The Radioactive Decay of K^{40}*

L. B. Borst and J. J. Floyd
Brookhaven National Laboratory, Upton, New York
September 10, 1948

FOR some time we have been interested in the radioactive decay of K^{40} and its geochemical significance. This note reports preliminary observations made on the β-ray from a sample of potassium enriched 14-fold in K^{40}, obtained from the Stable Isotopes Division of the U. S. Atomic Energy Commission,[1] and, in addition, a test for argon extractable from Permian sylvite obtained from the Department of Mineralogy, Harvard University.

Current interest in this problem is high because of the difficulty in establishing the partial disintegration constant for electron capture λ_e, because of the uncertainty in the partial disintegration constant for β-emission λ_β, and because of the uncertainty in the energy changes involved in electron capture and the β-processes and their relation to γ-emission.

Gleditsch and Gráf[2] and Birch[3] have pointed out that too low a half-life for decay will be difficult to accept in view of the existence of rocks of ages up to 2×10^9 yr.,[2b,4] which could not crystallize if the K^{40} heat output exceeds some reughly known upper limit. In particular the low value recently reported by Bleuler and Gabriel[5] for the total half-life of K^{40}, $T_{\frac{1}{2}}=(2.4\pm0.5)\times10^8$ yr., has been the subject of discussion[2,3] and of detailed criticism by Ahrens and Evans.[6] These last authors report a revised half-life of $(4.5\pm0.5)\times10^8$ yr. based on a reinterpretation of older counting data and an interpretation of the trends in the calcium to potassium ratio of old and young lepidolite samples.

The results reported below lead to a still higher value for the half-life for decay of K^{40} of $4\text{–}12\times10^8$ yr.

1. *Specific beta-activity.* A sample of KCl weighing 18.9 mg, and containing 0.15 ± 0.02 percent K^{40}, was studied in thin layer (2.5 mg/cm^2) mounted on thin aluminum (7 mg/cm^2), using a mica window counter and aluminum absorbers[7] (Fig. 1). The data plotted are individual counting rates after subtracting the counter background. The broken line indicates the γ-ray contribution evaluated from a sample of normal potassium metal. Using a geometry of 21 ± 1 percent, calibrated by UX_2 and RaE-RaF counting, the measured activity at zero total absorber was found to be 33 ± 1 counts/min., with back-scattering negligible. 19.4-mg normal KCl, measured under similar circumstances, showed an activity of 3.0 ± 0.6 counts/min., entirely consistent with the value for the enriched sample.

From the observed activity of the enriched sample the partial disintegration constant for β-emission is

$$\lambda_\beta=(3.9\pm0.4)\times10^{-10}\ \text{yr.}^{-1};$$

this value is slightly smaller than that of Mülhoff,[8] smaller than that derived in a complicated manner by Ahrens and Evans,[6] and also much smaller than that of Bleuler and Gabriel.[5] The difference in the last case may well be due to back-scattering.[6]

2. *Beta-energy.* Using standard techniques[7,9] the Al absorption curve of the β-rays from the enriched sample was compared with the β-rays UX_2 and RaE. Comparison with the latter by the Feather method up to 60 percent of the range led to an extrapolated range of 900 mg/cm^2. The Feather analysis is shown in the insert in Fig. 1. This range corresponds to a maximum β-energy of 1.9 ± 0.2 Mev, slightly higher than the new value of 1.70 ± 0.15 Mev reported by Franchetti and Giovanozzi,[10] and substantially higher than older values[2,6,11,12] of 1.3–1.4 Mev.

Within the limits of experimental accuracy, the absorption curve of K^{40} is of identical shape to that of RaE. In contrast it is of markedly different shape from that of UX_2, having many more low energy electrons. The K^{40} to Ca^{40} transition involves a spin change of four and is usually assumed to be three or fourfold forbidden.[13] Since high resolution β-ray spectra will not be available until much more highly enriched K^{40} samples are available, the present comparison between the RaE spectrum, usually considered singly or doubly forbidden,[14] and K^{40} provides the best key to the shape of highly forbidden spectra.

3. *Assignment of the gamma-transition.* Ahrens and Evans[6] have determined that the γ-ray energy output from normal potassium is 4.9 ± 0.1 Mev/g-sec. Using the energy 1.55 ± 0.05 Mev,[2a] the disintegration constant for emission of a hard γ-ray is

$$\lambda_\gamma=(0.62\pm0.02)\times10^{-10}\ \text{yr.}^{-1}.$$

Previous authors[2,6,11] have considered that this gamma-ray is associated with the electron capture decay rather than with electron emission because of the supposed deficiency of mass difference between K^{40} and Ca^{40}. Our energy experiments reopen the question.

Dr. C. D. Coryell has pointed out to us that a consideration of the packing fractions in the series Si^{28}, S^{32}, A^{36}, and Ca^{40} suggests that Ca^{40} is more stable than A^{40} (*M*

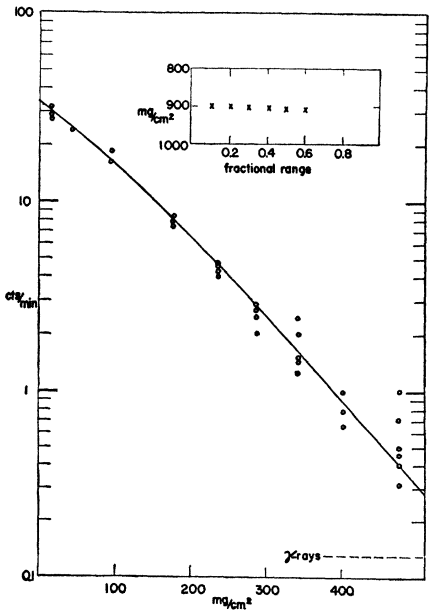

FIG. 1.

$=39.9755$),[15] making it more likely that γ-emission is associated with the emission of a very soft β-ray than with electron capture. This is also suggested by the twenty-shell argument given by Mayer.[16] The soft β-ray is probably not counted in the experiments in Part 1, nor in the coincidence studies of Meyer, Schwachheim, and deSouza Santos.[17] If this is true, λ_γ must be added to the experimental λ_β recorded in Part 1, whereas if the γ-emission is associated with a form of electron capture, λ_γ is included in λ_e calibrated by x-ray emission[5] or by argon production.[18]

4. *The yield of* A^{40}. Qualitative arguments and semi-quantitative calibrations[5,6] have shown that the partial disintegration constant for electron capture is of the same order of magnitude as that for electron emission, and that positron emission is less than 1 percent of electron emission.[5] Bleuler and Gabriel report $\lambda_e:\lambda_\beta=1.9\pm0.4$, and Ahrens and Evans report[7] 1.4 ± 0.2. Our modification of the value of λ_β would tend to support a higher ratio than the Ahrens and Evans analysis.

The argon content of a sample of Permian sylvite (KCl) of estimated age 2×10^8 yr. was determined by collection of the total permanent gas obtained on dissolving in air-free water. The total gas evolved was 0.0046 cm^3/g of K at S.T.P. No attempt was made to assay the argon content of the gas sample. If it be assumed that no A^{40} was lost from the sylvite, and that no other permanent gases were occluded in the mineral or introduced in the manipulations, the partial disintegration constant for electron capture would be

$$\lambda_e \simeq 2.8\times10^{-10}\ \text{yr.}^{-1},$$

if $\lambda_\beta+\lambda_\gamma=4.5\times10^{-10}$ is used for total β-emission. Essentially the same result is obtained if $\lambda_\beta=3.9\times10^{-10}$ is used, associating γ-emission with electron capture.

Contaminates in the extracted gases would make this value high whereas, more important, loss of A^{40} from the mineral would make this value low. Indeed, Suess[18] has reported much lower limits on the quantity of A^{40} in sylvine and carnallite, corresponding to $\lambda_e=3\times10^{-11}$ yr.$^{-1}$. It seems reasonable to suppose that λ_e lies between 2 and 12×10^{-10} yr.$^{-1}$ (between two-thirds and three times our observed value), corresponding to a range of $\lambda_e:\lambda_\beta$ from 0.5 to 3.

5. *Gross decay.* The summation of the decay constants λ_β, λ_γ, and λ_e gives the range $(5.9\text{–}16.5)\times10^{-10}$ yr.$^{-1}$ for the total disintegration constant, corresponding to a range in half-life $(4\text{–}12)\times10^8$ yr.; we have a subjective preference for a half-life of 6×10^8 yr. based on $\lambda_e=7\times10^{-10}$ yr.$^{-1}$ and $(\lambda_\beta+\lambda_\gamma)=4.5\times10^{-10}$ yr^{-1}. The estimated isotopic abundance of K^{40} at the time of nucleogenesis ($\sim3\times10^9$ B.C.) is, therefore, about 0.5 percent. The estimated current heat output of natural potassium from λ_β and an average β-energy of 0.6 Mev and the known γ-energy output[6] is now considered to be 20 microcal./yr. per gram of potassium, and the value at the time of petrogenesis ($\sim2.4\times10^9$ B.C.) was roughly sixteen times as great—300 microcal./yr. per gram of potassium.

* Research work done at Brookhaven National Laboratory under the auspices of the Atomic Energy Commission.

[1] The enriched K^{40} was produced by the Electromagnetic Separation Division, Carbide and Carbon Chemicals Company, Oak Ridge, Tennessee.

[2] (a) E. Gleditsch and T. Gráf, Phys. Rev. **72**, 640 (1947); (b) E. Gleditsch and T. Gráf, Phys. Rev. **72**, 641 (1947).
[3] F. Birch, Phys. Rev. **72**, 1128 (1947).
[4] C. Goodman and R. D. Evans, Bull. Geol. Soc. Am. **52**, 491 (1941).
[5] E. Bleuler and M. Gabriel, Helv. Phys. Acta **20**, 67 (1947).
[6] L. H. Ahrens and R. D. Evans, Phys. Rev. **74**, 279 (1948).
[7] C. D. Coryell, "*Use of Isotopes in Biology and Medicine*" (Univ. of Wisconsin Press, Madison, Wisconsin, 1948), Ch. VII, pp. 121, 128.
[8] W. Mühloff, Ann. d. Physik **7**, 205 (1930).
[9] L. E. Glendenin, Nucleonics **2**, 12 (1948).
[10] S. Franchetti and M. Giovanozzi, Rend. Acc. Linc. **1**, 1078 (1946); Phys. Rev. **74**, 102 (1948).
[11] O. Hirzel and H. Wäffler, Helv. Phys. Acta **19**, 216 (1946).
[12] B. Dzelepow, M. Kopjova, and E. Vorobjov, Comptes Rendus (U.S.S.R.) **52**, 121 (1946); Phys. Rev. **69**, 538 (1946).
[13] R. Marshak, Phys. Rev. **61**, 431 (1942).
[14] E. Greuling, Phys. Rev. **61**, 568 (1942).
[15] J. Mattauch, *Nuclear Physics Tables (1941)* (Interscience Publishers, Inc., New York, 1946).
[16] M. C. Mayer, Phys. Rev. **74**, 235 (1948).
[17] H. A. Meyer, G. Schwachheim, and M. D. deSouza Santos, Phys. Rev. **71**, 908 (1947).
[18] H. E. Suess, Phys. Rev. **73**, 1209 (1948).