

Cross Sections for the Production of Cl, K, Ca, and Sc Isotopes in the Bombardment of Pb and U with 3.0-Bev Protons*

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Cross sections for the formation of some nuclides in the mass number range 38–48 in the bombardment of Pb and U with 3.0-Bev protons have been measured. The maximum of the isobaric yield distributions for the nuclides from the Pb bombardment occurs at the stability line, while that from U is somewhat on the neutron-excess side. Total cross sections of 4–5 mb per mass number are deduced from the Pb data leading to a revision of the mass-yield curve in this region as proposed by Wolfgang *et al.* The cross sections from uranium are approximately twice as large as those from Pb. The data are shown to be consistent with a cascade-fission-evaporation mechanism.

INTRODUCTION

WOLFGANG *et al.*¹ have reported on the cross sections for the production of a variety of nuclides formed in the reaction of lead with protons of energies up to 3.0 Bev. The mass-yield curve obtained is markedly different at 3 Bev from that obtained at lower energies. In particular, at the lower energies (~ 300 –500 Mev) one finds a sharply peaked distribution with a maximum in the region $A=90$ –100.^{2,3} The results obtained at 3.0 Bev give no indication of a pronounced peak in this region but show a fairly flat distribution. At mass numbers below about $A=35$ the cross sections at 3.0 Bev were found to increase with decreasing A .

However, Wolfgang *et al.*¹ did not obtain data for mass numbers between 33 and 56. The shallow minimum in this mass region shown in Fig. 7 of their paper was obtained by interpolation. Therefore, it seemed worthwhile to measure cross sections for interactions of lead with 3-Bev protons leading to the formation of nuclides in this region. To obtain some indication of the variation of yield with target mass number, some of the nuclides isolated in the lead bombardments were also separated from uranium targets bombarded with 3.0-Bev protons. This was of interest because Caretto *et al.*⁴ had reported a pronounced rise in the cross sections for formation of Na²⁴ and F¹⁸ as the target mass number is increased (from Ta to U).

EXPERIMENTAL

Irradiations were performed in the circulating beam of the Brookhaven Cosmotron⁵ as described in reference 1. The reaction $\text{Al}^{27}(p,3pn)\text{Na}^{24}$ was used to monitor the proton beam. The cross section for this reaction was taken as 10.5 mb.⁶ Wolfgang *et al.* used an older value of 8.2 mb at 3 Bev. The thicknesses of the lead and uranium targets were ~ 90 and 50 mg/cm², respectively. In one irradiation a 325-mg/cm² sheet of uranium enriched to 94% U²³⁵ was used.

Both uranium and lead targets were dissolved in dilute HNO₃ containing the appropriate carriers. Whenever chlorine was to be isolated, AgCl was precipitated immediately. Otherwise, the first step was the removal of the target material by precipitation of PbCl₂ with gaseous HCl or by ether extraction of uranyl nitrate. Precipitation with NH₄OH yielded a fraction containing Sc, oxalate precipitation gave a Ca fraction. From the supernatant of either the CaC₂O₄ or AgCl precipitation, potassium was precipitated as potassium perchlorate. The further purification of the various fractions is described in the Appendix. After completion of the decay measurements, chemical yields in the separations were determined by suitable analytical methods.

The final precipitates were mounted on small filter paper disks⁷ and their activities were measured under end-window flow proportional counters of known geometrical efficiency. In addition, scintillation counter measurements were done, as described in the following paragraphs, to aid in the analysis of the complex isotopic mixtures obtained.

In the gamma spectra of the chlorine samples no 511-kev peak was detected, and an upper limit of a few percent could thus be set for the contribution of 32.4-min Cl³⁴ to the chlorine activity at end of bombardment. The chlorine decay curves obtained with the

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¹ Wolfgang, Baker, Caretto, Cumming, Friedlander, and Hudis, *Phys. Rev.* **103**, 394 (1956).

² W. F. Biller, University of California Radiation Laboratory Report UCRL-2067, 1952 (unpublished).

³ Vinogradov, Alimarin, Baranov, Lavrukina, Baranova, Pavlotskaya, Bragina, and Yakovlev, *Proceedings of the Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy*, July 1–5, 1955 (Akademiia Nauk, S.S.S.R., Moscow, 1955), pp. 97 and 132. (English translation by Consultants Bureau, New York; U. S. Atomic Energy Commission Report TR-2435, 1956, Part 2, pp. 65 and 85).

⁴ Caretto, Hudis, and Friedlander, *Phys. Rev.* **110**, 1130 (1958).

⁵ To complete this work during a long Cosmotron shutdown, one uranium irradiation was performed at the Berkeley Bevatron.

⁶ Cumming, Friedlander, and Swartz, *Phys. Rev.* **111**, 1386 (1958).

⁷ G. Friedlander and J. W. Kennedy, *Nuclear and Radiochemistry* (John Wiley and Sons, Inc., New York, 1955), p. 280.

proportional counters were therefore analyzed into two components, 37.3-min Cl^{38} and 55.5-min Cl^{39} . The method of analysis, described, e.g., by Wahl,⁸ makes use of the relation

$$A_t = A_1^0 e^{-\lambda_1 t} + A_2^0 e^{-\lambda_2 t},$$

where A_t is the total activity at time t , A_1^0 and A_2^0 are the activities of the two components at $t=0$, and λ_1 and λ_2 are the two decay constants. Then

$$A_t e^{\lambda_2 t} = A_2^0 + A_1^0 e^{(\lambda_2 - \lambda_1)t}.$$

Therefore, with accurately known values of λ_1 and λ_2 , one can plot $A_t e^{\lambda_2 t}$ vs $e^{(\lambda_2 - \lambda_1)t}$ and obtain a straight line with slope A_1^0 and intercept A_2^0 .

The potassium fractions obtained from lead were found to be radiochemically pure ($\leq 0.1\%$ of other activities). After subtraction of a small, nondecaying component accounted for by the natural K^{40} present, the decay curves could be analyzed by the method described above into components with half-lives of 22.4-hr (K^{43}) and 12.5-hr (K^{42}). The potassium fractions obtained from uranium contained, in addition, an impurity of approximately 35-day half-life, present to the extent of $\sim 1\%$ of total potassium activity at end of bombardment.

The calcium decay curves could be analyzed into two components ascribable to 4.7-day Ca^{47} and 152-day Ca^{46} , although the activity of the longer-lived component was so low (< 10 counts/min) that the presence of other long-lived impurities could not be positively ruled out, and rather large uncertainties had to be assigned to the Ca^{46} results. The growth of 3.4-day Sc^{47} was taken into account in the analysis.

Analysis of scandium decay data presents some difficulties. The long-lived component was pure 85-day Sc^{46} and this was subtracted from the total β -decay curve. Resolution of the residual curve was effected in the following way. The disintegration rate of the 2.4-day Sc^{44m} (in equilibrium with the 4-hr β^+ -emitter Sc^{44}) was followed by repeated comparison of the annihilation radiation intensity from the Sc sample with that from

TABLE II. Results obtained with U targets.

Product nuclide	Type of yield	Cross section (mb) Normal U	Cross section (mb) U^{235}	Estimated error ($\pm\%$)
K^{42}	<i>I</i>	4.0	3.4	20
K^{43}	<i>C</i>	4.1	3.9	20
Ca^{46}	<i>C</i>	3.7	3.7	50
Ca^{47}	<i>C</i>	1.2	0.5	75
Sc^{44m}	<i>I</i>	...	1.7	20
Sc^{46}	<i>I</i>	2.5	3.2	20
Sc^{47}	<i>I</i>	3.5	3.6	40
Sc^{48}	<i>I</i>	4.8	4.1	40

a calibrated Co^{56} source. For these measurements the sources, sandwiched between sufficient aluminum absorbers to ensure complete annihilation of positrons (Co^{56} and Sc^{44} have similar β^+ end points), were placed near a 2 in. \times 2 in. NaI(Tl) scintillation crystal connected to a 100-channel analyzer. The disintegration rates of Sc^{44m} so determined were reduced to the appropriate counting rates in the proportional counter, and this 2.4-day component was subtracted. The remaining curve could be analyzed into 3.43-day Sc^{47} and 44-hour Sc^{48} . One sample was isolated sufficiently early to permit approximate determination of a 3.9-hour component, presumably a mixture of Sc^{43} and Sc^{44} .

The disintegration rate for infinitely long bombardment was determined for each of the observed products from the measured counting rate and chemical yield, and from the known counting efficiency and length of irradiation. These disintegration rates were then translated into formation cross sections by comparison with the Na^{24} activity induced in the Al monitor foils.

RESULTS AND DISCUSSION

The results are listed in Tables I and II. The errors indicated are estimated probable errors based on considerations of the radiochemical purity of the samples, chemical yield determinations, decay curve analyses, and counting efficiency calibrations.

Of the cross sections measured, those of K^{42} , Sc^{44m} , Sc^{46} , Sc^{47} , and Sc^{48} represent yields for independent formation of these nuclides because their β -decay parents are stable or long-lived. In the case of Cl^{38} , the bombardment and separation time was comparable to the half-life of the β -decay parent (2.8-hour S^{38}). All other yields are cumulative yields for the mass chains involved. Examination of the data, in particular a comparison of the Sc^{47} and Ca^{47} yields, indicates that the peak yields for any isobaric chain occur near the stability valley. Thus one is justified in considering chain yields along with the independent yields, since the feed-in from β -decay precursors appears to contribute little to any measured chain yield.

It thus seemed instructive to plot cross sections vs distance from the stability line ($Z - Z_A$). If the mass-yield curve in the narrow mass region investigated is flat, such a plot should show a single distribution. The cross-section data for Pb bombardments are so plotted

TABLE I. Results obtained with Pb targets.

Product nuclide	Type of yield ^a	Cross section (mb)	Average	Estimated error ($\pm\%$)
Cl^{38}	<i>C-I</i>	1.26, 1.41	1.33	20
Cl^{39}	<i>C</i>	0.78, 0.85	0.81	20
K^{42}	<i>I</i>	1.58, 2.29, 2.16, 1.59, 1.49	1.82	20
K^{43}	<i>C</i>	1.39, 2.32, 2.02, 1.94, 2.33	2.00	20
Ca^{46}	<i>C</i>	1.2, 1.6	1.4	25
Ca^{47}	<i>C</i>	0.33, 0.33	0.33	40
Sc^{42+44}		0.6	0.6	50
Sc^{44m}	<i>I</i>	1.89, 1.28	1.58	20
Sc^{46}	<i>I</i>	2.51, 2.01	2.26	20
Sc^{47}	<i>I</i>	2.4, 1.1	1.7	40
Sc^{48}	<i>I</i>	0.7, 1.5	1.1	40

^a *C* = cumulative yield; *I* = independent yield; *C-I* indicates an intermediate situation.

⁸ A. C. Wahl, Phys. Rev. **99**, 730 (1955).

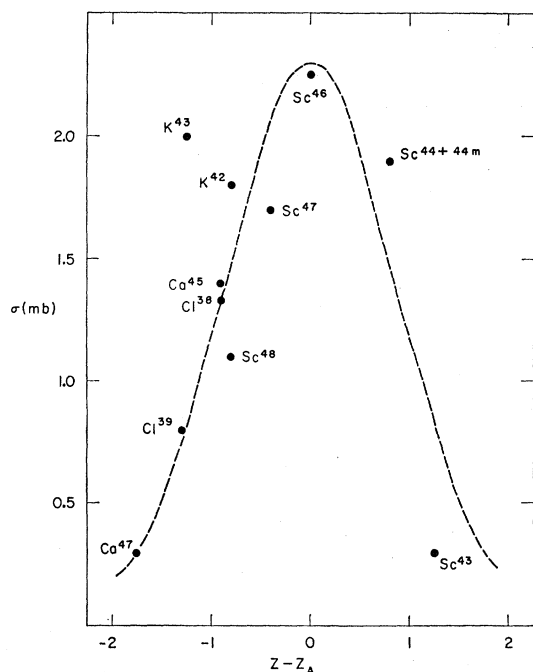


FIG. 1. Cross sections of nuclides formed in lead bombardments plotted against $(Z-Z_A)$. Z_A values follow those of Coryell.⁹ The observed yield of 4-hour Sc was arbitrarily divided equally between Sc^{43} and Sc^{44} . A Gaussian distribution with a peak value of 2.3 mb at $Z=Z_A$ is drawn for illustrative purposes.

in Fig. 1. It is seen that the data do indeed fall fairly well on a single distribution curve which happens to show a maximum at $Z=Z_A$. The Z_A values of Coryell⁹ were used here, but other Z_A choices were found to lead to curves of essentially the same shape. It should be noted that the right-hand (neutron-deficient) side of the curve is poorly defined, particularly since the distribution of the measured yield for 4-hr Sc between Sc^{43} and Sc^{44} is not known. The upper limit of ~ 0.2 mb for Cl^{34} ($Z-Z_A=0.95$) would indicate that the distribution curve falls off more steeply on the neutron-deficient than on the neutron-excess side.

The yields from uranium are in general higher than those from lead, for some nuclides by factors of 2 to 4. In view of the experimental errors, no significance can be attached at present to the apparent differences between U^{235} and normal uranium. The yield pattern for uranium appears to be markedly different from that for lead. The scandium cross sections increase monotonically with increasing A from Sc^{44} to Sc^{48} . The distribution in terms of $Z-Z_A$ is certainly broader than in the case of lead and appears to have its peak in the neighborhood of $Z-Z_A=-1$.

Using the "universal" isobar distribution of Fig. 1 one can now obtain the total yield at each mass number formed in the Pb interactions. This turns out to be

5.0 ± 0.1 mb at each mass number in the region $38 \leq A \leq 48$ if one assumes the Gaussian distribution of Fig. 1; if a steeper slope is taken on the neutron-deficient side in accordance with the argument above, the cross section per mass number reduces to about 4 mb. Wolfgang *et al.*¹ had predicted a rather flat valley at about 2 mb in this region of the mass-yield curve. The present work indicates clearly that this valley must be raised to 4–5 mb. A similar analysis of the data of Wolfgang *et al.*¹ in the mass region $56 \leq A \leq 72$ also leads to values in the neighborhood of 4 mb per mass number. Thus it appears that the entire valley in the 3-Bev mass-yield curve shown in Fig. 7 of reference 1 should be raised by about a factor of two.

Some conclusions may be drawn regarding the mechanism by which the nuclides investigated here arise in the interaction of lead and uranium with high-energy protons. Two extreme mechanisms may be considered, (a) that the observed products are formed as the primary fragments in some fission or fragmentation act or (b) that they are the residues of relatively long evaporation chains following fission or fragmentation. The first possibility can be ruled out quite readily. For a nucleus such as Sc^{46} to be formed as a primary fission fragment from Pb interactions the fissioning nucleus would presumably have to have approximately the same neutron-to-proton ratio (1.19) as Sc^{46} and a Z^2/A value sufficiently high for fission to occur; in the region of Pt to Pb this would require the pre-fission evaporation of 20 to 25 neutrons without any accompanying charged-particle evaporation or of at least 10 times as many neutrons as protons. According to the calculations of evaporation systematics by Dostrovsky *et al.*¹⁰ this is an entirely unrealistic postulate. The other extreme possibility would be the breakup of a highly excited nucleus resulting from the prompt cascade, followed by evaporation. According to the prompt-cascade calculations of Metropolis *et al.*,¹¹ the most probably cascade product from the interactions of 1.8-Bev protons with Pb^{206} is Au^{197} ; at 3-Bev bombarding energy it might be Pt^{195} . If this nucleus broke up into fragments of the same n/p ratio, the products would include such nuclei as K^{48} , Ca^{50} , Sc^{53} which could then, by evaporation of 7 or 8 neutrons, or of 8 or 9 neutrons plus one proton, reach the stability valley. According to the evaporation calculations of Dostrovsky *et al.*¹⁰ this would require excitations of about 90 to 120 Mev in the primary fragments, and at these initial excitations a ratio of $\sim 10:1$ for neutron-to-proton evaporation is expected in this region of A . Assuming that in the fission act the excitation energy is split in proportion to the fragment masses and without appreciable conversion of excitation to kinetic energy, one finally arrives at excita-

¹⁰ Dostrovsky, Rabinowitz, and Bivins, *Phys. Rev.* **111**, 1659 (1958).

¹¹ Metropolis, Bivins, Storm, Miller, Friedlander, and Turkevich, *Phys. Rev.* **110**, 204 (1958).

⁹ C. D. Coryell, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Palo Alto, 1953), Vol. 2, p. 305.

tions in the vicinity of 400 Mev for the fissioning nuclei. This again is in accord with the results of the prompt-cascade calculations¹¹ for the mean excitation in the average cascade product. Thus the Pb results can be accounted for by a consistent picture in terms of a cascade-fission-evaporation mechanism. It should be noted, however, that a certain amount of pre-fission evaporation cannot be excluded. Fissioning nuclei at higher excitations than indicated above may also contribute to the observed yields, since evaporation will proceed more or less along the stability valley once this has been reached.

The fact that the isobaric yield distributions from uranium have their peaks slightly more towards the neutron-excess side than those from lead gives a clue to the relative excitation energies involved in the two cases. On the average, the primary breakup products from uranium evidently do not have quite enough excitation to evaporate neutrons all the way to the line of β stability. Thus the fissioning nuclei apparently require less excitation energy for breakup into the fragments of interest and this fact, as well as the magnitude of the cross sections would indicate that the greater fissionability of the heaviest nuclei is discernible even in the somewhat exotic high-energy processes which are under discussion here.

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It is with particular pleasure that one of us (L. Y.) wishes to record his appreciation to the Brookhaven Chemistry Department for placing their equipment at

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APPENDIX. CHEMICAL PURIFICATION PROCEDURES

Chlorine.—The AgCl was dissolved in conc. NH_4OH , the solution made acid, and repeatedly scavenged with $\text{Fe}(\text{OH})_3$ precipitations. Bromide and iodide carriers were added and silver bromide and iodide repeatedly precipitated in the presence of 1M NH_4OH . AgCl was then precipitated from a Versene solution, scavenged with $\text{Fe}(\text{OH})_3$ and the process repeated several times. AgBr and AgI were again precipitated from NH_4OH , and AgCl was finally precipitated and mounted for measurement.

Calcium.—Calcium oxalate was dissolved, the oxalate destroyed, CaCO_3 was precipitated and redissolved, and repeated scavenging precipitations of $\text{Fe}(\text{OH})_3$, of PdS and CuS in acid solution, and of NiS and CoS in NH_4OH were performed. The oxalate was reprecipitated, dissolved, and scavenged with $\text{Ba}(\text{NO}_3)_2$ and $\text{Sr}(\text{NO}_3)_2$ precipitations. The oxalate and carbonate were reprecipitated, the carbonate dissolved and scavenged with $\text{La}(\text{OH})_3$. The calcium was finally precipitated as the oxalate.

Scandium.—The procedure used was similar to one used at Los Alamos.¹² In addition, to achieve proper decontamination it was found necessary to scavenge with CuS precipitations and then extract with TTA. The Sc was finally precipitated as K_3ScF_6 .

Potassium.—The potassium was repeatedly precipitated as KClO_4 , then separated as the cobaltinitrite, and scavenged repeatedly by FeS and $\text{La}(\text{OH})_3$ precipitations and BaCO_3 and CaCO_3 precipitations. After removal of NH_4^+ salts by fuming, KClO_4 was reprecipitated. Rb and Cs were repeatedly removed as the chlorostannates, SnO_2 removed from the K solution, and the procedure repeated. KClO_4 was finally precipitated in the presence of Na carrier.

¹² J. Kleinberg (Editor), Atomic Energy Commission Report LA-1721 (rev.), 1954 (unpublished).