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Photoproduction of calcium-47*

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Abstract

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PHOTOPRODUCTION OF CALCIUM-47*

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ABSTRACT

The irradiation of highly enriched $Ca^{48}CO_3$ with a bremsstrahlung beam of maximum energy 46 Mev produced Ca^{47} with a yield of 1.1 to 3.0 μc/mg Ca^{48} at half-saturation. The half-life of a sample measured through aluminum absorber to eliminate the contribution by the Sc^{47} daughter was 4.51 $\pm$ 0.02 days. By comparison with the $Cu^{63}(\gamma, n)Cu^{62}$ reaction the integrated cross section from 0 to 46 Mev for the sum of the reactions $Ca^{48}(\gamma, n)Ca^{47}$ and $Ca^{48}(\gamma, p)K^{47}$ was calculated to be 29 Mev-mbarn. The Ca^{46} content of the sample calculated to the end of the bombardment was ~0.04%, probably the result of the $Ca^{48}(\gamma, 3n)$ and $(\gamma, p2n)$ reactions.

INTRODUCTION

The production of radiochemically pure Ca^{47} for use as a tracer in medicine, agriculture and industry is of considerable interest.[(1)] Because of the presence at this Institute of a synchrotron capable of producing x-rays of 70 Mev energy, the possibility of producing Ca^{47} by a γ-n reaction on Ca^{48} was suggested by Oak Ridge National Laboratory. The use of isotopically pure Ca^{48} would lead to Ca^{47} essentially uncontaminated with Ca^{45}.

Several methods have been used for the production of Ca^{47}, including neutron irradiation of Ca^{46},[(2-5)] spallation reactions on copper, iron, vanadium and chromium,[(4, 6-8)] and other charged particle re-

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[1] Report of Second Consultant's Meeting on Calcium-47, IAEA, Vienna, December 1959 (unpublished).

[2] L. G. Cook and K. D. Shafer, Phys. Rev. 90, 1121 (1953).

[3] J. M. Cork, J. M. LeBlanc, M. K. Brice and W. H. Nester, Phys. Rev. 92, 367 (1953).

[4] L. J. Lidofsky and V. K. Fischer, Phys. Rev. 104, 759 (1956).

[5] W. S. Lyon and T. H. Handley, Phys. Rev. 100, 1280 (1955).

[6] R. E. Batzel, D. R. Miller and G. T. Seaborg, Phys. Rev. 84, 671 (1951).

[7] G. Rudstam, P. C. Stevenson and R. L. Folger, Phys. Rev. 87, 358 (1952).

[8] L. Marquez, Phys. Rev. 92, 1511 (1953).

actions.[5, 9] In most of these processes Ca^{45} is produced as an unwanted by-product. The only reported production of Ca^{47} by a γ-n reaction was in work done at the Royal Marsden Hospital, London.[1] In those experiments, calcium irradiated with electron-synchrotron bremsstrahlung of maximum energy 22 Mev, gave a yield of 0.1 microcuries of Ca^{47} per gram of Ca^{48} at infinite irradiation time.

EXPERIMENTAL

Oak Ridge National Laboratory provided on loan 125 mg of calcium carbonate with the following isotopic composition in atomic percent:

Ca^{40}	4.1
Ca^{42}	0.09
Ca^{43}	<0.05
Ca^{44}	0.2
Ca^{46}	<0.05
Ca^{48}	95.6 ± 0.1

Two irradiations of this material were performed with the Iowa State University electron-synchrotron.

First Irradiation

The enriched $CaCO_3$ was irradiated in a cylindrical holder of non-

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9 A. H. W. Aten, Jr., E. Greuell and W. J. van Dijk, Physica 19, 1049 (1953).

magnetic stainless steel. The holder was 3/8 in. in diameter and had a 1/8 in. hole drilled across the cylinder near the end. The $CaCO_3$ was packed in this hole which was then closed with two plugs of the same steel. The holder was inserted into a stainless steel probe which extended into the donut of the synchrotron. At the time of the irradiation the maximum energy of the synchrotron was 46 $\pm$ 2 Mev.

On March 17, 1960, the $Ca^{48}CO_3$ was dried for three hours at 105°C and 44.6 mg were loaded into the holder and irradiated for 68.8 hours.

After irradiation a portion of the material (37.2 mg) was transferred to a Plexiglas holder and counted in a Los Alamos-Sugarman Proportional Counter backed by a Radiation Counter Laboratory pulse amplifier and scaler. The window of the counter tube was 1.8 mg/cm^2 aluminized Mylar film and the counting gas was methane. The sample was counted with no added absorber and with 278 mg/cm^2 Al added absorber twice daily for 5 half-lives of Ca^{47}. A Sr^{90}-Y^{90} sample standardized in a 4π counter was mounted identically and counted during each period in order to normalize the calcium counting rates and to obtain an approximate geometry factor.

In order to obtain an approximate value for the cross-section, a separate irradiation under the same geometry was made on copper metal in the form of discs. These were counted in the same system and with the same geometry. After the irradiated calcium had decayed through 23 half-lives of Ca^{47} it was again counted in order to determine the yield of Ca^{45}.

Second Irradiation

A modification of the synchrotron designed to provide a more intense beam and cover a larger volume of the sample prompted the second irradiation. A different but similar holder was used to contain 33 mg of the material previously irradiated. The activity in this material had by this time (July 8, 1960) decayed to about 600 dpm. It was irradiated for four hours on July 9 and a portion of it was again transferred to a Plexiglas holder for counting. The counting system was similar to that used for the first irradiation.

CALCULATIONS

Since Ca^{47} decays to Sc^{47} which is also a beta emitter, the measurement of the yield of Ca^{47} in any nuclear process must take account of the relative counting efficiencies of the various beta groups. The decay scheme of Ca^{47} [(10)] shows a 1.94 Mev beta in 17% abundance and a 0.66 Mev beta (83%). In the decay of Sc^{47} there is a 0.44 Mev

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[10] D. Strominger, J. M. Hollander and G. T. Seaborg, Rev. Mod. Phys. 30, 585 (1958).

beta (60%) and a 0.60 Mev beta (40%). It was felt that a reasonable comparison with an absolute standard could be made with a $Sr-Y^{90}$ source which has beta rays of energy 0.55 and 2.3 Mev in equal amounts. On the basis of these energies, a correction factor for the absorption in the sample cover air and window was calculated. This factor, 1.57, represents the greater absorption of the Ca-Sc radiation relative to the Sr-Y radiation. Thus to obtain the yield, the total activity of the irradiated sample due to the decay of both Ca^{47} and Sc^{47} in transient equilibrium was compared with the decay rate of a $Sr-Y^{90}$ sample in secular equilibrium, the exact disintegration rate of which was known by 4π counting, and the absorption correction factor was applied.

First Irradiation

Measurements of the Ca^{47} decay with 278 mg/cm^2 of aluminum absorber between source and detector were used for the determination of the half-life of Ca^{47}. In this case the detection coefficient of Sc^{47} was considered to be negligible. Consideration of the various radiations from Sc^{47} and the efficiency of this counter for these irradiations would indicate that this assumption would not introduce an error of more than 0.5% in the half-life. Data taken on the decay of Ca^{47} over 5 half-lives (see Fig. 1) was treated by the method of least squares with a resultant value of 4.51 $\pm$ 0.01 days for the half-life. This error represents the precision of the data, a systematic error due to the presence of the Sc^{47} daughter could increase the

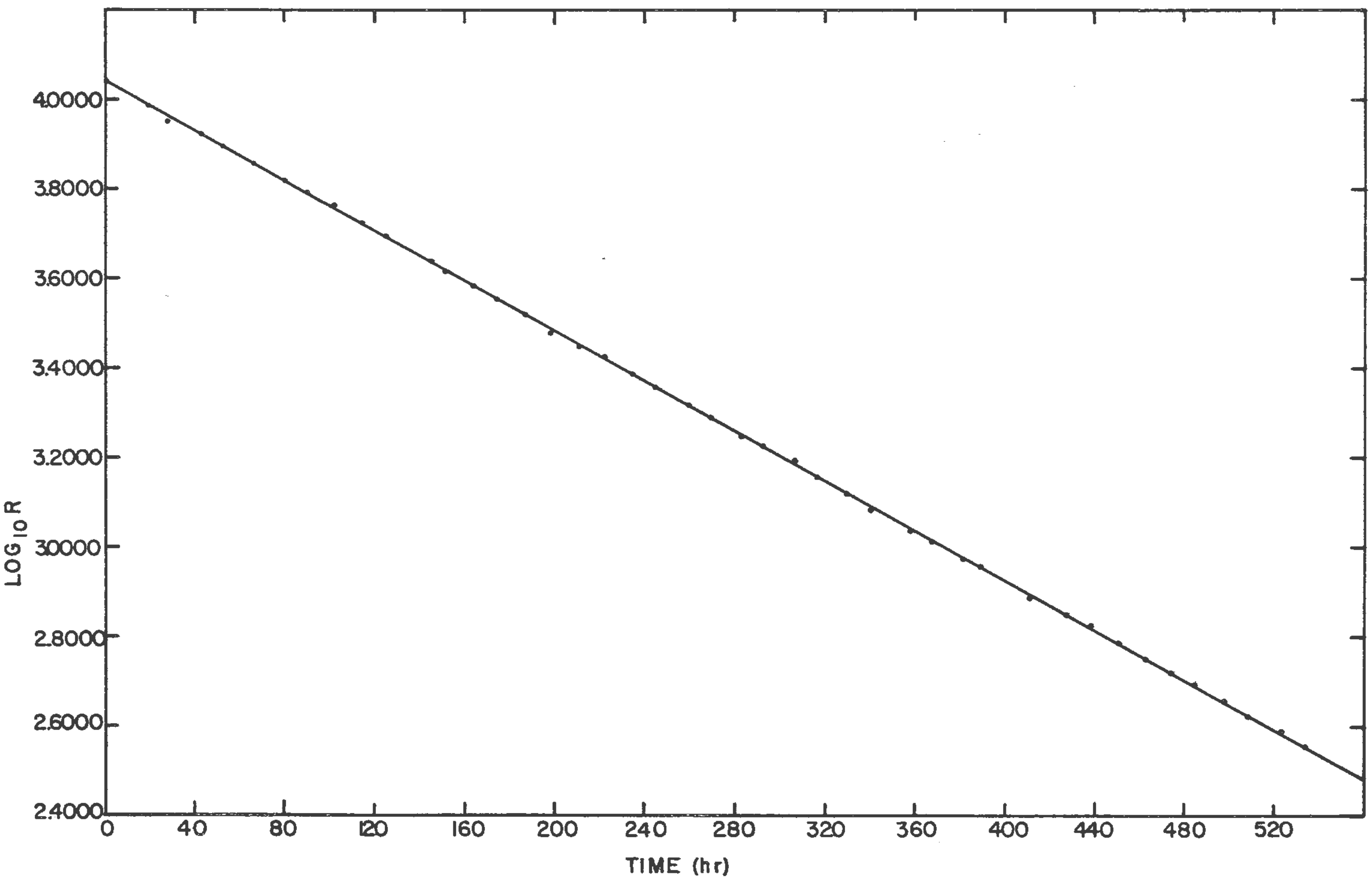

Fig. 1—Net counting rate of Ca^{47} through 278 mg/cm^2 of aluminum absorber. The solid line is the calculated least squares line.

over-all error to ± 0.02 days or roughly 0.5%.

The literature values for the half-life of Ca^{47} range from 4.3 to 5.3 days. Cook and Shafer[2] followed the decay of Ca^{47} produced by neutron irradiation of Ca^{46} by successively removing the Sc^{47} daughter and counting it. They report 4.8 ± 0.5 days for the Ca^{47} half-life. Lidofsky and Fischer[4] produced Ca^{47} in the same way and followed the decay with added absorbers for 5 half-lives, reporting a value of 4.7 ± 0.1 days. Lyon and Handley[5] produced Ca^{47} by bombarding CaO with 14 Mev protons and by irradiating enriched Ca^{46} with neutrons. They followed the decay with a well-type scintillation counter using 5 g/cm^2 of lead to cut out the Sc radiation and reported a half-life of 4.5 days. Other measurements include those by Marquez[8] of 4.3 ± 0.2 days, by Cork[3] of 5.35 ± 0.1 days and by Batzel[6] of 4.8 ± 012 days. The value reported here, 4.51 ± 0.02 days, thus compares favorably with earlier work but is possibly a little more accurate.

The calculation of the yield, which was made as indicated above by comparison with a standardized sample of $Sr\text{-}Y^{90}$, showed an average production rate during the irradiation of 7.63×10^7 atoms of Ca^{47} produced per minute. This corresponds to 12.3 μc produced in the measured sample in its 68-hr. irradiation or 35 μc for an infinite irradiation. On the basis of the Ca^{48} content of the measured sample, 15.82 mg, there were produced 0.77 μc Ca^{47}/mg Ca^{48} in 68 hrs. or 2.17 μc/mg at saturation.

These values are probably accurate to ± 10%.

The irradiation of copper discs was used to calculate the integrated cross-section for the production of Ca^{47} in the manner described by Penfold and Leiss[11] and Shupp, Colvin and Martin.[12] This involves a direct comparison of the yields of Cu^{62} and Ca^{47} and the use of the integrated cross-section of Cu^{62} which is well known. An NBS dosemeter was used to compare the integrated flux in the irradiations of copper and Ca^{48}. From these calculations an integrated cross-section for the production of Ca^{47} from Ca^{48} over the range 0 to 46 Mev was obtained of 29 ± 15 Mev-mbarn. It is noteworthy that this is much smaller than the cross-section for the $Cu^{63}(\gamma, n)Cu^{62}$ reaction which is 770 Mev-mbarn at this energy.[13] Also the observed cross-section for the production of Ca^{47} represents the sum of two processes, $Ca^{48}(\gamma, n)Ca^{47}$ and $Ca^{48}(\gamma, p)K^{47}$ although not listed in the isotope tables, must decay to Ca^{47} with a very short half-life.

The long-lived activity which remained in the sample 95 days after the irradiation was assumed to be Ca^{45} without any attempt at identifying it chemically or characterizing its radiation since the total activity was quite

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11 A. S. Penfold and J. E. Leiss, "Analysis of Photo Cross-Sections", Physics Research Laboratory, University of Illinois, May, 1958 (unpublished).

12 F. D. Schupp, C. B. Colvin and D. S. Martin, Jr., Phys. Rev. 113, 1095 (1959).

13 L. Marshall, A. H. Rosenfeld and S. C. Wright, Phys. Rev. 83, 305 (1951).

small. The measured activity due to Ca^{45}, extrapolated to the end of the irradiation, can be estimated as $5.74 \times 10^{-3}\%$ of the Ca^{47} activity. Corrections for the absorption of the lower energy beta rays from Ca^{45} bring the estimated Ca^{45} content up to approximately 0.04%.

In order to obtain an activity ratio of 4×10^{-4} for Ca^{45}/Ca^{47} in this irradiation, the ratio of production rates must have been about 1.2×10^{-2}. Since the isotopic ratio of Ca^{46}/Ca^{48} in the irradiated material is given as $< 5 \times 10^{-4}$, either the γ, n and γ, p cross-sections for Ca^{46} are much larger than those for Ca^{48} or the Ca^{45} is largely produced by the Ca^{48} (γ, 3n) or (γ, p2n) reactions.

Second Irradiation

The yield of Ca^{47} from the second irradiation was similarly determined with the result that 0.15 μc were produced per mg Ca^{48} in four hours. This corresponds to 5.9 μc/mg for irradiation to saturation, an increase in yield by a factor of 2.7 over the first irradiation.

CONCLUSIONS

From the results of the second experiment it can be seen that in a 4.5 day irradiation, a yield of about 3 μc of Ca^{47} per mg Ca^{48} could be obtained with negligible contamination by Ca^{45}. With the holders used in these irradiations, about 20 mg of Ca^{48} could be irradiated for a total yield of 60 μc in 4.5 days. It is estimated that this sample size could be increased by a factor of 3 to 150 mg of $CaCO_3$ or 64 mg Ca^{48}, with no loss in specific activity. If the Ca^{48} could be irradiated as metal, an additional improvement of a factor of 3 in the amount of Ca^{48} subjected to the beam could be obtained for a total yield of about 600 μc.

Certain changes in the synchrotron which are contemplated might increase the yield considerably. If the energy of the electron beam were raised to its design limit of 70 Mev, it is estimated that the yield would be increased by a factor of 4. A new injection system now under construction may increase the beam intensity by a factor of 6, for a total increase in specific activity of a factor of 24. Considering that all of these improvements would be possible, the over-all yield could be increased to approximately 14 mc in 200 mg of Ca^{48} or a specific activity of 70 mc/gm.

Thus it can be seen that if the production of Ca^{47} with a very low Ca^{45} content is highly desirable, the use of synchrotron produced x-rays to irradiate enriched Ca^{48} is a feasible method of accomplishing the task. It would appear that the high degree of enrichment in this material (95%) is not required. If the Ca^{46} content is less than 0.5% the Ca^{48} enrichment could be any value above 20% and material with very little Ca^{45} would result from the irradiation. This would, of course, reduce the total yield of Ca^{47} in an irradiation by the reduction in enrichment.

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