

TABLE I. Measured isotopic ratios of argon.

| Sample | A ⁴⁰ /A ³⁶ | A ³⁸ /A ³⁶ |
|--|----------------------------------|----------------------------------|
| Tracer | 1.172 ± 0.001 | 0.02242 ± 0.00002 |
| 4.90 cm ³ /cc. tracer +197.8 g sylvite | 2.633 ± 0.002 | 0.02239 ± 0.00002 |

indicate that the composition of the sample was constant within the errors of the experiment.

The argon content was determined by placing 197.8 g of sylvite in a quartz furnace and evacuating at room temperature for 12 hours. At this point 4.90 cc of tracer argon with an A⁴⁰/A³⁶ ratio of 1.172 was added to the volume and heating started. The temperature was held at 1050°C for one hour. At this temperature the KCl was refluxing vigorously. The gas over the molten sylvite was finally mixed by sorption and desorption from charcoal three times. The gases were then purified in a greaseless system. The purification train consisted of a CuO₂ furnace at 400°C, a KOH trap, an Mg(ClO₄)₂ dryer and finally a calcium furnace. The gas was treated in the system to constant volume. The excess A⁴⁰ in this sample was then determined by the increase at the A⁴⁰ position as measured on a mass spectrometer. The results are shown in Table I. Within the limits of error of the A³⁸/A³⁶ ratio shown in Table I there is no atmospheric argon in the sylvite. In order to check this more accurately the second aliquot of sylvite was run through the procedure without the addition of tracer material. The argon obtained was 99.0 ± 0.5 percent radiogenic. The chemical purity of the tracer argon was determined by dilution of a known volume of the tracer material with a known volume of Linde spectroscopic argon, and measurement of the mixture ratio on the mass spectrometer. Combining the factors the A⁴⁰ content of this particular sylvite sample is 0.360 ± 0.007 p.p.m. by weight.

To measure the Ca⁴⁰ content two samples containing 10.770 and 8.849 g, respectively, of sylvite were used. To each was added a weighed quantity of the separated isotope Ca⁴⁸, the purity of which was checked by dilution against normal CP calcium. The complete sample was then brought into solution to insure mixing. The first stage of chemical purification was fractional crystallization. After the KCl had been depleted by a factor of 10 the Ca was precipitated with oxalate^{8,9} and washed two times to reduce the potassium. It was then reprecipitated with oxalate and re-washed twice. The diluted Ca⁴⁸ was then compared with the tracer Ca⁴⁸ to determine the Ca⁴⁰ content and the normal calcium content of the sylvite. The results are shown in Table II. From the change in the Ca⁴⁴ peak the "normal" calcium impurity in the KCl was determined. The excess Ca⁴⁰ over this impurity was 2.82 ± 0.02 p.p.m. in the first case and 2.87 ± 0.03 p.p.m. in the second.

Combining the above individually determined radiogenic A⁴⁰ and Ca⁴⁰ contents gives the ratio of K-capture to beta as 0.1263 ± 0.0027. Due to systematic errors we do not believe this result to be of the precision accuracy given, but it is believed the result is good to ±4 percent. This error does not include errors due to possible loss of A⁴⁰ or Ca⁴⁰ from the sample during geologic times. It can, however, be considered as a lower limit since any alteration would affect the A⁴⁰ content more strongly than the Ca⁴⁰. The agreement of this value for the branching ratio with the most recent counting value of 0.136 determined by Sawyer and Wiedenbeck⁷ indicates that leakage in this sample was not serious. It

TABLE II. Measured isotopic ratios of calcium.

| Sample | 40/48 | 44/48 |
|--|---------------|-------------------|
| Ca ⁴⁸ spike | 0.585 ± 0.001 | 0.02042 ± 0.00005 |
| 91.5γ Ca ⁴⁸ +8.849 g sylvite Ca | 1.040 ± 0.002 | 0.02316 ± 0.00007 |
| 91.5γ Ca ⁴⁸ +10.770 g sylvite Ca | 1.157 ± 0.003 | 0.02396 ± 0.00007 |

should be noted that this branching ratio is sufficient to account for the A⁴⁰ in the atmosphere.

We wish to thank Professor W. W. Watson of Yale for supplying the A³⁶ tracer used in this experiment. The Ca⁴⁸ tracer was supplied from the Y-12 Plant of Oak Ridge on allocation from the Isotopes Division of the AEC.

- ¹ J. H. Reynolds, Phys. Rev. **79**, 789 (1950).
- ² E. Bleuler and M. Gabriel, Helv. Phys. Acta **20**, 67 (1947).
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- ⁵ H. E. Suess, Phys. Rev. **73**, 1209 (1948).
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- ⁷ G. A. Sawyer and M. L. Wiedenbeck, Phys. Rev. **79**, 490 (1950).
- ⁸ G. H. Ellis, Anal. Chem. **10**, 112 (1938).
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The Precision Determination of Some Half-Lives*

JAMES W. COBBLE AND R. W. ATTEBERRY

Chemistry Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee
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THE decay periods of several radioactive nuclides have been measured at this Laboratory using sources which were bombarded in the Oak Ridge National Laboratory pile and subsequently purified by ion-exchange column techniques using both cation and the new strong base anion exchangers. The activity was followed by a 4π-geometry ionization chamber filled to 40 atmos. with argon. This chamber was connected through a "vibrating reed" electrometer to a recording potentiometer which continuously records the *instantaneous* value of the activity as a function of time. The instrument had been previously calibrated to determine the effects of ion-recombination, scale drift, etc. Such a procedure is of great advantage with the shorter periods, and the arrangement is capable of a very high precision.

Results for four nuclides are given in Table I.

TABLE I. Half-lives of nuclides.

| Isotope | T _{1/2} (this research) | T _{1/2} (previously recorded values) |
|------------------|----------------------------------|--|
| Na ²⁴ | 15.10 ± 0.04 hr. ^a | 14.8 ± 0.1; ^b 15.04 ± 0.06 ^c |
| Cl ³⁸ | 37.29 ± 0.04 min. | 37.0 ± 0.3; ^b 38.5 ± 0.5 ^d |
| Br ⁸² | 35.87 ± 0.05 hr. | 33.9 ± 0.3; ^e 36.0 ± 0.1 ^f |
| Tc ⁹⁶ | 4.20 ± 0.04 days ^g | 4.2 ± 0.1; ^b 4.25 ± 0.05 ^h |

^a Errors estimated from reproducibility in duplicate determinations.

^b S. N. Van Voorhis, Phys. Rev. **49**, 889 (1936).

^c A. K. Solomon, Phys. Rev. **79**, 403 (1950).

^d N. Hole and K. Siegbahn, Arkiv Mat., Astron. Fys., **33A**, 9 (1946).

^e A. H. Snell, Phys. Rev. **52**, 1007 (1937).

^f E. Berne, Phys. Rev. **77**, 568 (1950).

^g Sample prepared from a Mo(d, n) reaction; the larger error in this isotope is due to uncertainties in the subtraction of the longer-lived Tc⁹⁵.

^h J. E. Edwards and M. L. Pool, Phys. Rev. **69**, 253 (1946).

ⁱ G. E. Boyd, private communication.

In the case of Na²⁴ and Br⁸², this work confirms the longer periods recently reported (references c and f of Table I).

* This document is based on work performed for the AEC at Oak Ridge National Laboratory.

The Virial Theorem and the Variation Principle

IRVIN ISENBERG

Institute for the Study of Metals, University of Chicago, Chicago, Illinois
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IT has been called to the author's attention that the theorem proven in a Letter to the Editor,¹ under the above title, was originally proven by V. Fock.² The author regrets not being familiar with this work prior to publication of the letter.

¹ I. Isenberg, Phys. Rev. **79**, 737 (1950).

² V. Fock, Zeits. f. Physik **63**, 855 (1930).