CROSS-SECTIONS FOR THE FORMATION OF Sb AND Sc ISOTOPES
BY IRRADIATION OF Y, La, Ta, AND Au WITH 18.2 GeV PROTONS

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ABSTRACT

The cross-sections have been measured for several Sb and Sc isotopes produced by irradiation of yttrium (Sc only), lanthanum, tantalum, and gold with 18.2 GeV protons. The shapes of the yield distribution curves for both Sb and Sc isotopes vary with the target element. If the previously known yield distributions with uranium as targets are decomposed into a "spallation" part and a fission part, the former will in both cases be about equal to the distribution with gold or tantalum as the target, whilst in shape the fission part closely resembles the distributions obtained when uranium is irradiated with 570-590 MeV protons. From the variation in shape of the distribution curves with target, this procedure of decomposition seems less justified for the scandium isotopes than for antimony isotopes.

The high spin to low spin isomeric yield ratios of the ^{44}Sc isomers were also determined, and show a very definite increase with mass difference between target and product.

Geneva - 21 June 1968

(Submitted to J.Inorg. and Nucl.Chem.)

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THE JOURNAL OF THE AMERICAN MEDICAL ASSOCIATION PUBLISHED WEEKLY

535 N. Dearborn Ave., Chicago 10, Ill.
Subscription price, \$5.00 per annum in advance.
Single copies, 15 cents.

Entered as Second-Class Matter, May 26, 1917.
Postpaid.

Acceptance for mailing at special rate of postage provided for in Act of October 3, 1917, authorized on July 26, 1934.
Postage paid at Chicago, Ill., and at additional mailing offices.
Postmaster: Send address changes in this journal to The Journal of the American Medical Association, 535 N. Dearborn Ave., Chicago 10, Ill.
This journal is published weekly, except during the months of December and January, when it is published bi-weekly.
The subscription price is \$5.00 per annum in advance.
Single copies, 15 cents.
The Journal of the American Medical Association is published by the American Medical Association, 535 N. Dearborn Ave., Chicago 10, Ill.
The Journal of the American Medical Association is published by the American Medical Association, 535 N. Dearborn Ave., Chicago 10, Ill.

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Printed at the Chicago Press, Chicago, Ill.

Published by the American Medical Association

535 N. Dearborn Ave., Chicago 10, Ill.

Subscription price, \$5.00 per annum in advance.
Single copies, 15 cents.

1. INTRODUCTION

This work is a direct continuation of work done by the present authors^{1,2)} and is also partly based upon the results of same. Ravn²⁾ studied the yields and ranges of scandium isotopes at 590 MeV and 18.2 GeV proton irradiation of uranium, and Hageb⁴⁾ studied yield distributions of antimony isotopes when uranium is bombarded with 159 MeV to 18.2 GeV protons. We extend here the yield studies at 18.2 GeV proton energy to include a number of lighter targets.

The question of mechanism for reactions taking place when a heavy nucleus is bombarded with very high energy protons is still not fully understood. It seems obvious that in such a bombardment there will be a certain possibility for almost any deposition energy, from the low-energy deposition of simple inelastic scattering reactions or peripheral reaction types, to the close to complete energy deposition leading to the most complex reactions. One can probably expect that any defined deposition energy interval will lead to a certain class of products.

Therefore, since the high-energy proton bombardments give a mixture of all possible reactions, it is necessary to amass a considerable amount of information in order to unravel the puzzle which is produced. When uranium is bombarded with protons of increasing energy, we know well the development of the product distributions up to 170 MeV³⁾, and many further details all up to about 600 MeV^{1,2,4-6)}. The yield distributions and momentum properties of the products are those typical for fission. We exclude here a small part of the total cross-section due to spallation products close to the target.

As the energy is increased into the GeV region, the mass-yield distribution in the fission product region broadens markedly, and products having properties other than the hitherto known fission products are produced together with the latter^{1,2,5-9)}. Fortunately, at least some of the new species have neutron-to-proton ratios different from those of the fission products, so that the properties of both may be compared directly.

Such comparisons are the primary reason why mechanisms other than that of fission have been proposed. In particular, the recoil ranges of these new products in uranium and other materials are shorter than the ranges of fission fragments^{6,7)}.

Rudstam and Sørensen⁵⁾ compared the yield distributions of iodine isotopes, and found that the lightest part of the distribution from uranium bombardment is almost identical to the distribution from gold or tantalum bombardment. From this they concluded that from uranium the light isotopes of iodine are spallation products, since gold and tantalum should not give fission products in the iodine region.

However, without mapping the product cross-section distributions in greater detail, almost any model will fit; thus the aim of the present work is to obtain additional relevant information about these extremely complicated processes.

2. EXPERIMENTAL TECHNIQUE

2.1 Targets and irradiations

The targets consisted of foils of uranium, gold, tantalum, lanthanum, and yttrium with thicknesses of about 90, 45, 47, 30, and 14 mg/cm², respectively. Several runs were made, always with uranium as one of the components, and with two or three other, different foils in each run. The target foils were separated by two 0.03 mm aluminium foils, in such a way that each target foil was covered by aluminium on both sides so as to catch the recoils. These catcher foils were then chemically treated, together with the target foils, to avoid any recoil losses. No aluminium monitor foils were used, as the monitor in this experiment is the uranium foil. The cross-sections were determined by measuring nuclide by nuclide relative to the ones produced in uranium, the latter being known from earlier work^{1,2)}.

To avoid edge effects coming from possible misalignment of the different foils in the stack, a circular part of the whole target, just back from the leading edge, was punched out after irradiation. The possibility of more secondary reactions taking place away from the leading edge of the target⁶⁾ was ignored, as the only reactions studied in the present work are reactions that need a very high energy deposition in the target nucleus and which cannot be induced by low-energy secondary particles.

The irradiations were performed in the circulating beam of the CERN Proton Synchrotron (PS) at an energy of 18.2 GeV.

2.2 Chemical separation methods

From some of the target stacks both Sc and Sb were separated from each target (only Sc from the Y target), whilst only Sb was separated from others. The dissolution and the very first steps of the chemical procedure were different for the different target elements with their catchers.

Lanthanum was dissolved in concentrated HCl containing 1 mg each of Sc, As, and Sb carriers. The solution was diluted to 10 ml with water, and the sulfides of arsenic and antimony precipitated by leading H_2S gas into the solution. After centrifuging, the sulfide was further purified as described below and the supernate made basic with NH_3 to precipitate $Sc(OH)_3$.

Tantalum was dissolved in a Pt crucible with a mixture of concentrated hydrofluoric, nitric, and hydrochloric acids containing the same carriers. After dilution and precipitation of the sulfides, the supernate was returned to the same crucible, 1 ml of concentrated sulfuric acid was added, and the solution evaporated to strong white fumes. It was then diluted and $Sc(OH)_3$ precipitated with NH_3 .

Gold was dissolved in about 10 ml aqua regia containing the same carriers. The solution was then made basic with 10 N NaOH solutions. The precipitate of $Sc(OH)_3$ was centrifuged off for further purification, and gold precipitated by adding about 1 ml 10% H_2O_2 . The new supernate was then made about 0.1 N in HCl and the arsenic and antimony sulfides precipitated.

Uranium was dissolved in dilute HCl containing some HNO_3 and carriers. The sulfides were precipitated by dilution and addition of H_2S gas. $Sc(OH)_3$ was precipitated from the supernate after boiling it down to a small volume and addition of 1M NaOH.

Yttrium was dissolved in concentrated HCl containing only Sc carrier. The solution was then made basic with NH_3 to precipitate the hydroxides.

The $Sc(OH)_3$ precipitates were further purified as described in detail by Ravn²⁾. The method consists of $Sc(OH)_3$ precipitation from a 1N NaOH solution containing some H_2O_2 and extraction from 0.1N HCl into 0.4M TTA in benzene. After back-extraction with 2N HCl, $Sc(OH)_3$ is again precipitated and washed. Counting was performed on a liquid sample obtained by dissolving this in 2N HCl.

The Sb and As sulfides were washed in H_2O , dissolved in 1 ml concentrated H_2SO_4 , and taken to thick white fumes. The solution was made ~30% in H_2SO_4 by dilution with water and poured onto granulated Zn held

at 100°C in an apparatus such as the one described by Greendale and Love¹⁰). The stibine (SbH_3) and arsine (AsH_3) produced were led in a quartz tube through an oven kept at 600°C, just outside of which antimony metal was deposited. A quartz wool plug ensured complete collection of the elemental antimony. The arsine was further decomposed by a burner placed after the plug.

In order to separate the antimony completely from the arsenic, the Sb deposit was dissolved in 9N HCl containing Br_2 and extracted into 10% isopropylether in benzene, washed three times with 9 N HCl and stripped from the organic phase with 3 ml H_2O , which were then transferred into small bottles for counting.

Care was taken that the SbH_3 production was always done simultaneously at a known time for all targets to facilitate corrections for growth from parent activities ($^{117}\text{Te} \rightarrow ^{117}\text{Sb}$).

Chemical yields were not in general determined absolutely, but only relative to the yield for Sb or Sc from U. For Sc the spectrophotometric method of Berman et al.¹¹) was used, which is a method based on the specific absorption of the Sc-xylene orange complex at 5530 Å in dilute perchloric acid. The chemical yield of Sb was determined by the spectrophotometric method of Van Aman et al.¹²) which uses the absorption peak at 5500 Å of the dithizone complex of antimony in isopropylether.

Two later runs were made in a completely different way to determine only the yield of $^{120\text{m}}\text{Sb}$. The targets (U, Au, Ta, and La) were dissolved very slowly and carefully to be certain that no loss of Sb in the form of very volatile SbCl_5 took place. In these runs ^{125}Sb tracer was added, and larger amounts of carriers. The sulfides precipitated as above were washed, digested in 9 N HCl to dissolve the antimony, which was then precipitated after centrifuging to separate them from arsenic. Well washed, this precipitate was dissolved in 9 N HCl and extracted with 10% isopropylether in benzene, and further purified as above.

As the chemical separation for these runs was done 5-10 days after irradiation only $^{120\text{m}}\text{Sb}$, and in the case of La, ^{122}Sb , could be measured. The amount of tracer ^{125}Sb used was quite small in order not to interfere too much with the gamma spectra from the reaction products, and the relative chemical yields were measured between two and three months after irradiation.

2.3 Counting of samples

Gamma-ray spectra were measured as a function of time using two different Li-drifted Ge-semiconductor detectors. The first of these was a 35 cm³ coaxially drifted detector having a resolution of about 8 keV at 1.33 MeV. This resolution was somewhat marginal for the experiment, and the next two runs were measured on a 14 cm³ planar (20 cm²) detector with a resolution of 4.0-4.3 keV at 1.33 MeV. The spectra were recorded with a 1024 channel pulse-height analyser. For some of the later runs the spectra were further transferred to magnetic tape. The spectra were plotted, and the area of the peaks belonging to each isotope present in the samples from Au, Ta, La, and Y-targets were compared to the areas of the same peaks in the samples from U-target. Integration of the peaks was done partly by hand and partly by the CDC 6600 computer using the data on magnetic tape and a FORTRAN program due to G. Astner^{*)}.

The gamma-spectra of the combined Sc activities were relatively simple and the identification of transitions due to the various isotopes was very easy using the data given in Nuclear Data Sheets¹³⁾.

On the other hand, the spectra due to Sb isotopes were extremely complicated, and the identification of prominent γ lines had to rely almost entirely upon a very recent scan of the γ spectra of different Sb isotopes obtained by bombardment of uranium, followed by chemical separation, and isotope separation as done by Hagebø and Stötzel¹⁴⁾.

The half-lives used for the calculations were those of Ravn²⁾ for the Sc isotopes, and those of Hagebø¹⁾ for the Sb isotopes.

3. RESULTS

The cross-sections obtained for Sc isotopes are given in Table 1 and the cross-sections for Sb isotopes in Table 2. In Tables 3 and 4 are given the cross-sections for Sc and Sb isotopes obtained by bombardment of uranium^{1,2)}, both for the sake of comparison and because these are the monitor cross-sections for the present work.

For all the numbers given, errors obtained by the comparison of γ lines, such as statistical counting errors, errors in the chemical yield

*) We are indebted to Dr. Astner for incorporating an extra routine in his program on our request.

measurements, reproducibility found by using several γ lines, and the errors of the Sc and Sb monitor cross-sections are included.

For the purpose of comparison of yield distributions from different targets, the errors in the monitor cross-sections which arise from the determination of these relative to the $^{27}\text{Al}(p,3pn)^{24}\text{Na}$ cross-section in Refs. 1 and 2 should not be taken into account. These errors are 7.3% and 8.5% for the scandium yields and the antimony yields, respectively.

4. DISCUSSION

4.1 Scandium cross-sections

Figure 1 shows the distributions determined in the present work. The curve drawn is only a manual fit to the points and a trial to determine the approximate position of the peak. We see a slight skewness of all the curves, the neutron excess side having a slightly smaller slope than the neutron deficient. If the mass yield curve increases towards higher mass numbers in this mass region, one expects a widening as observed. Figure 2 collects all the known distribution curves, the ones from copper and from uranium being taken from Ravn²⁾.

As already mentioned by Ravn²⁾, the curve showing the distribution from uranium at 18 GeV may be decomposed into two curves, one similar to the distribution from gold at 18 GeV and the other the Sc distribution obtained by irradiating uranium with only 570 MeV protons.

Plotting the full width at half maximum (FWHM) of the curves in Fig. 2 versus ΔA , the mass difference between target mass and the most probably scandium mass, gives Fig. 3, showing a steady increase in width over the full range. A common explanation for this increasing width is the increasing probability for neutron excess targets to give neutron excess products. We see also that the peak of the isotopic distribution shifts slowly towards higher neutron number as the mass of the target increases.

This rather steady increase in width makes less plausible the decomposition of the uranium curve into one spallation part and one fission part by using the curve from gold or tantalum targets as the prototype of spallation.

If we integrate the isotopic yield distribution curves to get the elemental yield⁵⁾ (Table 5), we see that while targets of copper, lanthanum, and gold give nearly equal yields, yttrium and uranium give much higher yields. From uranium we know that the higher yield is due to contributions from reactions involving lower deposition energies²⁾. The yttrium yield, which is a factor of two above the expected value, should be confirmed before it is further discussed.

Some isomeric yield ratios known for $^{44m}\text{Sc}(6+)/^{44}\text{Sc}(2+)$ are given in Table 6. We note that the absolute values for the isomeric yield ratio are similar to the ratio found by Matsuo and Sugihara¹⁶⁾ for $^{41}\text{K}(\alpha, n)^{44m}\text{Sc}$. Their peak value is ~ 2.0 at an α energy of 24-26 MeV.

Figure 4 shows the known ratios. The trend is for increasing ratios with increasing mass difference between target and product. This seems reasonable, as the ratios should be proportional to the angular momentum deposited by the protons. We do not know the relation between angular momentum and excitation energy imparted to the nuclei by such high-energy protons, but we would expect a relation similar to the one between linear momentum transfer and excitation energy, as calculated by Porile¹⁷⁾, to hold even for the angular momentum.

4.2 Sb isotopes

Figure 5 shows all the data plotted on one figure. It is not possible to distinguish between the gold and the tantalum curve. At 18 GeV the formation cross-section of ^{116}Sb is about the same for uranium, gold, tantalum, and lanthanum targets. In the case of lanthanum, it is probable that the widening towards higher mass numbers can be explained as the widening observed by Rudstam and Sørensen⁵⁾ for iodine isotopes from the same reaction. Among products close to the targets are also results of direct reactions giving residual nuclei with low excitation energies. These reactions may be of peripheral types, and give relatively neutron-rich products. The more common reactions involve more deposition energy, and the neutron evaporation gives products which in general lie on the far neutron-deficient side of stability. Furthermore, we see that if we subtract the gold/tantalum curve from the uranium curve we get the result which is shown in Fig. 6.

The suggestion by Rudstam and Sørensen is that by doing this, the difference curve should contain the fission contribution to the total curve of 18 GeV, whilst the spallation yields should be roughly equal for uranium and gold. For comparison, the relevant part of the distribution obtained at 570 MeV is also given in Fig. 6. The two curves compare reasonably well, although suggesting that the rather arbitrary energy of 570 MeV may be slightly too low to represent the typical fission distribution at very high energies. Otherwise, the present data support the arguments put forward by Rudstam and Sørensen; the most neutron-deficient products are spallation products. A further justification for this decomposition of the isotopic yield distribution curve is given by Brandt⁶⁾, who finds a much shorter range for the lightest iodine isotopes from uranium bombardment with 18 GeV protons than for the heavier fission products. Work in progress by the present authors¹⁸⁾ on the antimony isotopes supports the result of Brandt.

4.3 Conclusion

The present work generally supports the ideas of Rudstam and Sørensen in saying that there is a non-fission contribution to the isotopic yield distribution of antimony isotopes. Their way of decomposing the iodine curve into "spallation" and fission by means of the "spallation" curve from irradiation of gold is also applicable for the antimony distribution.

The scandium distribution, on the other hand, together with the isomeric yield ratios, make it less plausible that a similar decomposition may be performed. We see very steady changes in both yield distribution width and isomeric yield ratio with target or distance from target. While it seems less probable that fission of gold and tantalum gives rise to fission products in the antimony-iodine region, it is highly probable that these reactions produce scandium isotopes towards the neutron excess side.

A comparison of total iodine yields from gold and tantalum of 7.3 ± 1.2 mb and 6.9 ± 1.0 mb respectively⁵⁾ with the total scandium yield of 17.4 and 17.0 mb from the same targets also contradicts the classification of the scandium isotopes as spallation products. The total antimony yield integrated according to the estimation from Fig. 5 becomes very close to the iodine yields from the same two targets.

Acknowledgements

We are indebted to Prof. A.C. Pappas and to Drs. Brandt, Hudis, Kjelberg, and Rudstam for many discussions and for kind criticism of the manuscript. The skillful assistance of Mrs. R. Mohr, Mrs. W. Stötzel, and Mr. H. Bersinger at different stages of the work is gratefully acknowledged.

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Table 1

Yields of scandium isotopes obtained by
irradiation of different targets with 18.2 GeV protons

Target Nuclide	Yttrium	Lanthanum	Tantalum	Gold
	Cross-section mb	Cross-section mb	Cross-section mb	Cross-section mb
^{43}Sc *)	2.47 ± 0.24	0.67 ± 0.06	0.69 ± 0.11	0.50 ± 0.07
$^{44\text{m}}\text{Sc}$	4.59 ± 0.37	1.95 ± 0.15	1.66 ± 0.12	1.64 ± 0.12
$^{44\text{g}}\text{Sc}$	3.81 ± 0.29	1.35 ± 0.10	0.80 ± 0.07	0.76 ± 0.06
^{46}Sc	8.36 ± 0.69	4.23 ± 0.34	4.23 ± 0.32	4.42 ± 0.36
^{47}Sc	4.27 ± 0.32	2.72 ± 0.20	3.16 ± 0.23	3.23 ± 0.25
^{48}Sc	1.13 ± 0.09	1.15 ± 0.09	1.36 ± 0.10	1.53 ± 0.11

*) Cumulative yields.

Table 2

Yields of antimony isotopes obtained by
irradiation of different targets with 18.2 GeV protons

Target Nuclide	Lanthanum		Tantalum		Gold	
	No. of det.	Cross-section mb	No. of det.	Cross-section mb	No. of det.	Cross-section mb
^{115}Sb †)	2	4.95 ± 0.55	2	6.02 ± 0.69	1	5.70 ± 0.65
^{116}Sb	2	1.90 ± 0.26	2	1.82 ± 0.18	1	1.63 ± 0.17
^{117}Sb	2	2.2 ± 0.3	2	1.6 ± 0.4	1	$3.16^*)$
^{118}Sb	2	1.52 ± 0.15	2	0.644 ± 0.062	1	0.788 ± 0.076
^{120}Sb	3	0.49 ± 0.05	4	0.140 ± 0.016	2	0.150 ± 0.017
^{122}Sb	3	0.176 ± 0.018				

†) Cumulative yields.

*) Partly cumulative yield.

Table 3

Yields from Ref. 2 of scandium isotopes obtained by irradiation of uranium with 18.2 GeV protons

Nuclide	Cross-section mb
$^{43}\text{Sc}^*)$	0.467 ± 0.015
$^{44\text{m}}\text{Sc}$	1.70 ± 0.05
$^{44\text{g}}\text{Sc}$	0.87 ± 0.03
^{46}Sc	6.50 ± 0.21
^{47}Sc	6.16 ± 0.19
^{48}Sc	3.60 ± 0.11
$^{49}\text{Sc}^*)$	1.76 ± 0.05

*) Cumulative yields.

Table 4

Yields from Ref. 1 of antimony isotopes obtained by irradiation of uranium with 18.2 GeV protons

Nuclide	Cross-section mb
^{115}Sb	$4.03 \pm 0.41^*)$
$^{116\text{g}}\text{Sb}$	2.02 ± 0.18
^{117}Sb	2.85 ± 0.31
$^{118\text{g}}\text{Sb}$	3.17 ± 0.30
$^{120\text{m}}\text{Sb}$	2.65 ± 0.24
^{122}Sb	2.88 ± 0.24

*) Cumulative yield.

Table 5

Elemental yield of scandium for different targets
irradiated with 18.2 GeV protons

Target	Elemental yield mb
Cu	20.3
Y	36.3
La	17.6
Ta	17.0
Au	17.4
U	28.0

Table 6

Isomeric yield ratio for ^{44}Sc for different targets

Target	ΔA mass units	$\frac{\sigma(^{44\text{m}}\text{Sc } 6+)}{\sigma(^{44\text{g}}\text{Sc } 2+)}$
Copper	20	$0.58 \pm 0.02^{2)}$ $0.98 \pm 0.11^{15)}$
Yttrium	45	1.20 ± 0.13
Lanthanum	95	1.44 ± 0.15
Tantalum	137	2.08 ± 0.24
Gold	153	2.16 ± 0.23
Uranium	194	$1.96 \pm 0.09^{2)}$

Figure captions

Fig. 1 : Isotopic yield distributions of Sc isotopes obtained by irradiating targets of Y, La, Ta, and Au with 18.2 GeV protons. The solid lines connect the measured points.

Fig. 2 : Comparison of the Sc isotopic yield distribution curves from different targets. The Cu and U curves are taken from Ref. 2.

Fig. 3 : Full width at half maximum for the curves given in Fig. 2, as a function of mass difference between target and the peak position of the different curves.

Fig. 4 : Isomeric yield ratio $\sigma(^{44m}\text{Sc})/\sigma(^{44g}\text{Sc})$ from different targets irradiated with 18.2 GeV protons as a function of the mass difference between target and product.

Fig. 5 : Isotopic yield distributions of antimony isotopes obtained by irradiating La, Ta, and Au with 18.2 GeV protons. The curve from U is included for comparison (Ref. 1):

● : Lanthanum target ; ▲ : Tantalum target

□ : Gold target ; ○ : Uranium target¹⁾

Fig. 6 : Decomposition of the uranium curve into a spallation part (Ta curve) and a fission part (dotted line). The normalized distribution curve from uranium at 570 MeV¹⁾ is included for comparison.

FIG.1

FIG.2

FIG. 3

FIG. 4

FIG.6

1944-1945

1946-1947

1948-1949

1950-1951

1952-1953