Photoproduction of calcium-47*

M. S. Foster  
_Iowa State University_

D. L. Weaver  
_Iowa State University_

A. F. Voigt  
_Iowa State University_

Follow this and additional works at: [http://lib.dr.iastate.edu/ameslab_isreports](http://lib.dr.iastate.edu/ameslab_isreports)

Part of the Chemistry Commons

Recommended Citation

[http://lib.dr.iastate.edu/ameslab_isreports/27](http://lib.dr.iastate.edu/ameslab_isreports/27)

This Report is brought to you for free and open access by the Ames Laboratory at Digital Repository @ Iowa State University. It has been accepted for inclusion in Ames Laboratory Technical Reports by an authorized administrator of Digital Repository @ Iowa State University. For more information, please contact digirep@iastate.edu.
PHOTOPRODUCTION OF CALCIUM-47*

by

M. S. Foster, D. L. Weaver
and A. F. Voigt

AMES LABORATORY
RESEARCH AND DEVELOPMENT REPORT
U.S.A.E.C.
UNCLASSIFIED

IS-184

Chemistry-Radiation and Radiochemistry (UC-7)
TID 4500, August 1, 1959

UNITED STATES ATOMIC ENERGY COMMISSION
Research and Development Report

PHOTOPRODUCTION OF CALCIUM-47*

by
M. S. Foster, D. L. Weaver
and A. F. Voigt

August 1960

Ames Laboratory
at
Iowa State University of Science and Technology
F. H. Spedding, Director
Contract W-7405 eng-82

UNCLASSIFIED
This report is distributed according to the category Chemistry-Radiation and Radiochemistry (UC-7) as listed in TID-4500, August 1, 1959.

Legal Notice

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty of representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Printed in USA. Price $0.50. Available from the

Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.
ABSTRACT ............................... 5
INTRODUCTION .......................... 6
EXPERIMENTAL ............................ 7
    First Irradiation .......................... 7
    Second Irradiation ....................... 9
CALCULATIONS ............................. 9
    First Irradiation ....................... 10
    Second Irradiation .................... 14
CONCLUSIONS ............................. 14
ACKNOWLEDGEMENT .......................... 16
PHOTOPRODUCTION OF CALCIUM-47 *

M. S. Foster, D. L. Weaver and A. F. Voigt

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 $\mu$ c/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 ± 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$^{45}$ 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\textsuperscript{47} for use as a tracer in medicine, agriculture and industry is of considerable interest.\textsuperscript{(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\textsuperscript{47} by a γ-n reaction on Ca\textsuperscript{48} was suggested by Oak Ridge National Laboratory. The use of isotopically pure Ca\textsuperscript{48} would lead to Ca\textsuperscript{47} essentially uncontaminated with Ca\textsuperscript{45}.

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

\textsuperscript{8} L. Marquez, Phys. Rev. \textbf{92}, 1511 (1953).
actions.\(^{(5, 9)}\) In most of these processes \(\text{Ca}^{45}\) is produced as an unwanted by-product. The only reported production of \(\text{Ca}^{47}\) by a \(\gamma\)-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 \(\text{Ca}^{47}\) per gram of \(\text{Ca}^{48}\) 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:

\[
\begin{align*}
\text{Ca}^{40} & : 4.1 \\
\text{Ca}^{42} & : 0.09 \\
\text{Ca}^{43} & : <0.05 \\
\text{Ca}^{44} & : 0.2 \\
\text{Ca}^{46} & : <0.05 \\
\text{Ca}^{48} & : 95.6 \pm 0.1
\end{align*}
\]

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

**First Irradiation**

The enriched \(\text{CaCO}_3\) was irradiated in a cylindrical holder of non-

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 CaCO₃ 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 ± 2 Mev.

On March 17, 1960, the Ca⁴⁸CO₃ 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 Flexiglas 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/cm² aluminized Mylar film and the counting gas was methane. The sample was counted with no added absorber and with 278 mg/cm² Al added absorber twice daily for 5 half-lives of Ca⁴⁷. A Sr⁹⁰-Y⁹⁰ 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

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 \(4\pi\) 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\textsuperscript{47} range from 4.3 to 5.3 days. Cook and Shafer\textsuperscript{(2)} followed the decay of Ca\textsuperscript{47} produced by neutron irradiation of Ca\textsuperscript{46} by successively removing the Sc\textsuperscript{47} daughter and counting it. They report 4.8 ± 0.5 days for the Ca\textsuperscript{47} half-life. Lidofsky and Fischer\textsuperscript{(4)} produced Ca\textsuperscript{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\textsuperscript{(5)} produced Ca\textsuperscript{47} by bombarding CaO with 14 Mev protons and by irradiating enriched Ca\textsuperscript{46} with neutrons. They followed the decay with a well-type scintillation counter using 5 g/cm\textsuperscript{2} of lead to cut out the Sc radiation and reported a half-life of 4.5 days. Other measurements include those by Marquez\textsuperscript{(8)} of 4.3 ± 0.2 days, by Cork\textsuperscript{(3)} of 5.35 ± 0.1 days and by Batzel\textsuperscript{(6)} of 4.8 ± 0.12 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-Y\textsuperscript{90}, showed an average production rate during the irradiation of 7.63 x 10\textsuperscript{7} atoms of Ca\textsuperscript{47} produced per minute. This corresponds to 12.3 \mu c produced in the measured sample in its 68-hr. irradiation or 35 \mu c for an infinite irradiation. On the basis of the Ca\textsuperscript{48} content of the measured sample, 15.82 mg, there were produced 0.77 \mu c Ca\textsuperscript{47}/mg Ca\textsuperscript{48} in 68 hrs. or 2.17 \mu c/mg at saturation.
These values are probably accurate to $\pm 10\%$.

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 $29 \pm 15$ 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

---


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

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

Second Irradiation

The yield of $^{47}\text{Ca}$ from the second irradiation was similarly determined with the result that 0.15 $\mu$C were produced per mg $^{48}\text{Ca}$ in four hours. This corresponds to 5.9 $\mu$C/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 $\mu$C of $^{47}\text{Ca}$ per mg $^{48}\text{Ca}$ could be obtained with negligible contamination by $^{45}\text{Ca}$. With the holders used in these irradiations, about 20 mg of $^{48}\text{Ca}$ could be irradiated for a total yield of 60 $\mu$C in 4.5 days. It is estimated that this sample size could
be increased by a factor of 3 to 150 mg of CaCO₃ or 64 mg Ca⁴⁸, with no loss in specific activity. If the Ca⁴⁸ could be irradiated as metal, an additional improvement of a factor of 3 in the amount of Ca⁴⁸ 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 Ca⁴⁸ or a specific activity of 70 mc/gm.

Thus it can be seen that if the production of Ca⁴⁷ with a very low Ca⁴⁵ content is highly desirable, the use of synchrotron produced x-rays to irradiate enriched Ca⁴⁸ 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 Ca⁴⁶ content is less than 0.5% the Ca⁴⁸ enrichment could be any value above 20% and material with very little Ca⁴⁵ would result from the irradiation. This would, of course, reduce the total yield of Ca⁴⁷ in an irradiation by the reduction in enrichment.
ACKNOWLEDGEMENT

The interest of Mr. P. S. Baker, Superintendent of the Isotope Sales Department, Oak Ridge National Laboratory in suggesting that this method of producing Ca$^{47}$ be tried is gratefully acknowledged. The authors also wish to express their gratitude to the synchrotron group, especially Dr. A. J. Bureau, for help with the irradiation.