damage due to α particles and recoil nuclei originating from radionuclides within the decay series of U and Th (Weber 1990).