To be submitted to
Nuclear Physics

ISTITUTO NAZIONALE DI FISICA NUCLEARE
Laboratori Nazionali di Frascati

LNF-83/34(P)
14 Maggio 1983

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PRODUCTION CROSS SECTIONS FOR Sr AND Cs BOMBARDED BY 600 MeV PROTONS

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ABSTRACT

We have measured the direct cross sections for the production of various radionuclides by bombardment of Sr and Cs with 600 MeV protons. Some considerations are inferred about the production of radionuclides in the metastable state and in the fundamental state, which can be used for activation and trasmutation studies. The measured direct cross sections are compared with calculations performed using well known semiempirical formulae. We calculated also some cumulative cross sections by a complete transmutation computer program.

1. - INTRODUCTION

The production of a nuclide (Z_1, A_1) by inelastic interactions of high energy nucleons ($E > 100$ MeV) with a given target nuclide (Z,A) can proceed through different channels: the nuclide can be produced from the parent by different direct reactions with emission of several nucleons, free or compound to form heavier particles. A theoretical model that explains these reactions is the two steps mechanism: intranuclear cascade plus evaporation.

However, for general transmutation or activation studies, it is enough to know a total cross section for the formation of a product from a target which includes all the channels; semiempirical formulae, based on such a model, which allow to calculate these total cross sections have been proposed(1).

In addition to this direct mode of production, a given nuclide can also be produced from a target nuclide through successive steps of direct reaction and fast decay of intermediate products.

Whenever only the final products of an irradiation are at stake, it may be useful also for this mode of production to know just a "cumulative" fictive cross section for the production of a given isotope. It has to be kept in mind that such a "cross section" is a function of the irradiation time and only when the half lives of the possible intermediate products are very short compared to the irradiation time, this dependence can be ignored.

We have exposed Sr and Cs samples to 600 MeV protons and have measured, when it has been possible, the direct cross sections for the production of some radioisotopes. These cross sections are compared with calculations using the semiempirical formulae. We calculated also some cumulative cross sections by a transmutation computer program that follows the formation and decay of radioisotopes during the irradiation using the direct semiempirical cross sections.

2. - EXPERIMENTAL TECHNIQUES

2.1. - Target and Irradiation

We irradiated samples of natural Sr and Cs: natural Sr is composed by 0.55% of ^{84}Sr, 9.75% of ^{86}Sr, 6.96% of ^{87}Sr and 82.74% of ^{88}Sr while the natural Cs is composed by 100% of ^{133}Cs.

Pressed pellets 1 mm thick of $SrCO_3$, $Sr(NO_3)_2$ and CsCl where used as targets. The presence of C,O,N,Cl and other low mass number A impurities in the samples did not influence the production of the investigated nuclides because these latter have a mass number higher than the impurities and cannot be produced by them. The chemical analysis of the samples showed also the presence of traces of Pb in the negligible amount of 0.0005% in weight.

The targets were irradiated into the CERN Synchrocyclotron 600 MeV extracted proton beam. The proton flux was measured with thin Al foil, positioned in front and behind the target, by measuring the ^{24}Na produced in the reactions ^{27}Al(p,3p n) ^{24}Na and ^{27}Al(p,4p) Ne^{24}$\rightarrow$^{24}Na with a total cross section of (10.8 ± 0.7)mb[2].

2.2. - Measurements

The radionuclides produced have been identified through gamma spectroscopy. We used a Ge(Li) detector connected to a 4096 channel analyzer. In Table I we show the radionuclides' characteristics[3] used in the cross section calculations.

The computer program SAMPO[4] was used for identifying and calculating the activities of the produced radionuclides: the activities were checked by the various gamma lines, when present, taking into account the eventual superposition of lines from different radionuclides.

When a radioisotope is produced by direct interaction from the parent or when the half lives of the eventual intermediate radionuclides are very long compared to the irradiation time, the total production cross section is given by

$$\sigma = \frac{A(T_{irr})}{N_T(0)\,\phi\,\left(1 - e^{-\lambda . T_{irr}}\right)}$$

where:

TABLE I - Nuclear characteristics of the identified radionuclides.

Nucleus	Half-life	Energy of the specific γ-lines (keV)	Number of γ-rays per decay (%)
^{87m}Sr	2.805 h	388	82 (with IT)
^{87}Y	80.3 h	388	87.4
^{88}Y	106.61 d	1836	99.34
^{86}Y	14.74 h	1077	82.5
^{84}Rb	32.77 d	882	74 (with β^++EC)
^{82m}Rb	6.2 h	776	82.6
^{74}As	17.79 d	596	60 (with EC+β^+)
^{132}Cs	6.474 d	688	97.5 (with EC+β^+)
^{127}Cs	6.25 h	411	62
^{124}I	4.15 d	603	61
^{118m}Sb	5.00 h	1230	99.9

$A(T_{irr})$ = activity of isotope at the end of irradiation;
T_{irr} = irradiation time;
$N_T(0)$ = initial amount of target isotope;
λ = decay constant of isotope.

For the case of the products ^{87m}Sr and ^{87}Y, that have a γ-line in common, the decay scheme and the characteristic data are:

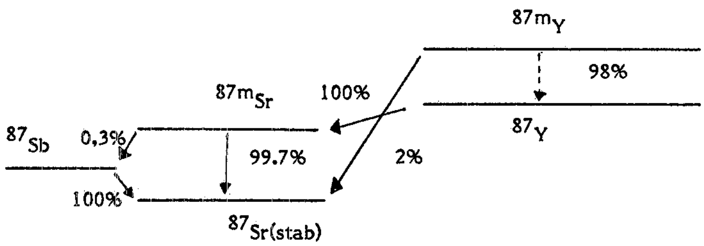

FIG. 1 - Decay scheme and characteristic data for ^{87m}Sr and ^{87}Y.

Nucleus	Half-life	Decay-type	Energy of the specific γ-line (keV)	Number of γ-rays per decay (%)
^{87m}Sr	2.805 h	IT 99.7% EC 0.3%	388	82 (with IT)
^{87}Y	80.3 h	EC, β^+	388	87.4

The equations for the activities are:

$$A_m^o = A_m^o(t)\, e^{\lambda_m t}$$

$$A_g(t) = A_m^o \frac{\lambda_g}{(\lambda_g - \lambda_m)} e^{-\lambda_m t} + (A_g^o - A_m^o \frac{\lambda_g}{(\lambda_g - \lambda_m)}) e^{-\lambda_g t}$$

$$A_p(t) = \left[A_p^o - \frac{\lambda_p \lambda_g A_m^o}{(\lambda_p - \lambda_m)(\lambda_g - \lambda_m)} - \frac{\lambda_p}{(\lambda_p - \lambda_g)} (A_g^o - \frac{A_m^o \lambda_g}{(\lambda_g - \lambda_m)}) \right] e^{-\lambda_p t} +$$

$$+ \frac{\lambda_p}{(\lambda_p - \lambda_m)} \frac{A_m^o \lambda_g}{(\lambda_g - \lambda_m)} e^{-\lambda_m t} + \frac{\lambda_p}{(\lambda_p - \lambda_g)} (A_g^o - A_m^o \frac{\lambda_g}{(\lambda_g - \lambda_m)}) e^{-\lambda_g t}$$

where:

A^o = activity at the end of irradiation;

λ = decay constant

t = time from the end of irradiation;

$\bar{P}$ = number of γ per decay (in %)

and the subscripts, g, m and p refer to ^{87}Y, ^{88m}Y and ^{87m}Sr respectively.

For the intensity of the 388 keV γ-line we have

$$I(t) = \bar{P}_p A_p(t) + \bar{P}_g A_g(t)$$

Thus, the measurement of $A_m(t)$ and two measurements of I(t), at different times, provide all the necessary informations for the determination of A_g^o and A_p^o.

2.3. - Contribution of Secondary Processes

From the thickness of the target we calculated that the probability that a nucleon formed by interaction of the primary proton interacts with a nucleus in the target is about 1/100 of that of the primary interaction. Secondary interactions were, then, neglected in the cross section calculations.

3. - RESULT AND DISCUSSION

The measured and calculated cross sections for the direct production of some radionuclides from Sr and Cs are shown in Table II and Table III respectively; calculated cumulative cross sections are shown in Table IV and Table V.

The errors in the cross sections have been estimated by compounding the errors of the γ-lines percentages, of the measured proton fluxes and of the measured intensities. This last error includes the statistical, energy calibration and efficiency calibration errors of the detector.

The cross sections for the production of ^{86}Y and ^{84}Rb in Table II are slightly overestimated because also their metastable states (indicated in brackets) contribute to their production and, in some cases, they have not half lives long compared to the irradiation time.

TABLE II - Measured and calculated cross sections for the direct production of some radionuclides from interaction of 600 MeV protons with isotopes of natural Sr.

Target isotope	Reaction	Produced radionuclides	Measured cross-section (mb)	Calculated cross-section (mb)
^{88}Sr	(p,pn)	^{87m}Sr	14.4 ± 4.6	52.0(1)
^{88}Sr	(p,n)	^{88}Y	1.5 ± 0.3	1.2
$^{88}Sr+^{87}Sr$	several	^{87}Y	2.0 ± 0.4	2.7(2)
$^{88}Sr+^{87}Sr+^{86}Sr$	several	$^{86}Y+(^{86m}Y)$	2.3 ± 0.4	1.4
$^{88}Sr+^{87}Sr+^{86}Sr$	several	$^{84}Rb+(^{84m}Rb)$	21.2 ± 3.3	20.9
$^{88}Sr+^{87}Sr+^{86}Sr+^{84}Sr$	several	^{82m}Rb	17.5 ± 1.9	56.9(3)
$^{88}Sr+^{87}Sr+^{86}Sr+^{84}Sr$		^{74}As	10.6 ± 1.6	6.9

(1) Includes the cross section for ^{87}Sr production

(2) Includes the cross section for ^{87m}Y production

(3) Includes the cross section for ^{82}Rb production

TABLE III - Measured and calculated cross sections for the production of some radionuclides from interaction of 600 MeV protons with ^{133}Cs.

Target isotope	Reaction	Produced radionuclides	Measured cross section (mb)	Calculated cross section (mb)
^{133}Cs	(p,pn)	^{132}Cs	59.2 ± 6.2	61.1
^{133}Cs	(p,p 6 n)	^{127}Cs	18.2 ± 2.6	14
^{133}Cs	(p, 3p 7n) or (p, spall)	^{124}I	10.8 ± 1.5	9.5
^{133}Cs	(p.spall)	^{118m}Sb	3.8 ± 0.7	5.7(1)

(1) Includes the cross section for ^{118}Sb production.

TABLE IV - Calculated "cumulative" cross sections for production of some radionuclides from ^{88}Sr.

Radionuclide	"Cumulative" cross section (mbarn)
^{83}Sr	0.033
^{79}Kr	42.97
^{77}Br	35.50
^{71}As	5.95
^{69}Ge	6.343
^{69m}Zn	0.19

TABLE V - Calculated "cumulative" cross sections for the production of some radionuclides from ^{133}Cs.

Radionuclide	"Cumulative" cross section (mbarn)
^{122}Xe	68.3
^{125}Xe	69.6
^{129}Cs	24.1
^{121m}Te	34.2
^{127}Xe	18.6

We wrote a computer program[6] that uses the semiempirical formulae[1], adjusted with more experimental data[7], for calculating the direct production cross sections; this program is used as a subroutine in the transmutation program[5]. These calculated cross sections are shown in Tables VI and VII.

In Table II for each produced radionuclide are indicated the natural isotopes of Sr that can contribute to its production; when more than one isotope of Sr may contribute, the calculated cross section is a weighted average of the cross sections for each of the isotopes.

For the calculations with the trasmutation program, when isomers are present in the products, the value of the cross section has been divided into equal parts amongst the isomers. Tables VIII and IX, which show a comparison between calculated and measured activities of various identified radionuclides, confirm that assumption and this makes possible the calculation of activities of metastable nuclides. By combining measured and calculated activities, it is possible to roughly evaluate the ratio of the cross sections for the production of the nuclide in the fundamental state (σ_g) and that in the metastable state (σ_m).

These ratios are shown in Table X and they show that, in general, the production of the fundamental state does not look to be privileged over the metastable one: then a $(\sigma_g/\sigma_m) \simeq 1$ is an acceptable assumption.

TABLE VI - Cross sections for peripheral and spallation reaction of 600 MeV protons with ^{88}Sr.

Reaction / A of product	(p,xn)	(p,pxn)	(p,2pxn)	(p,3pxn)	(p,spall)	(p,spall)	(p,spall)	(p,spall)	(p,spall)	(p,spall)
88	1.24	T								
87	2.64	52.01	22.5							
86	1.24	0.18	26.9	0.35						
85		0.10	27.6(x)	1.75						
84		0.05	9.47	1.95						
83		0.03	41.3	33.6						
82		0.02(x)	57.1	58.8(x)						
81		4.61	39.4	25.5						
80		1.34	14.4	54.2	4.72					
79			4.65	38.8	12.2	0.82	0.02			
78				21.5	22.3	3.27	0.07			
77				5.64	29.7	7.02	0.31	0.01		
76				1.82	14.3	18.9	1.02	0.04		
75				0.30	5.39	23.6	3.16	0.14		
74				0.07	1.39	19.9	7.12	0.67	0.01	
73						6.18	13.6	1.71	0.05	
72						2.25	12.1	5.60	0.2	0.01
71						0.41	5.57	9.13	0.73	0.02
70						0.10	1.64	15.0	1.97	0.12
69							0.42	6.00	4.76	0.37
Z (symbols of product)	39 (Y)	38 (Sr)	37 (Rb)	36 (Kr)	35 (Br)	34 (Se)	33 (As)	32 (Ge)	31 (Ga)	30 (Zn)

T : Target nucleus $^{88}_{38}Sr$.

(x) For isotoper with A smaller than this, the reaction is a (p,spall).

TABLE VII - Cross sections for peripheral and spallation reaction of 600 MeV protons with ^{133}Cs.

Reaction / A of product	(p,xn)	(p,pxn)	(p,2pxn)	(p,3pxn)	(p,spall)	(p,spall)	(p,spall)	(p,spall)	(p,spall)	(p,spall)
133	1.45	T								
132	3.10	61.1	25.8							
131	1.45	49.0	28.2	0.27						
130		33.9	27.6(*)	0.52						
129		24.6	1.18	1.38						
128		18.3	3.94	3.47						
127		14.0	7.62	7.00						
126		10.9(*)	20.6	10.9(*)						
125		48.4	31.2	4.26						
124		62.7	62.3	9.51	0.56					
123		44.7	61.9	18.6	1.25	0.04				
122			50.0	30.9	3.93	0.14	0.01			
121			19.0	40.6	7.06	0.43	0.02			
120			9.29	28.6	17.3	1.10	0.07			
119			2.48	13.9	23.0	2.83	0.18			
118			0.90	5.50	37.0	5.73	0.65	0.02		
117				1.85	19.8	11.2	1.35	0.06		
116					11.7	15.9	3.93	0.17	0.01	
115					3.61	16.4	6.36	0.50	0.02	
114						8.30	13.4	1.20	0.09	
113						3.69	14.3	2.80	0.22	0.01
Z (symbol) of product	56 (Ba)	55 (Cs)	54 (Xe)	53 (I)	52 (Te)	51 (Sb)	50 (Sn)	49 (In)	48 (Cd)	47 (Ag)

T: Target nucleus $^{133}_{55}Cs$.

(*) For isotopes with A smallet than this, the reaction is a (p,spall).

TABLE VIII - Radionuclides identified from irradiation of natural Sr. We compare the measured activity of the radionuclide and the activity calculated by the transmutation program using the cross sections of Table VI. The activities indicated are normalized to 10^{10} nuclei of the target isotope. Errors in the measured activities are within the limit of ±10%.

Identified radionuclite	Measured activity (Ci)	Calculated activity (Ci)
^{86}Y	0.87×10^{-7}	0.52×10^{-7}
^{87}Y	0.25×10^{-7}	0.23×10^{-7}
^{88}Y	0.35×10^{-8}	0.27×10^{-8}
^{83}Sr	0.47×10^{-7}	0.96×10^{-7}
^{85}Sr	0.65×10^{-8}	0.16×10^{-8}
^{87m}Sr	0.15×10^{-6}	0.24×10^{-6}
^{82m}Rb	0.60×10^{-6}	0.99×10^{-6}
^{83}Rb	0.87×10^{-8}	0.89×10^{-3}
^{84}Rb	0.14×10^{-6}	0.14×10^{-6}
^{79}Kr	0.08×10^{-5}	0.18×10^{-5}
^{77}Br	0.10×10^{-5}	0.15×10^{-5}
^{73}Se	0.40×10^{-8}	0.35×10^{-8}
^{75}Se	0.65×10^{-7}	0.61×10^{-7}
^{71}As	0.17×10^{-6}	0.11×10^{-6}
^{74}As	1.29×10^{-7}	0.84×10^{-7}
^{66}Ge	0.26×10^{-8}	0.53×10^{-8}
^{69}Ge	0.16×10^{-6}	0.16×10^{-6}
^{69m}Zn	0.81×10^{-8}	0.68×10^{-8}

TABLE IX - Radionuclides identified from irradiation of natural Cs. We compare the measured activity and the activity calculated by the trasmutation program using the cross section of Table VII. The activities indicates are normalized to 10^{10} nuclei of the target isotope. Errors in the measured activities are within the limit of $\pm 10\%$.

Identified radionuclide	Measured activity (Ci)	Calculated activity (Ci)
^{131}Ba	0.65×10^{-7}	0.45×10^{-7}
^{127}Cs	0.22×10^{-5}	0.17×10^{-5}
^{129}Cs	0.46×10^{-5}	0.45×10^{-5}
^{132}Cs	0.22×10^{-5}	0.28×10^{-5}
^{122}Xe	0.31×10^{-5}	1.57×10^{-5}
^{125}Xc	0.72×10^{-5}	1.82×10^{-5}
^{127}Xe	0.15×10^{-6}	0.23×10^{-6}
^{123}I	0.27×10^{-4}	0.32×10^{-4}
^{124}I	0.64×10^{-6}	0.56×10^{-6}
^{131}I	0.07×10^{-7}	0.11×10^{-7}
^{119}Te	0.43×10^{-5}	0.45×10^{-5}
^{119m}Te	0.09×10^{-5}	0.15×10^{-5}
^{121}Te	0.85×10^{-8}	0.72×10^{-3}
^{121m}Te	0.54×10^{-8}	0.20×10^{-8}
^{118}Sb	0.49×10^{-9}	0.35×10^{-9}

Nuclide	σ_g/σ_m
^{82}Rb	2.25
^{118}Sb	0.5
^{87}Sr	2.6

TABLE X - Ratio of the cross section for the production of ground to metastable state of some radionuclides.

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