Mass Difference of K^{40} and Ar^{40}†

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(Received 24 August 1967)

The mass difference K^{40}-Ar^{40} and the electron-capture (EC) decay energy of K^{40} to the first excited state (2+) of Ar^{40} have been obtained from a measurement of the Q value of the exothermic reaction $K^{40}(n_{th},p)Ar^{40}$. The following reactions have been observed and their Q values determined: $K^{40}(n,p_0)Ar^{40}$ (2.270±0.005 MeV), $K^{40}(n,p_1)Ar^{40}$ (0.811±0.005 MeV), $K^{40}(n,\alpha)Cl^{37}$ (3.866±0.007 MeV), and, for comparison, $Cl^{35}(n,p)S^{35}$ (0.612±0.004 MeV). From the reaction $K^{40}(n,p_0)Ar^{40}$ the values 1.488±0.005 MeV and 27±5 keV, respectively, have been obtained for the K^{40}-Ar^{40} mass difference and the EC decay energy of K^{40}. The cross sections of the observed reactions were determined relative to the $N^{14}(n,p)C^{14}$ reaction cross section, yielding the values: $K^{40}(n,p_0)Ar^{40}$ (4.30±0.34 b), $K^{40}(n,p_1)Ar^{40}$ (0.127±0.016 b), $K^{40}(n,\alpha)Cl^{37}$ (0.388±0.032 b), and $Cl^{35}(n,p)S^{25}$ (0.446±0.040 b).

I. INTRODUCTION

THE orbital electron capture by K^{40} is of considerable geophysical importance. It has recently been pointed out[1] that, since orbital electron capture rates depend on the electronic environment, the branching ratio $K^{40} \rightarrow Ar^{40}$ must to some extent depend on the chemical state and pressure. Hence the chemical and pressure history of a sample may have to be taken into account in age determinations by means of the potassium-argon ratio. In order to estimate the importance of this effect, an exact knowledge of the disintegration energy of K^{40} to the 1.46-MeV 2+ state in Ar^{40} is necessary. Since a recent study[2] of the $Ar^{40}(p,n)K^{40}$ process led to a considerably lower electron capture energy (19±5 keV) than the previously accepted value[3–5] (49±6 keV), an independent determination of this energy by means of the exothermic $K^{40}(n_{th},p)Ar^{40}$ process seemed worthwhile, since the only previous published investigation[6] of this process did not achieve sufficient accuracy, yielding the value 47±100 keV. A new determination of this energy has therefore been performed.

In the course of this investigation not only has the proton branch (p_0) leading to the ground state of Ar^{40} been confirmed (see Fig. 1), but also a branch to the first excited state (n,p_1), and the reaction $K^{40}(n,\alpha)Cl^{37}$, have been observed. The Q value of the (n,α) reaction is known from the inverse $Cl^{37}(\alpha,n)K^{40}$ reaction[7] and from mass determinations.[8] Lastly, a measurement was performed to determine the thermal cross sections of the reactions observed relative to the $N^{14}(n,p)C^{14}$ reaction, whose thermal cross section is well known.[9]

II. APPARATUS AND EXPERIMENTAL PROCEDURE

The measurement of the charged particle reactions observed from thermal neutron capture in K^{40} has been performed using a well-collimated, thermalized beam from the Brookhaven National Laboratory graphite reactor. This neutron beam was incident on a 1 cm² aluminum foil of effective thickness 3.5 mg/cm² coated by evaporation with 40 μg/cm² of enriched[10] KCl. A surface-barrier gold-silicon detector of effective area 1 cm² capable of detecting protons of energy up to 3.0 MeV was mounted, in a vacuum chamber, 1 cm away from the target and outside the neutron beam. The pulses from the detector were fed into a low-noise charge-sensitive FET preamplifier, amplified and recorded on a 4096-channel pulse-height analyzer. A digital stabilizer in conjunction with a precision temperature controlled dual Decapot pulser[11] was employed to stabilize both the zero and gain of the analyzer.[12] The counter and source arrangement are shown in Fig. 2 together with a block diagram of the associated electronics.

† Work performed under the auspices of the U. S. Atomic Energy Commission.

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[1] M. Goldhaber and G. Scharff-Goldhaber (unpublished).

[2] J. Beyea, L. Mo, A. R. Sayres, and L. J. Lidofsky, Bull. Am. Phys. Soc. **10**, 610 (1965).

[3] *Nuclear Data Sheets*, compiled by K. Way *et al.* (U. S. Government Printing Office, National Academy of Sciences—National Research Council, Washington, D. C., 1966), NRC 59-4-42 K^{40}-2. Later values (Refs. 4 and 5) are in agreement with the mean value stated here.

[4] P. B. Parks, P. M. Beard, E. G. Bilpuch, and H. W. Newson, Nucl. Phys. **85**, 504 (1966).

[5] M. F. McCann, G. M. Lewis, and K. M. Smith, Nucl. Phys. **A98**, 577 (1967).

[6] J. Rossel and J. Weber, Helv. Phys. Acta **31**, 727 (1958).

[7] A. M. Smith and F. E. Steigert, Phys. Rev. **122**, 1527 (1961).

[8] J. H. E. Mattauch, W. Thiele, and A. H. Wapstra, Nucl. Phys. **67**, 32 (1965).

[9] J. R. Stehn, M. D. Goldberg, B. A. Magurno, and R. Wiener-Chasman, Brookhaven National Laboratory Report No. BNL 325, second edition, suppl. 2, 1966 (unpublished).

[10] K^{39} (63.3±0.2%), K^{40} (30.3±0.2%), K^{41} (6.49±0.2%); [measured impurities: Ba (0.01%), Ca (0.5%), Li (<0.01%), Na (0.5%)].

[11] G. L. Miller (unpublished).

[12] M. A. Poole (unpublished).

The mode of operation was as follows: Before and after each K^{40} run the system was calibrated using the natural α particles from Am^{241} and Cf^{252}. This procedure not only furnished a preliminary energy calibration but also served as a check on the stability of the system during the runs. At the end of a set of measurements a 20 $\mu g/cm^2$ U^{235} source[13] was employed to provide a subsequent energy calibration. At the conclusion of these sets of runs a known amount of air was introduced into the vacuum chamber in order to identify, by their energy loss in air, the nature of the charged particles observed. The target was then inverted so as to see if any of these reactions were due to impurities in the aluminum foil. None were detected. Lastly, in order to identify further the events arising from the K^{40} reactions, the KCl foil was replaced by a second foil, evaporated on an identical aluminum backing, whose enrichment[14] in K^{40} was an order of magnitude smaller.

In order to determine the cross sections of the above reactions relative to the $N^{14}(n,p)C^{14}$ reaction, a 60 $\mu g/cm^2$ KNO_3 target enriched[14] in K^{40} was prepared by evaporation on an aluminum foil identical to those used in the previous measurements and introduced in place of the KCl target. A series of runs were then made in which no energy calibrations were performed since only the relative number of counts in each peak was of interest.

III. EXPERIMENTAL DATA

A typical spectrum of the charged particles observed in one of the runs with the enriched KCl target is seen in Fig. 3. This figure shows the charged particle groups arising from the decay of the compound nucleus K^{41} to the ground states of Ar^{40} and Cl^{37}, plus a group at 0.595 MeV from the known reaction $Cl^{35}(n,p)S^{35}$.

Figure 4 represents the sum of a series of runs obtained with the highly enriched KCl target. As can be seen from this figure, besides the groups observed in the individual runs, two α-particle groups and a triton group at 1.431, 2.029, and 2.714 MeV, respectively, are observed, which do not belong to the reactions under study. Furthermore, an indication of the protons arising from the transition to the 2^+ state in Ar^{40} is seen above the background at 0.791 MeV. An analysis of these groups will be described in the following section.

The energy scale in the above figures has been obtained from the calibration performed, between the several sets of runs, with U^{235}, Am^{241}, and Cf^{252}. A typical spectrum, that of Cf^{252}, obtained during one of the calibration runs, is shown in Fig. 5. This figure shows the energy resolution for α particles of the detector employed, as well as a pulser peak showing the resolution of the electronic system.

Lastly, Fig. 6 shows the proton spectrum obtained with the KNO_3 target indicating, besides the transition

[13] U^{234} (0.06%), U^{235} (99.76%), U^{236} (0.06%), and U^{238} (0.09%).
[14] K^{39} (92.19±0.1%), K^{40} (1.90±0.05%), and K^{41} (5.92±0.1%). [Measured impurities: Na (0.07%), B (<0.01%)].

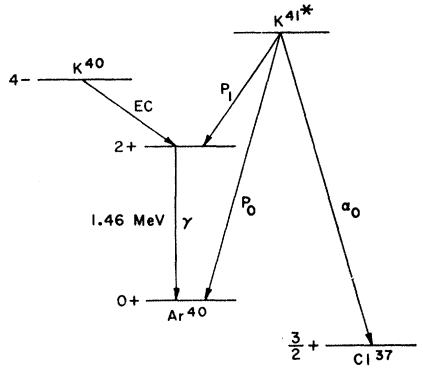

FIG. 1. Reactions produced by slow neutrons in K^{40} and decay branch of K^{40} by electron capture.

to the ground state of Ar^{40} arising from thermal neutron capture in K^{40}, the $N^{14}(n,p)C^{14}$ reaction which has been employed in the determination of the relative cross sections of the K^{40} reactions under study.

IV. ANALYSIS

The analysis of the data obtained in the present experiment consists of three parts: (A) calibration and linearity of the system, (B) the energetics of the charged particles observed from neutron capture in K^{40} and, (C) determination of the cross sections of these processes relative to the reaction $N^{14}(n,p)C^{14}$.

A. Calibration and Linearity of the System

The calibration of the system was performed, as mentioned previously, by means of an analysis of the natural α particles from U^{234}, U^{235}, Am^{241}, and Cf^{252}. This analysis is based on the assumption that the energy

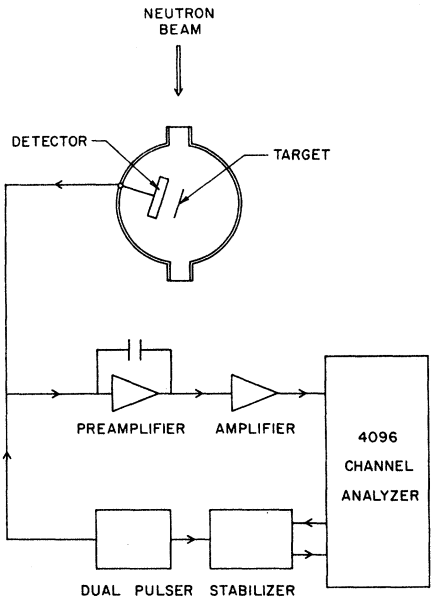

FIG. 2. Schematic view of experimental arrangement and electronics equipment.

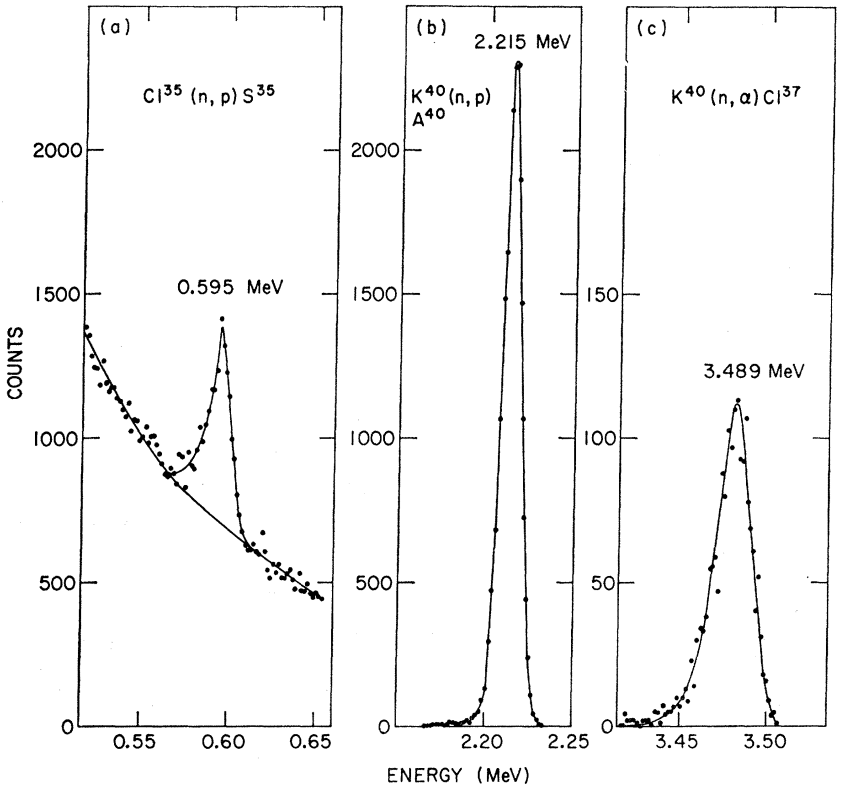

FIG. 3. Typical spectra of charged particles observed in a three-day run from a KCl target (enrichment of K^{40}: 30.3%) bombarded by thermal neutrons.

necessary to produce an electron-hole pair is identical for protons and α particles. The excellent resolution of the system for α particles, 18 keV full width at half-maximum (FWHM) (Fig. 5), permitted an unambiguous identification of the α-particle energy groups observed in U^{234}, Am^{241}, and Cf^{252}. In the case of U^{235} only the group at 4.210 MeV was employed for calibration, since the main group in U^{235} represents five unresolved lines.[15] The energy values used in this calibration were those of Lederer *et al.*[16] and are shown in Table I. These values have to be corrected for source thickness as well as for the finite thickness, 50 μg/cm^2, of the gold window of the detector employed.

TABLE I. α-particle energies used for calibration.[a]

Isotope	% abundance	Energy (MeV)
Cf^{252}	84.3	6.119±0.005
	15.5	6.076±0.005
Am^{241}	86	5.486±0.001
U^{234}	72	4.773±0.002
	28	4.722±0.002
U^{235}	5.7	4.210±0.005[b]

[a] Values taken from C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes*, (John Wiley & Sons, Inc., New York, 1967), 6th ed.

[b] This value was obtained by subtracting from the energy of the α branch populating the ground state of Th^{231}, 4.597±0.005 MeV, the energy of the level populated by this α branch (0.387 MeV). This energy is in good agreement with the energies listed in other tables, whereas it disagrees with the α-particle energy quoted in Ref. (a) (4.216 MeV).

The extreme thinness of the Am^{241} and Cf^{252} sources, the latter being made by vacuum implantation, introduces a negligible correction, while that of U^{234} and U^{235}, 20 μg/cm^2, introduces a 1-keV correction due to loss in the target.[17] The finite thickness of the detector window introduces corrections of approximately 2 keV to the Cf^{252} and Am^{241} α-particle energies and of approximately 3 keV to the U^{234} and U^{235} α energies.[17] These corrections have been introduced in the final energy calibration of the system. The errors in these corrections are negligible as they are well within the errors to which the α-particle energies are known.

The linearity of the system was measured using a precision mercury pulser[11] and a least-squares fit to the data obtained was performed. This showed that the system used was linear to better than 0.04% while the identification of each calibration point could be reproduced to within 0.8 keV (half a channel).

B. Charged Particles Observed from Thermal Neutron Capture in K^{40}

The analysis of the charged particle reactions observed from thermal neutron capture in K^{40} proceeded along the following lines: Every set of individual runs

[15] R. C. Pilger, F. S. Stephens, Jr., F. Asaro, and I. Perlman, Bull. Am. Phys. Soc. 2, 394 (1957); University of California Radiation Laboratory Report No. UCRL-3877 (unpublished).

[16] C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes* (John Wiley & Sons, Inc., New York, 1967), 6th ed.

[17] H. A. Bethe and J. Ashkin, in *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley & Sons, Inc., New York, 1953), vol. 1, p. 166.

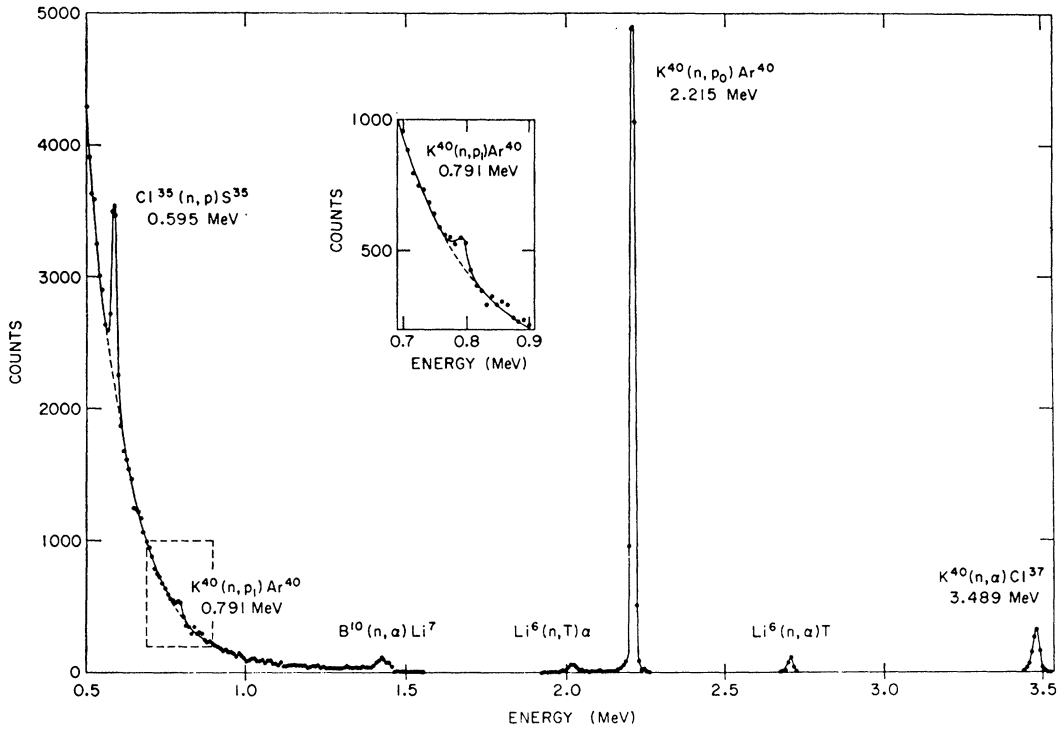

FIG. 4. Sum of four runs (representing ten days running time) with KCl target (enrichment of K^{40}: 30.3%). Insert shows proton group populating the 2+ state in Ar^{40}.

which, as explained previously, were preceded and followed by α-particle calibrations, was studied, and only those which showed no observable shift between the two calibration runs were retained. Each such set of runs was then individually analyzed.

A shape analysis was performed on each of the charged particle peaks observed to determine its centroid. This could be done in each case to better than 0.8 keV. (Fig. 3). Once the position of these peaks was determined a least-squares fit from the corresponding α-particle calibrations yielded the desired energy calibration for the charged particle reactions observed. These partial runs furnished, furthermore, a check on the reproducibility of the measurements. A weighted average was then performed among the individual runs. These values were then corrected for energy loss in the finite gold window of the detector employed.[18]

Lastly, in order to obtain a check on these final energy values, a determination was made of the energy of the transition to the first excited state of Ar^{40}. This was accomplished by adding all the individual runs and determining the centroid of the peak. Its energy was then obtained from a weighted average of each of the α-particle calibration runs employed and by taking into consideration the energy loss of this low-energy group in the detector window.[19]

The identification of the charged particle groups observed as belonging to the reactions under study was accomplished, as mentioned above, by first introducing a known amount of air into the chamber. This yielded a knowledge of the nature of the charged particles observed from their energy loss in air.[17] The enriched KCl target[10] was then substituted by one containing an order of magnitude less K^{40}.[14] This permitted a comparison, in the two targets, of the intensities of the charged particles from the reactions $K^{40}(n,p_0)Ar^{40}$ and $K^{40}(n,\alpha)Cl^{37}$ to that of the protons from the $Cl^{35}(n,p)S^{35}$ reaction. These procedures led therefore to an unequivocal identification of the reactions under study. They furthermore permitted us to establish the nature of the three weak charged particle groups (Fig. 4) as arising, respectively, from the alphas of the $B^{10}(n,\alpha)Li^{7}$ reaction, and the alphas and tritons of the $Li^{6}(n,\alpha)T$ reaction, probably due to impurities in the tape holding the target in place. The energy loss suffered by these groups is due to the finite thickness of the film covering this tape.

TABLE II. Q values obtained in the present experiment.

Reaction	Particle energy (MeV)	Q value (MeV)
$Cl^{35}(n,p)S^{35}$	0.595±0.004	0.612±0.004
$K^{40}(n,p_1)Ar^{40}$	0.791±0.005	0.811±0.005
$K^{40}(n,p_0)Ar^{40}$	2.215±0.005	2.270±0.005
$K^{40}(n,\alpha)Cl^{37}$	3.489±0.007	3.866±0.007

[18] R. M. Sternheimer, in *Nuclear Physics*, edited by L. C. L. Yuan and C-S. Wu (Academic Press Inc., New York, 1961), part A, p. 1.
[19] H. A. Wilcox, Phys. Rev. 74, 1743 (1948).

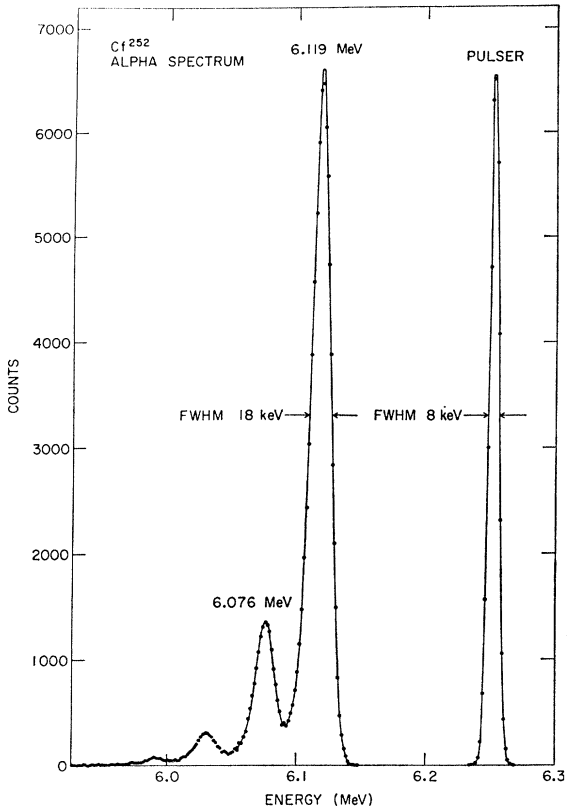

FIG. 5. α spectrum of Cf^{252} used for calibration and pulser peak.

C. Determination of Cross Sections Relative to that for the $N^{14}(n,p)C^{14}$ Reaction

The analysis of the charged particle reactions observed from thermal neutron capture in KNO_3 permits the determination of the cross section of the reaction $K^{40}(n,p_0)Ar^{40}$ relative to that of $N^{14}(n,p)C^{14}$.

In order to determine this value several sets of runs were added. The spectra thus obtained for the two reactions under consideration are shown in Fig. 6. As can be seen from this figure the total number of events in the $K^{40}(n,p)Ar^{40}$ reaction can be unambiguously determined. This, however, is not the case with the $N^{14}(n,p)C^{14}$ reaction as the number of events under the peak has to be determined in a region where the background is rapidly varying. The determination of this background [shaded area, Fig. 6(a)] therefore introduces the greatest source of uncertainty (8%) in the determination of the cross section.

Once the total number of counts under each peak was determined these numbers were normalized by taking the chemical and isotopic composition of the sample into account. The ratio of these normalized counts multiplied by the known[9] cross section of the $N^{14}(n,p)C^{14}$ reaction ($\sigma=1.81\pm0.05$ b) yielded the desired cross section for the $K^{40}(n,p_0)Ar^{40}$ reaction.

In order to obtain the cross sections of the reactions $K^{40}(n,p_1)A^{40}$, $K^{40}(n,\alpha)Cl^{37}$, and $Cl^{35}(n,p)S^{35}$, the data

TABLE III. Comparison between measured mass differences and EC decay energies.

Experimental method	K^{40}-Ar^{40}[a] (MeV)	E_{EC}[b] (keV)	References
$K^{40}(n,p)Ar^{40}$	1.51 ±0.10	47±100	[c]
	1.488±0.005	27±5	[d]
$Ar^{40}(p,n)K^{40}$	1.522±0.006	61±6	[e]
	1.505±0.001	44±1	[f]
	1.478±0.005	17±5	[g]
L/K ratio	1.481±0.003	20±3	[h]
	1.502±0.006	41±6	[i]
Mass spectr.	1.505±0.003	44±3	[j]

[a] The values in this column have been obtained in the following way: Those corresponding to the $K^{40}(n,p)Ar^{40}$ and $Ar^{40}(p,n)K^{40}$ reactions correspond to the observed Q value minus the (n,p) difference (Ref. 8) of 782 keV. The L/K ratio values arise from the reported E_{EC} to which has been added the γ-ray energy of 1.461 MeV (Ref. 20). The last value in this column is a direct measurement.
[b] All the values in this column, except the ones from the L/K ratio, have been obtained by subtracting the γ-ray energy of 1.461 MeV (Ref. 20) from the K^{40}-Ar^{40} value of the previous column.
[c] Reference 6.
[d] Present experiment.
[e] R. E. Holland and F. J. Lynch, Phys. Rev. 113, 903 (1959).
[f] Reference 4.
[g] Reference 2.
[h] B. L. Robinson and R. W. Fink, Rev. Mod. Phys. 32, 117 (1960).
[i] Reference 5.
[j] C. F. Giese and J. L. Benson, Phys. Rev. 110, 712 (1958).

from the 30.3% enriched KCl target were analyzed, as these data permit a comparison of these reactions relative to $K^{40}(n,p_0)A^{40}$ (Fig. 4). Here again the number of counts under the $K^{40}(n,p_0)Ar^{40}$ and $K^{40}(n,\alpha)Cl^{37}$ peaks can be uniquely determined while the peaks corresponding to the $K^{40}(n,p_1)Ar^{40}$ and $Cl^{35}(n,p)S^{35}$ reactions are found in a region of rapidly varying background. This background contributes the main error in the determination of the relative cross sections for the $K^{40}(n,p_1)Ar^{40}$ and $Cl^{35}(n,p)S^{35}$ reactions.

The number of counts under the four peaks of interest were then normalized, taking into account the

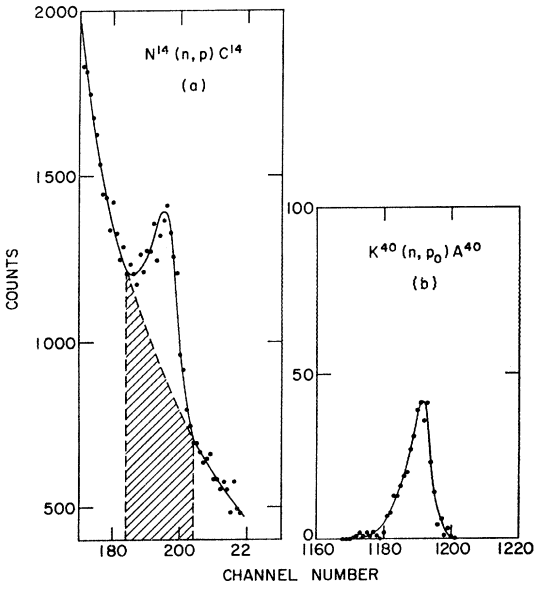

FIG. 6. Peaks from the reactions (a) $N^{14}(n,p)C^{14}$ and (b) $K^{40}(n,p)$-Ar^{40} used for cross-section determination.

chemical and isotopic composition of the sample. The ratio of the normalized counts $K^{40}(n,p_1)Ar^{40}/K^{40}(n,p_0)Ar^{40}$, $K^{40}(n,\alpha)Cl^{37}/K^{40}(n,p_0)Ar^{40}$, and $Cl^{35}(n,p)S^{35}/K^{40}(n,p_0)Ar^{40}$ multiplied by the previously obtained value for the $K^{40}(n,p_0)Ar^{40}$ reaction yielded the corresponding cross sections for these reactions.

V. RESULTS AND DISCUSSION

A. K^{40}-Ar^{40} Mass Difference

The results obtained, from the final energy calibration for the charged particle reactions observed in the present experiment, are given in Table II. This table shows the energies of the charged particles observed (column 2) plus the corresponding Q values for the reactions concerned (column 3).

The Q value for the reaction $K^{40}(n,p_0)Ar^{40}$ leads to a value of the mass difference $(K^{40}-Ar^{40})$, as $(K^{40}-Ar^{40})=Q-(n-H_1)$. Using the value[8] 782.45±0.07 keV for $(n-H_1)$, one finds $(K^{40}-Ar^{40})=1488\pm5$ keV. Furthermore, as K^{40} decays by electron capture to the first excited state (2+) of Ar^{40} (Fig. 1), which decays to the ground state by emission of a 1460.75±0.06 keV γ ray,[20] one obtains the value 27±5 keV for the energy available for the decay of K^{40} to Ar^{40} via electron capture. These values are in disagreement with some of the previously published higher-energy values as shown in Table III.

The reaction $K^{40}(n,\alpha)Cl^{37}$ has not been measured previously. The Q value reported here (see Table II) may be compared to the Q value for the inverse $Cl^{37}(\alpha,n)K^{40}$ reaction[7] (−3.86±0.06 MeV) and to the value found from adjusted Q values[8] (3.878±0.002 MeV). The difference between this last value and that found in the present experiment is 12±7 keV. If one compares the adjusted Q value[8] for the $K^{40}(n,p)Ar^{40}$ reaction (2.287±0.001 MeV) with the value given in Table II, one finds the value 17±5 keV.

On the other hand, the Q value obtained for the $Cl^{35}(n,p)S^{35}$ reaction (Table II) is in agreement with the value (615.0±0.2 keV) deduced from the S^{35} dis-

[20] J. D. King, N. Neff, and H. W. Taylor, Nucl. Instr. Methods **52**, 349 (1967).

TABLE IV. Thermal cross sections obtained in the present experiment.

Reaction	Cross section relative to $K^{40}(n,p_0)Ar^{40}$	Cross section (b)
$K^{40}(n,p_0)Ar^{40}$	1	4.30 ±0.34
$K^{40}(n,p_1)Ar^{40}$	0.0296±0.0030	0.127±0.016
$K^{40}(n,\alpha)Cl^{37}$	0.0902±0.0018	0.388±0.032
$Cl^{35}(n,p)S^{35}$	0.1038±0.0045	0.466±0.040

integration energy.[21] A further consistency, in the present results, is shown by the values obtained for the Q values of the reactions $K^{40}(n,p_0)Ar^{40}$ and $K^{40}(n,p_1)Ar^{40}$, as their difference yields the value 1459±7 keV in agreement with the γ-ray transition energy from the 2+ to the 0+ state in Ar^{40}.[20] Therefore, the discrepancies found in the reactions $K^{40}(n,p)Ar^{40}$ and $K^{40}(n,\alpha)Cl^{37}$ between the adjusted Q values and those measured in the present experiment have to be considered as real, and can be ascribed to a possible error in the determination of the mass of K^{40}.

B. Cross-Section Values

The analysis of the results obtained with the KNO_3 target yields a value for the cross section of the $K^{40}(n,p_0)Ar^{40}$ reaction of 4.30±0.34 b. This is based on the value for the $N^{14}(n,p)C^{14}$ reaction, $\sigma=1.81\pm0.05$ b. Our cross section is in good agreement with the value ($\sigma=3.8\pm0.7$ b) given by Rossel and Weber.[6]

The results of the relative measurements obtained with the KCL target are shown in the second column of Table IV. The values for the corresponding reaction cross sections, normalized to the $K^{40}(n,p_0)Ar^{40}$ reaction, are given in the third column of this table. The cross sections for the $K^{40}(n,p_1)Ar^{40}$ and $K^{40}(n,\alpha)Cl^{37}$ reactions have not been measured before. The present value obtained for the $Cl^{35}(n,p)S^{35}$ reaction is in good agreement with previously published results, whose weighted mean value[9] is $\sigma=400\pm100$ mb.

[21] R. D. Conner and I. L. Fairweather, Proc. Phys. Soc. (London) **70A**, 769 (1957); **70A**, 909 (1957).