Simulating 100 million years of radiation damage in six years

John M. Hanchar
Department of Earth Sciences, Memorial University of Newfoundland, St. John's, NL, A1B 3X5.

Zircon ($ZrSiO_4$) is an important mineral in understanding the Earth's crustal evolution. The stability of zircon in nature over long periods of time has led researchers to focus on zircon as the preferred mineral for geochronology. In order to provide an explanation for zircon U-Pb analyses that are isotopically disturbed, it is desirable to acquire a better understanding of radiation damage mechanisms and processes that have led to alteration of the zircon structure and the enhanced mobility of the parent or daughter isotopes. Zircon has also been proposed as a potential storage material for actinides, including weapons-grade plutonium from dismantled nuclear weapons.

For investigations of self-irradiation damage effects taking place in zircon, one option is to use single crystals of zircon doped with ^{238}Pu. Due to the high alpha-emitting activity of ^{238}Pu ($t_{1/2}$ = 87.74 years, specific activity = 17.3 Curies/gram), accelerated alpha-induced radiation damage in zircon crystals on the laboratory time scale (months to years) can be investigated. This is opposed to thousands to tens of thousands of years for ^{239}Pu-doped ceramics ($t_{1/2}$ = 24 100 years, specific activity = 0.063 Curies/gram), or millions to hundreds of millions of years for natural zircon samples (depending on the initial U content and the geologic history of the sample).

In our investigation, single crystals of synthetic zircon doped with ^{238}Pu, up to 3.5 mm in size, were grown for the first time ever using a Li-Mo flux. The crystals were transparent, of pink-brown color, and free of inclusions of separated Pu-oxide phases. Approximately five months after zircon synthesis, the crystals changed color to gray-brown, and after 14 months, the gray color in the crystals increased. After 24 months, the crystals were still transparent and free of inclusions of separated Pu phases. The development of cracks in the crystals increased since crystallization, due to the cumulative dose of self-irradiation from the decay of ^{238}Pu to ^{234}U. Cathodoluminescence spectroscopy and single crystal and powder X-ray diffraction measurements also reveal a loss of long-range order in the zircon crystals as a function of dose of the ^{238}Pu alpha-induced radiation damage. Additionally, our results suggest that the damage incurred by the zircon crystals at different times, and thus resulting doses, is greater than what is predicted by currently accepted models for the behavior of radiation damage in natural zircon.

Atlantic Geology, 2006, Volume 42, Number 2-3