to the l=2 reduced width. Thus the  $\frac{5}{2}$  - assignment is excluded, and the level is established definitely as  $\frac{5}{2}$ +.

The transition involved is presumably, therefore, a  $d_{\frac{1}{2}} \rightarrow s_{\frac{1}{2}}$  neutron transition outside a C<sup>14</sup> core.<sup>28</sup> The observed transition speed is about 0.4 times the Weisskopf estimate for a single proton. This implies configuration mixing in the wave function of the odd neutron, and the participation of parent states in C<sup>14</sup> having a substantial E2 radiative width to the  $C^{14}$  ground state. Two 2+ states in C<sup>14</sup> are predicted<sup>29</sup> at excitations of 7.01 and 8.32 Mev, but too little is known about their radiative widths to enable quantitative estimates to be made of their effect on the  $C^{15}$  lifetime.

On the weak-coupling collective model, the transition is considered to take place through oscillations of the nuclear surface induced by the odd neutron. Thus the present result for C<sup>15</sup> should be directly related to lifetimes for other  $d_{\frac{1}{2}} \leftrightarrow s_{\frac{1}{2}}$ , single neutron transitions, provided the surface deformability is the same in each nucleus considered. The model has been examined in detail for nuclei in this mass region by Raz.<sup>2</sup> Taking the value for the surface deformability parameter C deduced by Raz for the  $s_{\frac{1}{2}} \rightarrow d_{\frac{1}{2}}$  neutron transition in O<sup>17</sup>, the C<sup>15</sup> lifetime is predicted<sup>30</sup> to be  $(5.20\pm0.37)$  nsec. A reasonable explanation for this discrepancy is that the param-

<sup>28</sup> C.f. E. C. Halbert, Ph.D. thesis, University of Rochester, <sup>29</sup> E. K. Warburton and W. T. Pinkston, Phys. Rev. 118, 733

(1960).

<sup>30</sup> In reference 2, the value of the matrix element of the surface coupling term,  $\langle 2s | k | 1d \rangle$ , was assumed to be constant for nuclei in this mass region. In the present analysis, values of this matrix element computed by G. R. Satchler (private communication through B. J. Raz) were used. The computed values deviate from constancy by about 20% over the three nuclei considered here. eter C is lower for the  $C^{14}$  core in  $C^{15}$  than for the  $O^{16}$ core in  $O^{17}$  by a factor 0.85. This corresponds to a more easily deformable core, and a change in this direction is expected when moving away from a closed shell.

#### SUMMARY OF RESULTS

The results of the present experiments are summarized here. The lifetime for the  $\frac{5}{2} + \rightarrow \frac{1}{2} +$  transition in C<sup>15</sup> was found to be

$$\tau_m = (3.73 \pm 0.23)$$
 nsec,

and for the  $1 + \rightarrow 3 +$  transition in B<sup>10</sup>,

 $\tau_m = (1.04 \pm 0.02)$  nsec.

Alternatively, as suggested by Wilkinson,<sup>1</sup> these may be expressed in terms of the Weisskopf units with

$$r_0 = 1.2 \times 10^{-13} \text{ cm}$$
:

For C<sup>15</sup>,

and for B10,

 $\Gamma_{\gamma} = (3.17 \pm 0.07) \Gamma_{W}.$ 

 $\Gamma_{\gamma} = (0.431 \pm 0.026) \Gamma_{W};$ 

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## Positron Emission in the Decay of $K^{40}$ <sup>†</sup>

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Positron emission has been detected in the decay of  $K^{40}$  in a triple coincidence experiment using a liquidscintillation positron detector and two NaI(Tl) annihilation gamma detectors. A ratio of  $\beta^+/\beta^- = (1.12 \pm 0.14)$  $\times 10^{-5}$  was found. This corresponds to a ratio of the squared matrix elements of  $M_{+2}^2/M_{-2}^2 = 0.18 \pm 0.03$ which is in substantial agreement with a theoretical estimate of  $\frac{1}{2}$ . The effect of this result on the decay constants of K40 is discussed.

### INTRODUCTION

NUMBER of investigators<sup>1-5</sup> have searched for positron emission from  $K^{40}$ . The most nearly definitive result was obtained by Tilley and Madansky<sup>5</sup>

who reported an upper limit of  $(1.3\pm0.7)\times10^{-5}$  on the ratio of positron to negatron emission. In their experiments a large scintillation crystal of KI (natural abundance) which served as both the source and positron detector was placed between two NaI(Tl) scintillation counters which detected annihilation quanta. Pair production in the KI crystal by the 1.46-Mev gamma of K<sup>40</sup> set the ultimate limit to their detection sensitivity. With potassium highly enriched in K<sup>40</sup> readily available it seemed worthwhile to repeat their experi-

<sup>†</sup> Based on work performed under the auspices of the U.S. Atomic Energy Commission.

 <sup>&</sup>lt;sup>1</sup> P. R. Bell and J. M. Cassidy, Phys. Rev. 77, 409 (1950).
<sup>2</sup> P. R. Bell and J. M. Cassidy, Phys. Rev. 79, 173 (1950).
<sup>3</sup> S. A. Colgate, Phys. Rev. 81, 1063 (1951).
<sup>4</sup> M. L. Good, Phys. Rev. 81, 1058 (1951).
<sup>4</sup> D. B. Ciller and L. McLacher, Phys. Rev. 70, 126 (1951).

<sup>&</sup>lt;sup>5</sup> D. R. Tilley and L. Madansky, Phys. Rev. 116, 413 (1959).

TABLE I. Composition of scintillator solutions.

Sample No.	1	2
Observed $\beta^-/\min$	141 100	81 900
K <sup>40</sup> (mg)	9.91	5.75
Total K (mg)	33.4	19.4
Na (mg)	16.4	0
C (g)	4.75	4.75
H (g)	0.45	0.45
Volume (ml)	6.0	6.0

ments using  $K^{40}$  dissolved in a small central liquid scintillation counter to minimize pair production.

Much of the interest in positron branching in  $K^{40}$ stems from the use of  $K^{40}$  in dating minerals by means of  $Ar^{40}/K^{40}$  ratios. Although the above upper limit shows that positron emission itself cannot contribute appreciably to the formation of  $Ar^{40}$ , positron emission could serve as an indicator of a possible *K*-capture branch to the ground state of  $Ar^{40}$  (see Fig. 6). Calculations by Brosi and Ketelle<sup>6</sup> show that the electron capture branch to the ground state should be 155 times as intense as the positron branch for a maximum positron energy (including rest mass) of  $1.961m_0c^2$ .

## EXPERIMENTAL PROCEDURE

A block diagram of the apparatus is shown in Fig. 1. Potassium<sup>7</sup> enriched to  $(29.7 \pm 0.1)\%$  in K<sup>40</sup> in the form of the octoate salt was dissolved in liquid scintillator {3g/liter terphenyl+0.1 g/liter 1, 4-di-[2-(5-phenyloxazolyl)]-benzene (POPOP) in toluene} in the central beta counter. Annihilation quanta were detected by two  $2\frac{1}{2}$ -in. diam  $\times 2\frac{1}{4}$ -in. high NaI(Tl) crystals on either side of the central beta detector. The singlechannel analyzers associated with the NaI(Tl) detectors accepted pulses in an energy band 70-kev wide centered on the 0.51-Mev photopeaks. If the coincidence and anticoincidence requirements of an event were met, the pulse from the liquid scintillation counter was recorded in the 256-channel analyzer. The positron detection efficiency was checked periodically by substituting for the K40 cell a similar cell containing Na22 dissolved in liquid scintillator. The efficiency varied from 2.7% to 2.8% during the course of the experiment. Two samples enriched in K<sup>40</sup> gave net coincidence rates of 0.10 and 0.06 count per minute above a background of 0.006 count per minute with normal potassium in the cell. In one experiment one of the single-channel analyzers was deliberately set above the 0.51-Mev position, at 0.64 Mev. A counting rate equal to background was obtained, showing that the coincidences obtained with K<sup>40</sup> are associated with gammas of about 0.51 Mev and not with a continuous distribution in the region of 0.51 Mev.



FIG. 1. Block diagram of apparatus.

Energy calibration of the liquid scintillators was accomplished by means of the Compton edges of the 0.511- and 1.277-Mev gammas from an external source of Na<sup>22</sup>. The method was checked by comparing the Na<sup>22</sup> Compton edges in a liquid scintillator containing Sn<sup>113</sup> with the position of the *K*-conversion electron line from the 392-kev gamma ( $Kx-e^-$  coincidences were used to isolate the *K* line). It was found that the channel number indicated by the midpoint on the slope of the Compton edge should be decreased by 3% to obtain the proper energy calibration factor. It has been shown<sup>8</sup> that the response of the liquid scintillator to monoenergetic electrons is linear above 100 kev with a positive intercept of 20 kev on the energy axis. The relationships used in this paper are:

Pulse height = k(E-20) for single electrons or positrons, pulse height = k(E-40) for positron-electron pairs,

where E is the electron or total pair energy and k is the calibration factor for a particular solution. The intercept for positron-electron pairs is twice that for a single particle since we are considering two particles, each of which has an effective energy 20 kev less than its actual energy.

#### RESULTS

Two different  $K^{40}$  liquid-scintillator solutions having the compositions listed in Table I were prepared. The presence of sodium in the initial sample was discovered when gross beta-counting data on sample 1 gave a much lower specific activity than should have been expected. Spectroscopic analysis showed that essentially the only impurity was sodium. The material was then purified by precipitation of potassium as KClO<sub>4</sub> from an alcohol-HClO<sub>4</sub> solution, leaving the sodium in solution. Sample 2, made from the purified potassium, showed the expected specific activity. The presence of inactive material in sample 1 does not affect the results since the amount of  $K^{40}$  in each solution was determined from the integral beta-counting rate in the liquid scintillator using a specific activity of  $28.0\beta^{-}$  dis/sec

<sup>&</sup>lt;sup>6</sup> A. R. Brosi and B. H. Kettelle (private communication).

<sup>&</sup>lt;sup>7</sup> The enriched potassium was supplied by the Stable Isotopes Division of the U. S. Atomic Energy Commission, Oak Ridge, Tennessee.

<sup>&</sup>lt;sup>8</sup> K. F. Flynn, L. E. Glendenin, E. P. Steinberg, and P. Wright (to be published).



FIG. 2. External pair spectrum from 1.53-Mev gamma of K42.

per gram of natural K, a K<sup>40</sup> natural abundance of 0.0118%<sup>9</sup> and 29.7% for the K<sup>40</sup> abundance in our enriched material.

The only significant source of interfering positrons to be expected is from internal and external pairs created by the 1.46-Mev gamma in the electron capture branch. Pairs accompanying the  $\beta^-$  branch should have an intensity less than 10<sup>-9</sup> per beta.<sup>10</sup> Pairs created within the liquid scintillator should appear as a monoenergetic line since the total kinetic energy of the pair equals the gamma energy minus 1.02 Mev or 0.44 Mev. Figure 2 shows the pair spectrum obtained by exposing a liquid scintillator containing normal potassium to a collimated beam of 1.53-Mev gammas from an external source of K<sup>42</sup>. Coincidence requirements were the same as in the K<sup>40</sup> experiment so that only events producing positrons should have been detected. The spectrum consists of a full-energy peak containing 88% of the pulses and a low-energy tail arising from those events



FIG. 3. Gross positron spectrum of K<sup>40</sup>.

9 Way, Everling, Fuller, Gove, McGinnis, and Nakasima, "Nuclear Data Sheets, No. 59-4-36," National Academy of Sciences—National Research Council, Washington, D. C. <sup>10</sup> K. Huang, Phys. Rev. **102**, 422 (1956).

in which some of the energy of the pair is lost outside the scintillator volume. The pulses above the full energy peak are probably due to traces of radioactive impurities in the  $K^{42}$  sample.

Counting data for the two K<sup>40</sup> samples are given in Table II. Since pair production contributes heavily to the total positron count the results are rather sensitive to the estimation of the pair production. Details of the pair production calculation are given in the Appendix. Fortunately, most of the pair pulses occur in a single peak near the expected upper limit of the nuclear positron spectrum so that the two sources of positrons may be distinguished. The error attached to the final  $\beta^+/\beta^-$  ratio should be interpreted as a standard deviation, and is due partly to counting statistics and partly to estimated errors in the cross sections and other constants used in calculating the pair peak intensity.

The gross triple coincidence spectra from both samples of  $K^{40}$  are summed in Fig. 3. The background spectrum and the calculated pair spectrum for the 1.46-Mev gamma are also shown. The K<sup>40</sup> pair spectrum



FIG. 4. Net positron spectrum of K<sup>40</sup>.

was assumed to have the same shape as the K<sup>42</sup> external pair spectrum, Fig. 2, normalized to the peak position and to the total number of counts calculated theoretically for internal and external pair production in K<sup>40</sup>. After subtracting background and pair spectra the spectrum of Fig. 4 is obtained representing 786 net positron counts. Since it did not seem worthwhile to attempt a Fermi analysis with so few counts, the  $N_E$ spectrum was compared with the  $N_E$  spectrum expected for a third forbidden transition with an end point of 491 kev.

The theoretical spectrum was computed using the  $C_{3A}$  correction term for a third-forbidden unique positron spectrum<sup>11</sup> and has been corrected for finite detector resolution and normalized to the same area as the experimental spectrum. Within the wide limits imposed by counting statistics and the uncertainty introduced by subtraction of the pair spectrum, the agreement is satisfactory. The cross-hatched region represents an estimate of the number of counts missed by the multichannel analyzer in the low-energy region.

<sup>&</sup>lt;sup>11</sup> E. Greuling, Phys. Rev. 61, 568 (1942).



Some spectral distortion might be expected as a consequence of the escape of beta particles from the liquid scintillator. To determine the significance of this effect, the positron spectrum of Na<sup>22</sup> (allowed shape,  $E_{\rm max}$ =541 kev) was measured in the same way as the K<sup>40</sup>. It was necessary to subtract a small background due to occasional summing of the 1.28-Mev gamma Compton distribution with the positron pulses. After correction for finite detector resolution<sup>12</sup> the Kurie plot of Fig. 5 was obtained. The linearity of the plot demonstrates that no serious spectral distortion is caused by particle escape for maximum beta energies up to at least 0.5 Mev.

TABLE II. K<sup>40</sup> counting data.

Sample	1	2
Extrapolated beta+gamma singles rate		
(dis/min)	141 700	82 200
Calculated gamma singles rate (dis/min)	600	-300
Net beta singles rate (dis/min)	141 100	81 900
Coincidence counting time (min)	6950	19 050
Gross coincidence rate (counts/min)	0.1023	0.0640
Background coincidence rate, natural K		
(counts/min)	0.0045	0.0045
Net coincidence rate (counts/min)	0.0978	0.0595
Positron detection efficiency	0.027	0.028
Positrons plus pairs (dis/min)	3.62	2.12
Calculated pair rate (dis/min)	2.08	1.21
Net positron rate (dis/min)	1.54	0.91
Estimated $\beta^+$ missed at low energies		
(dis/min)	0.03	0.02
Total $\beta^+$ rate (dis/min)	1.57	0.93
$(\beta^+/\beta^-) \times 10^5$	1.11	1.14
$\tilde{A}$ verage $(\beta^+/\beta^-) \times 10^5$	1.1	$2 \pm 0.14$

 $^{12}$  G. E. Owen and H. Primakoff, Phys. Rev. 74, 1406 (1948). Only the second derivative term was used since it has been found empirically (see reference 8) that this treatment gives the best results.

### DISCUSSION

The decay scheme of Fig. 6 is based on data presented by Way *et al.*<sup>9</sup> and on our results for the intensity of the positron branch. An electron-capture-to-ground-state to positron ratio of 155 was assumed. The electroncapture rate to the ground state is then 1.6% of the electron-capture rate to the 1.46-Mev excited state and is, therefore, of only slight significance for mineral dating by the Ar<sup>40</sup>/K<sup>40</sup> method. Since both the  $\beta^+$  and  $\beta^-$  transitions are third-forbidden unique, it is possible to calculate the ratio of the squares of the matrix elements  $M_+^2/M_-^2$  from the measured  $\beta^+$  to  $\beta^-$  ratio. Following the procedure outlined by Tilley and Madansky<sup>5</sup> and using the decay energies shown in Fig. 6 we arrive at the value:

# $M_{+}^{2}/M_{-}^{2}=0.18\pm0.03.$

The percentage error assigned to the ratio of the squared matrix elements is larger than that of the  $\beta^+/\beta^-$  ratio



FIG. 6. Decay scheme of K<sup>40</sup>.

because of the additional uncertainty introduced by possible errors in the decay energies.

Kurath<sup>13</sup> has shown that if the ground states of Ar<sup>40</sup>,  $K^{40}$ , and  $Ca^{40}$  are assigned pure jj configurations, the ratio of the squared reduced matrix elements  $M_{+}^{2}/M_{-}^{2}$ should equal  $\frac{1}{8}$ . The difference between the theoretical value and the experimental result is within the range of possible experimental error.

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The authors wish to thank Dr. Dieter Kurath for calculating the theoretical ratio of the matrix elements for  $\beta^+$  and  $\beta^-$  decay and to Dr. F. Porter and Dr. M. S. Freedman for assistance in formulating the calculation of the beta-spectrum shape-correction factors.

#### APPENDIX

The number of external pairs produced in the liquid scintillator per K40 disintegration depends upon the number of 1.46-Mev gammas per  $\beta^-$ , the fraction of these gammas stopped in the liquid scintillator, and the ratio of pair production to total cross sections for the gammas in the liquid scintillator. The number of 1.46-Mev gammas per  $\beta^-$ , 0.123, was taken from the NRC data tables.<sup>9</sup> Case et al.<sup>14</sup> present a table of the probability of an interaction within a uniform spherical source as a function of a/l, where a is the radius of the sphere and l is the mean free path of the radiation in the material. They show that the interaction probability Pis quite insensitive to the exact geometrical shape:

<sup>13</sup> D. Kurath, Argonne National Laboratory Physics Division Summary Report for November and December, 1961 (unpublished).

<sup>14</sup> K. M. Case, F. de Hoffman, and G. Placzek, Introduction to the Theory of Neutron Diffusion (U.S. Government Printing Office, Washington, D. C., 1953), Vol. 1.

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TABLE III. Pair production in liquid scintillator.

a (6 ml volume)	1.126 cm
l (for toluene, 1.46-Mey $\gamma$ )	20.28 cm
P (cylinder)	0.0382
$\sigma_{\rm nair}/\sigma_{\rm total}$ 5.95×10 <sup>-5</sup> /4.93×10 <sup>-2</sup> =	$= 1.207 \times 10^{-3}$
External pair production in liquid	
scintillator	4.61×10 <sup>-5</sup> per gamma
External pair production in walls	$0.16 \times 10^{-5}$ per gamma
Internal pair production	7.2 $\times 10^{-5}$ per gamma
Total pair production	12.0 ×10 <sup>-5</sup> per gamma
1 1	1 0

P(hemisphere) is 0.945 times P(sphere) and P(tetrahedron) is 0.944 times P(sphere) for equal volumes. For our liquid scintillator which approximated a cylinder of height equal to its radius, we assumed that P(cylinder)was 0.945 times P(sphere) of the same volume. All cross sections were interpolated from the tables of Davisson and Evans,<sup>15</sup> applied to scintillator solutions having the compositions listed in Table I, and include pair production in the potassium and sodium. A rough estimate was also made of the probability that a pair would be produced in the glass walls of the liquid scintillator cell and that at least one member of the pair would reach the liquid scintillator. This estimate is entered in Table III. Exact calculations of internal pair-production cross sections have been made by Jaeger and Hulme<sup>16</sup> for Z=0and Z=84 for E2 radiation. The values at 1.46 Mev for Z=0 and Z=84 were read from their graph and introduced into Brimberg's<sup>17</sup> interpolation formula which yielded an average value of  $7.2 \times 10^{-5}$  pairs per gamma at Z = 18.

<sup>15</sup> C. M. Davisson and R. D. Evans, Revs. Modern Phys. 24, 79

(1952). <sup>16</sup> J. C. Jaeger and H. R. Hulme, Proc. Roy. Soc. (London) A148, 708 (1935). <sup>17</sup> S. A. S. Brimberg, Phys. Rev. 87, 150 (1952).

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## Photoproton Reaction in Be<sup>9</sup><sup>†</sup>

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The reaction  $Be^{9}(\gamma, p)Li^{8}$  was measured from threshold (16.89 Mev) up to 57 Mev using the bremsstrahlung beam from the Iowa State University electron synchrotron. The data were taken in energy steps of  $\sim 0.050$  Mev for the first several Mev and in energy steps of  $\sim 1.0$  Mev for the remainder of the yield curve. A number of small resonances near threshold was observed. The giant resonance cross section reaches a peak value of  $2.64\pm0.30$  mb at an energy of 23 Mev, and it possesses a large high-energy tail. The integrated cross section to 56.8 Mev is 41.4±4.6 Mev-mb.

### INTRODUCTION

N 1953 Haslam et al.<sup>1</sup> published the results of their measurement of the  $Be^{9}(\gamma, p)Li^{8}$  cross section from the threshold for this reaction (16.89 Mev) up to 26

Mev. They observed the usual giant resonance and reported a peak cross section of 2.7 mb at 22.2 Mev, an integrated cross section of 13 Mev-mb up to 26 Mev, and a yield of  $2.3 \times 10^4$  counts/mole roentgen at a bremsstrahlung energy of 26 Mev. Several years later Cohen et al.<sup>2</sup> measured the photoproton yield from Be<sup>9</sup>

<sup>2</sup>L. Cohen, A. K. Mann, B. J. Patton, K. Reibel, W. E. Stephens, and E. J. Winhold, Phys. Rev. **104**, 108 (1956).

<sup>&</sup>lt;sup>†</sup>Contribution No. 1066. Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission. <sup>1</sup> R. N. H. Haslam, L. Katz, E. H. Crosby, R. G. Summers-Gill, and A. G. W. Cameron, Can. J. Phys. **31**, 210 (1953).