

misgivings as to the validity of the assumption that $N(E)$ is constant in the region of the absorption line. It should be pointed out that, in view of the symmetry of σ_D with respect to E_r , $N(E)$ does not need to be constant over the region of the absorption line as long as it is linear, i.e., as long as the higher derivatives of $N(E)$ with respect to E are small. Some estimate of the magnitude of the higher derivatives of $N(E)$ may be obtained by a study of the change in the resonance fluorescence effect upon heating of the nickel scatterer. This temperature effect depends on the higher derivatives in closely the same way as the self-absorption experiment. A small temperature dependence of the resonance effect implies a small correction to the simple expression (4) used for the analysis of the self-absorption.

In a separate experiment the scatterer was heated from room temperature to 300°C and the change in the resonance fluorescence effect was found to be smaller than 5 percent. This indicates that the error introduced in the self-absorption experiment by assuming $N(E)$ to be constant over the region of the absorption line is small indeed.

CONCLUSIONS

The resonance fluorescence self-absorption method allows the determination of lifetimes even if the line shape of the exciting radiation is not known because of the complexity of the recoil phenomena.

The mean life of $(1.1 \pm 0.2) \times 10^{-12}$ second measured for the 1.33-Mev excited state of Ni⁶⁰ is approximately five times shorter than the single-particle estimate.⁹ Compared with other electric quadrupole transitions in the weak coupling region of the periodic table,^{10,5} this transition is slowed down by a factor of from two to five. This slowing down is not unexpected in view of the magic number of protons in Ni⁶⁰.

ACKNOWLEDGMENTS

The author wishes to thank Mr. W. B. Todd for his help in taking the data and in maintaining the equipment, Dr. Leonard Eisenbud for valuable discussions, and Mr. F. B. Thiess for aid with the computations.

⁹ V. F. Weisskopf, *Phys. Rev.* **83**, 1073 (1951).

¹⁰ G. M. Temmer and N. P. Heydenburg, *Phys. Rev.* **99**, 1609 (1955).

Decay Constants of K⁴⁰ as Determined by the Radiogenic Argon Content of Potassium Minerals

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(Received March 28, 1956)

It is shown that the potassium-argon age of young minerals depends almost linearly on the decay constant for electron capture in K⁴⁰ and is very insensitive to the decay constant for beta emission. This fact permits calculation of λ_e by comparing the concordant uranium-lead age of cogenetic uraninite with A⁴⁰/K⁴⁰ ratios found in young samples of mica. It is found that $\lambda_e = (0.557 \pm 0.026) \times 10^{-10} \text{ yr}^{-1}$. Similar comparisons with older mica samples indicate that satisfactory agreement with the uraninite ages are obtained by use of this value of λ_e together with $\lambda_\beta = (0.472 \pm 0.05) \times 10^{-9} \text{ yr}^{-1}$. It is concluded that there is no conflict between the decay constants inferred by this geological method and those found by direct counting experiments.

I. INTRODUCTION

A NUMBER of papers have appeared in which the branching ratio of K⁴⁰ has been determined by measurement of the radiogenic argon content of potassium minerals. Aldrich and Nier¹ showed, by semi-quantitative measurements of the ratio of radiogenic A⁴⁰ to K⁴⁰ in minerals of approximately known geological age, that the branching ratio was of the order of 0.1.

Earlier counting experiments² had indicated that the branching ratio was of the order of 1. Inghram, Brown, Patterson, and Hess³ determined the branching ratio to be 0.126 by measuring the ratio of radiogenic argon to radiogenic calcium in sylvite (KCl). Subsequently Mousuf⁴ and Russell, Shillibeer, Farquhar, and Mousuf⁵ published a branching ratio of 0.06 determined by comparing A⁴⁰/K⁴⁰ ratios from feldspars with various age

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¹ L. T. Aldrich and Alfred O. Nier, *Phys. Rev.* **74**, 876 (1948)..

² E. Bleuler and M. Gabriel, *Helv. Phys. Acta* **20**, 67 (1947).

³ Inghram, Brown, Patterson, and Hess, *Phys. Rev.* **80**, 916 (1950).

⁴ A. K. Mousuf, *Phys. Rev.* **88**, 150 (1952).

⁵ Russell, Shillibeer, Farquhar, and Mousuf, *Phys. Rev.* **91**, 1223 (1953).

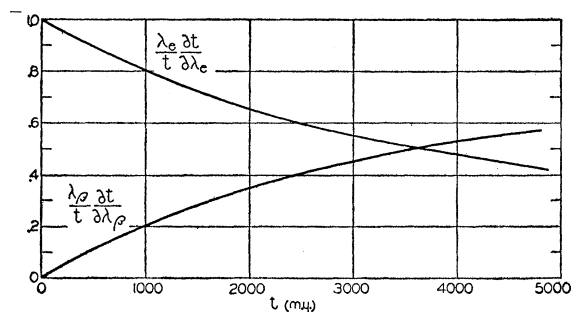


FIG. 1. Graph showing the relative sensitivity of the measured age t (in millions of years) to changes in the decay constants λ_e and λ_β .

determinations on minerals believed to be cogenetic. Wasserburg and Hayden⁶ then showed that these workers failed to extract all of the argon from the minerals and that when the uncertainties in the comparison ages are considered, the geological measurements were consistent with a branching ratio as high as 0.13.

Later, Wasserburg and Hayden⁷ showed by a number of argon determinations on feldspars compared with the concordant Pb-U ages of cogenetic uraninites ($t_{U235} = t_{U238}$) that a branching ratio of 0.085 fitted their data well over a large span of geological time. In this work Wasserburg and Hayden assumed the β -disintegration constant $\lambda_\beta = 5.07 \times 10^{-10} \text{ yr}^{-1}$, and their branching ratio then corresponds to a value of $\lambda_e = \lambda_\gamma = 0.43 \times 10^{-10} \text{ yr}^{-1}$ corresponding to a specific gamma activity of 2.5 gamma rays per second per gram of potassium. While this is a very low value for the specific activity, it is difficult to exclude it in view of the uncertainties in the counting experiments. It was then found by Wetherill, Aldrich, and Davis⁸ that mica samples characteristically had higher A^{40}/K^{40} ratios than feldspar samples obtained from the same rock. This observation has subsequently been confirmed in other laboratories.^{9,10} For every pair of cogenetic micas and feldspars investigated, it was found that the micas gave the greater A^{40}/K^{40} ratio. The authors believe that this effect is the result of a loss of radiogenic A^{40} from potassium feldspars in comparison to micas.

The alternative hypothesis that micas have preferentially "adsorbed" A^{40} during their formation would seem to be excluded by the regularity of the results obtained from various mica samples of different ages.

When these measurements were reported, there was an insufficient number of comparisons between con-

cordant uranium-lead ages and mica A^{40}/K^{40} determinations to permit a re-evaluation of the question of the decay constants. A number of these comparisons has now been made independently by the Chicago and Carnegie Institution workers, and their data are combined in this report.¹¹

II. CALCULATION OF THE DECAY CONSTANTS FROM GEOLOGIC DATA

The sensitivity of the calculated age

$$t = \frac{1}{\lambda_e + \lambda_\beta} \ln \left\{ 1 + \frac{A^{40}}{K^{40}} \left(\frac{\lambda_e + \lambda_\beta}{\lambda_e} \right) \right\}$$

to changes in the decay constants λ_e and λ_β may be seen from the graphs of $(\lambda_e/t)(\partial t/\partial \lambda_e)$ and $(\lambda_\beta/t)(\partial t/\partial \lambda_\beta)$ as functions of t (Fig. 1). It may be shown that

$$\left[\frac{\lambda_e}{t} \frac{\partial t}{\partial \lambda_e} + \frac{\lambda_\beta}{t} \frac{\partial t}{\partial \lambda_\beta} \right] = -1.$$

While the curves are drawn for a particular value of λ_e and λ_β ,¹² the qualitative effects are the same for any reasonable value of these decay constants. It is seen that for young minerals $(\lambda_e/t)(\partial t/\partial \lambda_e) \sim -1$, indicating that an increase in λ_e will cause a proportionate decrease in t . For older minerals the dependence of t on λ_e decreases, while the dependence of t on λ_β increases. At $t = 3500$ million years, a 10% change in either λ_e or λ_β will cause a 5% change in t . It may also be seen that for young minerals, t is relatively independent of λ_β . For this reason it is most desirable to use the geological data to compute λ_e instead of λ_e/λ_β , as earlier workers have done. Thus, comparison of the A^{40}/K^{40} ratio of a young mica with a concordant uranium-lead age will permit the calculation of λ_e with very little regard to λ_β . This has been done for samples 1-4 in Table I, and gives an average $\lambda_e = (0.557 \pm 0.026) \times 10^{-10} \text{ yr}^{-1}$ corresponding to a specific gamma activity of 3.24 ± 0.15 gamma rays per second per gram of natural potassium. This may be compared with recent values of the specific activity shown in Table II.

There is no corresponding direct way of determining λ_β from radiogenic argon measurements on available mineral samples. In Table I, we list argon ages calculated by using $\lambda_e = 0.557 \times 10^{-10} \text{ yr}^{-1}$ as determined from the young minerals, and $\lambda_\beta = 0.472 \times 10^{-9} \text{ yr}^{-1}$ which is the average calculated by Endt and Kluver in a review of previous studies of these decay con-

⁶ G. J. Wasserburg and R. J. Hayden, *Phys. Rev.* **93**, 645 (1954).

⁷ G. J. Wasserburg and R. J. Hayden, *Geochim. et Cosmochim. Acta* **7**, 51 (1955).

⁸ Wetherill, Aldrich, and Davis, *Geochim. et Cosmochim. Acta* **8**, 171 (1955).

⁹ G. J. Wasserburg and R. J. Hayden, *Geochim. et Cosmochim. Acta* (to be published).

¹⁰ J. Reynolds, paper presented at Pennsylvania State College Conference on Nuclear Geophysics (unpublished).

¹¹ Argon analyses were made using mass spectrometric isotope dilution technique, while potassium, uranium, and lead analyses were made either using isotope dilution or by gravimetric chemical analysis. Lead isotope abundances were determined with a mass spectrometer. More complete discussion of the experimental methods and the geological significance of the results will be published in the geological literature.

¹² Using $\lambda_e = 0.549 \times 10^{-10} \text{ yr}^{-1}$ and $\lambda_\beta = 0.474 \times 10^{-9} \text{ yr}^{-1}$.

stants.¹³ Although sample No. 9 shows evidence for loss of argon, the general agreement of the argon ages with concordant uranium-lead ages is seen to be good over a great range of time. However, as can be seen from Fig. 1, this agreement is not very sensitive to the choice of λ_β . The agreement would still be acceptable if λ_β were changed by 10%. Taking into consideration the uncertainty in both λ_β and λ_e , the geological evidence gives a branching ratio of 0.117 ± 0.015 , the principal source of uncertainty being the uncertain value of λ_β .

One of the most serious problems to be faced in the determination of mineral ages by the argon method is the question of whether or not the radiogenic argon has been quantitatively retained over the great expanse of geologic time. Any loss of argon will result in the calculation of a value of λ_e which is too low. In the earlier work on feldspars,⁷ it was not possible to distinguish between the two possibilities, that argon has been lost from the mineral or alternatively that the true value of λ_e was actually near the lower limit set by counting experiments. The situation has now improved insofar as the value of λ_e required by the radiogenic argon content of the micas is near the average of the values determined by counting experiments.

III. CONCLUSIONS

The value of the specific electron capture rate of potassium as determined from potassium-argon measurements of mica from young dated pegmatites is 3.24 ± 0.15 electron captures per gram per second, assuming that radiogenic argon has been quantitatively retained by these samples.

TABLE I. Comparison of K-A and U-Pb ages (using $\lambda_e = 0.557 \times 10^{-10} \text{ yr}^{-1}$ and $\lambda_\beta = 0.472 \times 10^{-9} \text{ yr}^{-1}$).

Sample	A ⁴⁰ /K ⁴⁰	K-A age (10 ⁶ yr)	U-Pb age (10 ⁶ yr)
1. Portland, Conn.	0.0158	265 ± 8	267 ± 5
2. Glastonbury, Conn.	0.0156	259 ± 8	255 ± 5
3. Spruce Pine, N. C.	0.0213	349 ± 15	375 ± 10
4. Branchville, Conn.	0.0236	382 ± 11	367 ± 7
5. Parry Sound, Ontario	0.0705	970 ± 30	994 ± 20
6. Cardiff Twp., Ontario	0.0729	1000 ± 50	1020 ± 20
7. Wilberforce, Ontario	0.0695	960 ± 50	1030 ± 30
8. Keystone, S. Dakota	0.140	1600 ± 80	1600 ± 30
9. Keystone, S. Dakota	0.119	1430 ± 70	1600 ± 30
10. Viking Lake, Sask.	0.175	1850 ± 80	1890 ± 40
11. Bikita, S. Rhodesia	0.300	2550 ± 150	2650 ± 100 ^a
12. Forest City Stone Meteorite	0.996	4370 ± 60 ^b	4500 ± 100 ^c

^a A. Holmes, Nature 173, 612 (1954).
^b G. J. Wasserburg and R. J. Hayden, Phys. Rev. 97, 86 (1955).
^c C. C. Patterson, Geochim. et Cosmochim. Acta 7, 151 (1955).

¹³ P. M. Endt and J. C. Kluyver, Revs. Modern Phys. 26, 95 (1954).

TABLE II. Determinations of the specific gamma and beta activity of natural potassium.

Investigator	γ /g sec	β /g sec
Gleditsch and Graf ^a	3.6 ± 0.8	
Graf ^b		26.8 ± 1.2
Ahrens and Evans ^c	3.42 ± 0.07 ^r	
Hess and Roll ^d	2.6	
Stout ^e		30.6 ± 2.0
Sawyer and Wiedenbeck ^f	2.88 ± 0.3 ^s	
Spiers ^g	2.97	30.5
Faust ^h	3.6 ± 0.4	31.2 ± 3.0
Graf ⁱ	3.4 ± 0.5	
Houtermans, Haxel, and Heintze ^j	3.1 ± 0.3	27.1 ± 1.5
Smaller, May, and Freedman ^k		22.5 ± 0.7
Sawyer and Wiedenbeck ^l		28.3 ± 1.0
Good ^m		27.1 ± 0.6
Delaney ⁿ		32.0 ± 3
Burch ^o	3.37 ± 0.09	
Suttle and Libby ^p	2.96 ± 0.3	29.6 ± 0.7
McNair, Glover, and Wilson ^q	3.33 ± 0.15 ^r	
This paper	3.24 ± 0.15	

^a E. Gleditsch and T. Graf, Phys. Rev. 72, 640 (1947).
^b T. Graf, Phys. Rev. 74, 831 (1948).
^c L. H. Ahrens and R. D. Evans, Phys. Rev. 74, 279 (1948).
^d V. F. Hess and J. D. Roll, Phys. Rev. 73, 916 (1948).
^e R. W. Stout, Phys. Rev. 75, 1107 (1949).
^f G. A. Sawyer and M. L. Wiedenbeck, Phys. Rev. 76, 1535 (1950).
^g F. W. Spiers, Nature 165, 356 (1950).
^h W. R. Faust, Phys. Rev. 78, 624 (1950).
ⁱ T. Graf, Rev. Sci. Instr. 21, 285 (1950).
^j Houtermans, Haxel, and Heintze, Z. Physik 128, 657 (1950).
^k Smaller, May, and Freedman, Phys. Rev. 79, 940 (1950).
^l G. A. Sawyer and M. L. Wiedenbeck, Phys. Rev. 79, 490 (1950).
^m M. L. Good, Phys. Rev. 83, 1054 (1951).
ⁿ C. F. G. Delaney, Phys. Rev. 82, 158 (1951).
^o P. R. A. Burch, Nature 172, 361 (1953).
^p A. Suttle and W. F. Libby, Analyt. Chem. 27, 921 (1955).
^q McNair, Glover, and Wilson, Phil. Mag. 1, 199 (1956).
^r Recalculated using $E_\gamma = 1.46$ Mev.
^s Recalculated using $\lambda_\gamma/\lambda_\beta = 0.20$ for K⁴².
^t Recalculated using their $\lambda_e/\lambda_\beta = 0.121$ and their specific beta activity of 27.5/g sec.

Using data obtained from older pegmatites, a specific beta activity of 27.6 ± 3.0 β particles per gram per second is seen to be consistent with the data. Thus geological measurements give a branching ratio of 0.117 ± 0.015 . The agreement of these values with the results of counting experiments indicates that the potassium-argon ages of micas are close to being absolute ages and that leakage of argon has not been very serious.

ACKNOWLEDGMENTS

Some of these results were obtained in connection with the joint program of mineral age determination of the Department of Terrestrial Magnetism and the Geophysical Laboratory, Carnegie Institution of Washington. Part of the work reported here was supported by the U. S. Atomic Energy Commission and the National Science Foundation. Stable isotopes used in the isotope dilution analyses of potassium, uranium, and lead were obtained on loan from the U. S. Atomic Energy Commission.