

## DIFFERENTIAL HELIUM RETENTION IN ZIRCONS: IMPLICATIONS FOR NUCLEAR WASTE CONTAINMENT

Robert V. Gentry,<sup>1\*</sup> Gary L. Gush,<sup>2</sup> and Eddy R. McBay<sup>2</sup>

<sup>1</sup>Physics Department, Columbia Union College, Takoma Park, ND 20012

<sup>2</sup>Analytical Chemistry Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830

**Abstract.** A very sensitive helium leak detector was utilized to measure the helium liberated from groups of zircons extracted from six deep granite cores. The observed low differential loss of gaseous helium down to 2900 m (197°C) in these ancient Precambrian rocks is easily attributable to the greater diffusion of He at higher temperatures rather than losses due to corrosion of the zircons. This fact strongly suggests that deep granite burial should be a very safe corrosion-resistant containment procedure for long-term waste encapsulation.

Recent mass spectrometric studies (Gentry, et al. 1982) have revealed that lead has been retained in zircons extracted from deep (960 m to 4310 m) granite cores where the ambient temperature increases from 105°C to 313°C at the greatest depth. As a follow-up to those experiments we now report the results of differential helium retention in similar zircons extracted from the same granite core samples which were used in the lead analyses (Laney and Laughlin, 1981).

The procedure for separating the zircons from the six different granite cores (from depths of 960, 2170, 2900, 3502, 3930, and 4310 m) was the same as that used in the previous experiments. The high-density fractions, obtained by passing the crushed core samples through different methylene iodide separating funnels, were thoroughly washed with acetone before being placed on a standard microscope slide. A fine-tipped needle was used to pick out the individual zircons with the aid of a polarizing microscope. Groups of these separated zircons, usually about 10 in number, were then loaded onto the platinum filament of the thermal inlet probe of the mass spectrometer for differential helium analysis.

The helium measurements were performed on a Leybold-Heraeus model F helium leak detector that had a Chemical Data Systems Pyrolysis unit interfaced to the test port. The leak detector has a detection limit of less than  $10^{-10}$  cm<sup>3</sup>/sec when operating in the dynamic mode. (The instrument could have been operated in a near-static mode with increased sensitivity down to  $\sim 10^{-11}$  cm<sup>3</sup>/sec of He, but our experiments did not necessitate this increased sensitivity.)

\*Also: Research Assistant Professor, Physics Department, University of Tennessee, Knoxville, TN 37916.

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In our initial series of measurements our spectrometer was calibrated against a  $5 (\pm 0.5) \times 10^{-8}$  cm<sup>3</sup>/sec standard He leak. A subsequent recalibration with a more precise  $5 (\pm 0.5) \times 10^{-10}$  cm<sup>3</sup>/sec standard He leak revealed the total helium liberated during these initial measurements was slightly underestimated. The general procedure was to measure helium evolution from a group of zircons at progressively higher temperatures of 400°C, 600°C, and 1000°C for 20 sec intervals. (Previous studies of helium diffusion (Magomedov, 1970) from zircons indicated 1000°C was sufficient to liberate the helium with an activation energy of 15 kcal/mol.) We did not include the small amount of He observed at 1100°C in the total He summation because of possible atmospheric contamination. Between six and eight groups of zircons were analyzed at each depth. Runs were repeated at a given temperature until background helium levels were observed. Data recordings and integration under the peaks were done with a Nicolet 1170 signal averager.

The third column in Table 1 shows, as a function of depth, the total amount of He liberated per  $\mu\text{g}$  of zircon for zircon groups comprised of approximately equal-size ( $\sim 50\text{--}75 \mu\text{m}$ ) zircons. The fourth column in Table 1 shows the ratio of the amount of He actually measured in zircons from any particular depth to the estimated amount of He which should have accumulated in those same zircons assuming negligible diffusion loss. For the zircons taken from a surface outcrop we assumed this ratio was one because the specimens we used were small fragments from the interior of larger zircon crystals.

For the other zircons from the granite and gneiss cores, we made the assumption that the radiogenic Pb concentration in zircons from all depths was, on the average, the same as that measured (Zartman, 1979) at 2900 m, i.e.,  $\sim 8$  ppm with  $^{206}\text{Pb}/^{207}\text{Pb}$  and  $^{206}\text{Pb}/^{208}\text{Pb}$  ratios of ten (Gentry, et al., 1982; Zartman, 1979). Since every U and Th derived atom of  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$ , and  $^{208}\text{Pb}$  represents 8, 7, and 6  $\alpha$ -decays respectively, this means there should be  $\sim 7.7$  atoms of He generated for every Pb atom in these zircons.

Knowledge of the zircon mass and the appropriate compensation factor (to account for differences in initial He loss via near-surface  $\alpha$ -emission) enabled us to calculate the theoretical amount of He which could have accumulated assuming negligible diffusion loss. This compensating factor is necessary because the larger ( $150\text{--}250 \mu\text{m}$ ) zircons lost a smaller proportion of the total He generated within the crystal via near-surface  $\alpha$ -emission than did the smaller ( $40\text{--}50 \mu\text{m}$ ) zircons. For the smaller zircons we estimate as many as 30-

