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A contribution to the waste transmutation study

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HIGH ENERGY PROTON INTERACTIONS WITH Sr AND Cs

A contribution to the waste transmutation study

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SUMMARY

We studied the production of stable and radioactive isotopes by irradiation of ^{90}Sr and ^{137}Cs in high energy particle beams.

A fast computer program has been written which follows the yield of the various nuclides through successive trasmutation and decay processes. The cross sections for the high energy reactions were calculated using semiempirical equations.

Samples of Sr and Cs have been exposed to a 600 MeV proton beam.

The comparison of experimental and theoretical results confirms the validity of the semiempirical formulas as well as of the transmutation code.

This work is a contribution to the study of the scientific feasibility of using charged particle beams for transmutation to reduce the radioactivity of high level nuclear waste materials.

1. - INTRODUCTION

The possibility of using high energy accelerator beams for transmuting radioactive waste material is presently investigated at several places. An International Conference on Nuclear Waste Transmutation was held in 1980 at the University of Austin, Texas, USA[(1)].

However, many fundamental data on reactions of different particles with radioactive isotopes are needed for evaluating the feasibility of such a possibility.

We present the results of a theoretical and experimental study on the interactions of 600 MeV protons with Sr and Cs.

We calculated the total inelastic cross sections and the cross sections of spallation and peripheral reactions of Sr, Cs and neighbouring isotopes with high energy protons.

A computer program was written which follows the interactive transmutation and decay chains of Sr and Cs yields for different irradiation and decay times.

We exposed samples of natural Sr and Cs to a beam of 600 MeV protons and singled out a number of radionuclides by γ-ray spectroscopic analysis.

This allowed us to verity the calculated cross sections as well as the transmutation program.

2. - TOTAL INELASTIC, PERIPHERAL AND SPALLATION CROSS SECTIONS

We limited our transmutation study to elements with $A \geqslant 66$ for the parent Sr and with $A \geqslant 116$ for the parent Cs. We calculate the peripheral and spallation cross sections from and to the considered isotopes using the semiempirical formulas of Rudstam as correct by Silberberger and Tsao[2]. The total inelastic cross sections were approximated to the total geometrical absorption cross sections[3]

$$\sigma = \pi (1{,}26 \times 10^{-13} \times A^{1/3})^2 \qquad \text{(in cm}^2\text{)} \qquad (1)$$

The cross sections for (p,n), (p,2n), (p,3n) reactions were calculated using data found in bibliography (4,5). For isotopes that have also metastable states, the cross section was shared into equal parts between the fundamental and the metastable states of the isotope.

In Table I and II, as example, are reported the production cross sections of the various isotopes starting from ^{88}Sr and ^{133}Cs respectively.

TABLE I - Cross sections for peripheral and spallation reactions 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(*)	1.75						
84		0.05	9.47	1.95						
83		0.03	41.3	33.6						
82		0.02(*)	57.1	58.8(*)						
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 (symbol) 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.

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

TABLE II - Cross sections for peripheral and spallation reactions 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$^{(x)}$	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$^{(x)}$	20.6	10.9$^{(x)}$						
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	13.5	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.
(x) For isotopes with A smaller than this, the reaction is a (p,spall).

3. - THE TRANSMUTATION PROGRAM

A computer program has been written that follows the transmutation-decay chain of a target isotope. It has presently been used for following the chain of reactions of Sr and Cs with high energy protons.

The program solves a system of differential equations of the type:

$$\frac{dN_i}{dt} = - N_i (\phi \ _i + \lambda_i) + \sum_k N_k (\phi \ _{k,i} + \lambda_{k,i}) \qquad (2)$$

where

N_i = amount of isotope i
ϕ = interacting particle flux ($cm^{-2}s^{-1}$)
λ_i = decay constant of isotope i
σ_i = total inelastic cross section of the isotope i
$\sigma_{k,i}$ = production cross section from isotope k to isotope i
$\lambda_{k,i}$ = decay constant of isotope k into isotope i (if that decay is possible)

Equation (2) can be written as

$$\frac{dN_i}{dt} = \sum_k N_k A_{k,i} \qquad (3)$$

where

$$A_{k,i} = \begin{cases} \phi\sigma_{k,i} + \lambda_{k,i} & \text{for } k \neq i \\ -(\phi\sigma_i + \lambda_i) & \text{for } k = i \end{cases}$$

The numerical solution has been obtained be developind $N_i(t)$ into Taylor series in each step of the integration:

$$N_i(t+h) = N_i(t) + \sum_{j=1}^{P} \frac{h^j}{j!} N_i^{(j)}(t) \qquad (4)$$

where

$$N_i^{(j)} = {}_{k} N_k^{(j-1)} A_{k,i}$$

$$N_k^{(0)} = N_k$$

The calculation algorithms and the computer program are fully described in ref. (6).

Such a program can be used for calculating transmutation chains for the interaction of any isotopes with any particles once the reactions, cross sections and decay contants are fed in.

4. - EXPERIMENTAL RESULTS

Samples of $Sr(NO_3)_2$, $SrCO_3$ and CsCl of about 1 g each were compressed to form a solid pellet and exposed for different times into the 600 MeV proton beam at the CERN Synchrocyclotron. The intensity of the beam at the sample was about 10^{13} protons s^{-1} as measured by the activation of Al foils in front of the samples.

The radioactive isotopes formed in the sample were identified by gamma spectroscopy and by decay half life. We used a Quartz et Silice GeLi detector (55 cm^3) connected to a Camberra multichannel analyzer. Also the SAMPO computer program was used for identifying the peaks.

5. COMPARISON OF THEORETICAL AND EXPERIMENTAL RESULTS

A radionuclide can be produced by transmutation of the main parent and/or of a product nuclide as well as by decay of product nuclides. By comparing the amount of each radionuclide formed, as calculated by the computer program with the amount measured one can verify the validity of the program and the precision of the calculated cross sections.

In Table III and IV we show the calculated and measured activities of different radionuclides produced from Sr and Cs respectively. For some reactions, where the radioisotope production chain is simpler, it has been possible to calculate from the experimental data the cross section for the production reaction. In Table V and VI are shown the measured and the calculated cross sections.

The data of Tables III to VI experimentally confirm the correctness of the computer program as well as the validity of the Silberberger and Tsao semiempirical formulas for the calculation of the cross sections for targets in our range of Z and A.

TABLE III - 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 I. The activities indicated are normalized to 10^{10} nuclei of the target isotope.

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 IV - Radionuclides identified from irradiation of natural Cs. We compare the measured activity and the activity calculated by the transmutation program using the cross section of Table II. The activities indicated are normalized to 10^{10} nuclei of the target isotope.

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}

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

Target isotope	Reaction	Produced isotope	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.

6. - WASTE TRANSMUTATION APPLICATION

An as example of the use of the computer code, we studied the yield of a 600 MeV proton beam on ^{90}Sr (h.l.: 28,8 y) and ^{137}Cs (h.l.: 30,17 y). For simplifying the calculations we used rather hypothetical parameters: a beam intensity of 10^{17} p cm^{-2} s^{-1}, very thin pure ^{90}Sr and ^{137}Cs targets such that we considered only interactions with the primary beam, and short irradiation times (14 day for the ^{90}Sr and 10 days for the ^{137}Cs).

We are aware that these conditions are unrealistic: however the results, that are rather interesting, will be used for optimizing the parameters in more realistic case studies. We calculated the amount and the activity of each produced radionuclide as well as the total activity at different times during the irradiation period and after several decay times. The detailed results are reported in refs. (6) and (7).

We summarize some of the results:

a) ^{90}Sr case. We limited the yield to 157 nuclides of atomic mass between 66 and 90. After the 14 days irradiation period, 11,5% of the ^{90}Sr has disappeared. The total activity of the products has increased by a factor of about 20 as compared to the initial activity of the ^{90}Sr (+^{90}Y). However the vast majority of the products have very short half lives. In Fig. 1 we show the build up and decay of the total activity of the radionuclides produced. After some 2.000 days of decay, the total activity has decreased to about 18% of the initial activity of ^{90}Sr.

The very long half life radionuclides produced are ^{87}Rb (4.8×10^{10} y), ^{81}Kr (2.1×10^{5} y) and ^{79}Se (6.5×10^{4} y) : their activities are respectively 9×10^{-13}, 4×10^{-7} and 4×10^{-8} of the initial activity of ^{90}Sr.

b) ^{137}Cs case. The yield was limited to 157 nuclides of atomic mass between 119 and 137. After 10 days of irradiation 10,9% of the ^{137}Cs has been "burned", the total radiactivity of the products is about 30 times that of original ^{137}Cs but it is formed mainly by short life radionuclides. Fig. 2 shows the decay of the total radioactivity of the products. After some 2.000 days it is reduced to about 3×10^{-5} of the initial activity of ^{137}Cs. The very long lived radionuclides produces are ^{121m}Sn (55 y), ^{135}Cs (3×10^{6} y), ^{129}I (1.6×10^{5} y) and ^{126}Sn (10^{5} y) which have an activity of respectively, 6×10^{-7}, 1.1×10^{-8}, 2×10^{-11} and 1.2×10^{-12} of the initial activity of the ^{137}Cs.

This computer code will be used for studying and optimizing more realistic cases of waste transmutation.

FIG. 1 - The decay of the radionuclides formed by irradiation of ^{90}Sr with 600 MeV protons.

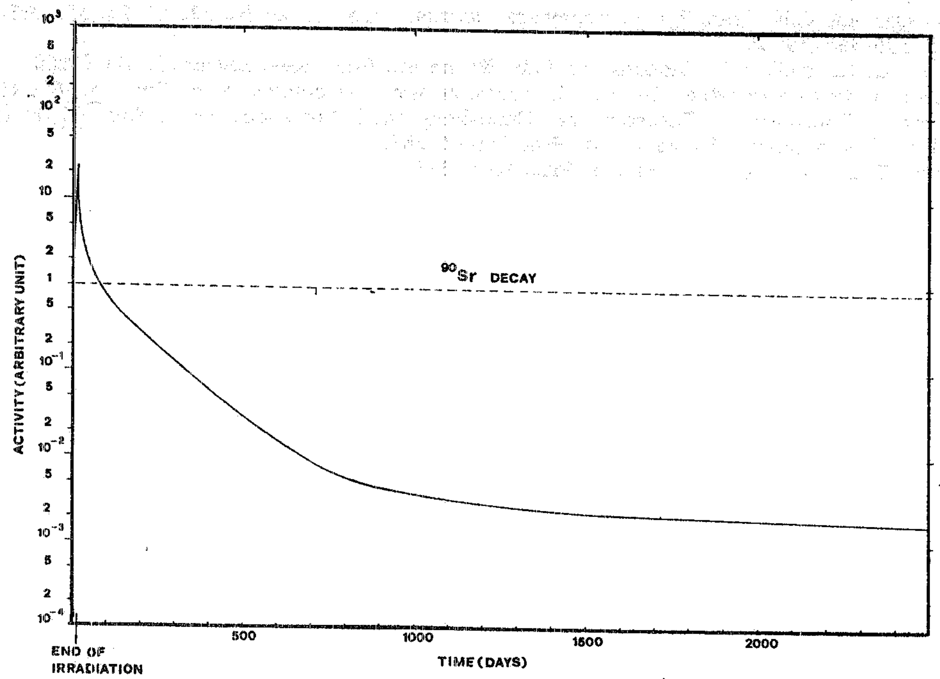
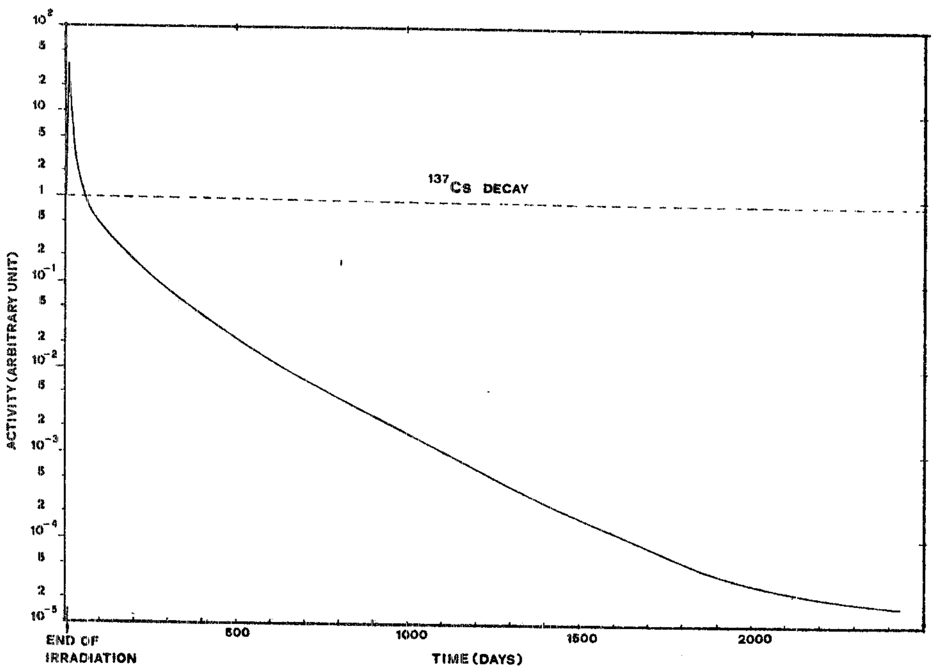


FIG. 2 - The decay of the radionuclides formed by irradiation of ^{137}Cs with 600 MeV protons.



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