

## Reinvestigation of the $\alpha$ -activity of Conway granite

ADAMS *et al.*<sup>1,2</sup> reported evidence for an unidentified 4.4 MeV  $\alpha$ -activity in certain core sections taken from Conway granite in New Hampshire. A similar  $\alpha$ -activity has also been reported by Cherdyn'tsev *et al.*<sup>3</sup> and by Brukl *et al.*<sup>4</sup> in different materials, but in neither case was it ever confirmed. We report here our reinvestigation of this phenomenon, and that we were unable to confirm the evidence of a 4.4 MeV  $\alpha$ -activity in the Conway granite.

Because it was thought that a previous failure<sup>5</sup> to confirm the existence of this activity in the Conway granite might have been due to subtle differences in sample preparation, considerable effort was made to obtain the same core material and to follow the same preparation techniques used by Adams *et al.*<sup>1,2</sup>. On the other hand, because no record was kept of the depth of the particular core section which yielded evidence for the 4.4 MeV  $\alpha$ -emitter, it is not known whether the Conway granite core sections used in these experiments were from the same depth as the core section in question. Other pieces of granite from this same general area were also obtained from T. P. Kohman, and were given the same experimental treatment as the original cores obtained by Adams.

The experimental treatment, with minor variations from time to time, consisted of first crushing the 30.5 cm long, 2.5 cm diameter cores to about 1 mm size, followed by magnetic separation of the biotite in a standard Franz isodynamic magnetic separator. The biotite flakes obtained from this magnetic separation were then crushed using a mortar and pestle. To extract the small radioactive inclusions from this crushed biotite, a heavy liquid separation was carried out using methylene iodide ( $\rho=3.4 \text{ g cm}^{-3}$ ). The crushed biotite was left in the methylene iodide for several hours to allow ample time for the small (10–200  $\mu\text{m}$ ) radioactive mineral inclusions ( $\rho>3.5 \text{ g cm}^{-3}$ ) to separate from the less dense biotite ( $\rho\approx 3 \text{ g cm}^{-3}$ ). The methylene iodide residue was subsequently removed from the inclusions using an acetone rinse.

After drying, the inclusions were sprinkled on to the surface of 1 x 3 inch Eastman Kodak NTA emulsion track plates. These NTA plates had an emulsion thickness of 25  $\mu\text{m}$  and were sensitive to  $\alpha$  particles without recording  $\alpha$  rays. The plates with inclusions were then placed in a light tight box under refrigeration for periods ranging from three weeks to three months. Subsequent development and microscopic scanning of these plates revealed the  $\alpha$ -activities of the various individual inclusions on the plate. At this point microscopic techniques were utilized to pick out only the inclusions which exhibited the greatest cluster of  $\alpha$  tracks. These highly radioactive inclusions, which varied in size from about 10 to 200  $\mu\text{m}$ , were then placed either singly or in groups of up to 25 on to Pt or stainless steel disks. The initial experiments involved dissolution of these inclusions on the Pt disks with drops of concentrated HF and HNO<sub>3</sub>, the Pt disk itself serving as the source for the  $\alpha$ -spectrometer. It was soon found that better energy resolution could be obtained by subsequent crushing of the dissolved residue, and this technique was followed until it was found that the acid dissolution could be dispensed with entirely. From then on, the higher activity inclusions were mounted on stainless steel disks, crushed, and then counted for periods ranging from about one day to a week.

The measurements were made on an  $\alpha$ -spectrometer with a 2 cm diameter, gold-covered surface barrier Si-detector of 300- $\mu\text{m}$  depth mounted in a vacuum chamber. Sample disks were placed in the bottom of a polyethylene cap which fitted over the detector so that the sample was approximately 1 mm from the face of the Si-diode. A preamp was mounted directly on the base of the detector (bias  $\sim 100 \text{ V}$ ) and its output was fed to an amplifier. The pulse-height spectrum was measured with a multichannel analyzer. Although the system exhibited a resolution of 30–35 keV (FWHM) for a very thin source, samples prepared by the above technique exhibited a typical resolution of 50–60 keV. Measurements generally spanned the

range of  $\sim 2$  to 10 MeV over 2,048 channels. Energy drift and background were negligible.

About 50 different  $\alpha$ -spectra were obtained on the various cores and samples from the Conway granite. The  $\alpha$ -spectra from the multichannel analyzer were recorded on punched tape, which then, by means of a computer program, generated an accurate plot of counts per channel against channel number. Energy calibration was accomplished by means of a pulser and standard  $\alpha$ -sources.

The  $\alpha$ -spectra generally showed evidence of both the <sup>238</sup>U and <sup>232</sup>Th  $\alpha$ -decay chains, and in some cases the abundance ratios of the two elements and their respective daughters were noted to vary from inclusion to inclusion. This was not surprising, for the inclusion selection process was not specific for a particular mineral, hence different U/Th ratios were to be expected as dissimilar mineral inclusions were analyzed. In some cases disequilibrium in the U—Th chains was observed in the  $\alpha$ -spectra but no attempt was made to determine whether this condition arose from slight variations in the sample preparation procedure or from an inherent disequilibrium within the sample before crushing. The reason for this disequilibrium condition was not pursued because the main point of the experiments was to determine whether any 4.4 MeV  $\alpha$ -activity could be detected.

With the possible exception of one sample, which exhibited poor statistics because of a relatively weak source, the experiments did not reveal any evidence for the 4.4 MeV  $\alpha$ -emitter reported by Adams *et al.*<sup>1</sup>. A redetermination of that particular sample subsequently gave no evidence whatsoever for the 4.4 MeV activity. Thus we were unable to find any confirmatory data for the existence of a 4.4 MeV  $\alpha$ -activity in the Conway granite.

This research was supported by Union Carbide Corporation under contract with the Division of Basic Energy Sciences of the Department of Energy. Separation of the biotite from the crushed granite was carried out using the magnetic separator located in the Geology Department of the University of Tennessee, Knoxville. Mr. Mirza Beg and later Dr. Otto Kopp supervised this phase of the experiment.

R. V. GENTRY  
J. H. HALPERIN  
B. H. KETELLE  
G. D. O'KELLEY  
R. W. STOUGHTON

*Chemistry Division,  
Oak Ridge National Laboratory,  
Oak Ridge, Tennessee 37830*

J. A. S. ADAMS

*Geology Department,  
Rice University,  
Houston, Texas 77001*

Received 11 January; accepted 9 March 1978.

1. Adams, J. A. S. & Rogers, J. J. W. TID—20506(1964).
2. Cherry, R. D. Richardson, K. A. & Adams, J. A. S. *Nature* **202**, 639 (1964).
3. Cherdyn'tsev, V. V., Zvenev, V. L., Kuptsov, V. M. & Kisilitsina, G. I. *Geokhimiya* **4**, 395 (1968).
4. Brukl, A., Hennegger, F. & Hilbert, H. *Oesterr. Akad. Wis. Abt. 2A* **160**, 129 (1951).
5. Petrzhak, K. A., Yakunin, M. I. & Ter-Akop'yan, G. M. *Atomn. Energ.* **32**, 179 (1972).