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UTILIZATION OF AN INTENSE BEAM OF 800 MEV PROTONS TO PREPARE RADIONUCLIDES

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Abstract

Since the early 1970's, a program has been underway at this institution to employ the excess proton beam emerging from the major experimental areas of the LAMPF accelerator to make a wide variety of radioactive nuclides. This paper presents a review of the targets irradiated, cross section data, and nuclide yield measurements.

1. Introduction

Early in the development of the Clinton P. Anderson Meson Physics Facility (LAMPF), concerted efforts were underway to develop a number of non-meson-physics programs that could utilize the unique resources of this accelerator. One of these efforts was concerned with designing and building a targeting system (1), to be located immediately upstream of the main LAMPF beam stop, that would allow a variety of target materials to be inserted into the excess proton beam emerging from the main experimental areas. This was an immense and exciting challenge to those of us that were involved in the program, as the beam would reach a power level of 800 kW, and massive shielding would be required for the ejected neutrons and gamma radiation, yet the operation of the facility, later named the Isotope Production Facility (IPF), was required to have minimal impact on the overall operation of the accelerator.

The objective of this paper is to briefly summarize the preparatory research carried out with 590 MeV protons. This will be followed by a review of the results that were obtained during the 15-year period I was associated with and directed the Medical Radioisotope Research Program. Obviously the constraints on the length of this paper limit the amount of data that can be presented here, but numerous publications will be cited for those wishing more extensive data.

2. Spallation Reaction Studies at 590 MeV

To prepare for this program, a small group of nuclear chemists and physicists was formed and commenced to irradiate a variety of target elements in the low intensity 590 MeV proton beam available from the synchrocyclotron at the Space Radiation Effects Laboratory located in Newport News. These data would provide a planning basis for the LAMPF radioisotope production program. Thin target foils of the following elements were irradiated: C, Mg, Al, Si, Ti, V, Cr, Fe, Co, Ni, Cu, Zn, Ge, As, Se, Nb, Mo, In, Sn, Ba, La, Ce, Tb, Lu, Hf, Ta, W, Au, Pb, Bi, and U. Ge(Li) spectrometry was employed to measure the yields of the major gamma photon emitting nuclides produced in the irradiated targets. Chemical separations were carried out on the higher Z targets, as needed. Many of the targets studied by us at this energy have not been studied by others at similar energies to permit comparisons. Some publications (2,3) contained product yield data for Ti, Fe, and Cu targets bombarded with 600 MeV protons, and comparisons showed wide disagreements (see Tables 1, 2, and 3). Because of this, an interlaboratory comparison was carried out in which identically irradiated foils of Fe and Cu, plus Al and C monitor foils, were distributed to the three participating laboratories for measurement(4). The resulting data showed generally good agreement.

Published data (5,6) from Fe and Cu targets, bombarded with 1 and 3 GeV protons in a thick target array, indicate a buildup in the effective cross sections for products relatively close in mass to that of the target. At 1 GeV the buildup factors, which are attributed to the increase in particle flux due to the production of secondaries, reach their maximum for the 0 to 2.5 cm radial zone at approximately 40 g cm^{-2}. A similar experimental arrangement was used by us to measure thick target yields in Fe, Nb, and Bi targets in an iron array at 590 and 300 MeV, since our planned LAMPF targets were to be on the order of 2.5 cm thick. The resultant curves, obtained by plotting the effective cross section vs depth in the array, were found to be similar to those obtained at the higher proton energies, with the exception that the maxima are shifted to approximately 30 g cm^{-2} depth(see Figure 1 for an example). Thus one could expect enhanced yields for some product nuclides in the LAMPF targets.

3. LAMPF Target Irradiations

The LAMPF is centered around a half-mile long linear accelerator designed to accelerate a high intensity beam of protons to energies well beyond the pion production threshold (7,8). The IPF, as mentioned above, contains nine independent target stations in series immediately upstream of the main LAMPF beam stop. Targets are typically on the order of 6.4 cm OD by 2 cm thick and are encased in a bolted container, which has been made of gold-plated copper, stainless steel, or aluminum. In addition, a number of different target encapsulation methods have been developed and tested (9).

In contrast with low-energy charged-particle nuclear reactions, loss of particle energy in target windows and coolant is not a great consideration at 800 MeV. Physical strength, high heat transfer, and resistance to radiation damage are important factors in selecting materials for use in encapsulation systems.

When operations at the IPF were initiated in October, 1976, LAMPF was delivering 800 MeV protons at a current of 300 microamperes. Initially only pure metal targets were selected for irradiation. These included Mo, Ni, Ta, Si, V, Al, and Cu, which ranged in mass from a few to several hundred grams. One program required a multicurie quantity of

^{88}Y that necessitated the irradiation and processing of more than 5 kilograms of Mo metal. As experience was gained, it was learned that oxide and salt targets could safely be irradiated, and we eventually developed a targeting system that permitted the sweeping and cold trapping of radioxenon gas from a molten CsCl during irradiation (10).

4. Spallation Reaction Studies at 800 MeV

The measurement of 800 MeV proton spallation cross sections has been an important activity of the Los Alamos Medical Radioisotope Research Group. Knowledge of the formation cross sections of isotopes of interest allows the calculation of estimated production yields in thick targets, the monitoring of hot cell procedures, and an evaluation of isotopic interferences in a particular LAMPF product. Because of the non-specific nature of nuclear spallation, for isotope production (11), chemical separation alone is sometimes insufficient for the requisite purity (12).

Thin targets of Ni, As, Pb, and Bi were irradiated with (800 ± 5) MeV protons to an integrated intensity of 1 microampere hr, together with Al monitor foils (12). Beam intensity measurements were based on the yields and known cross sections of the following activities in Al: ^{24}Na, 10.785 mb; ^{22}Na, 16.3 mb; and ^{7}Be, 6.4 mb. Radiochemical separations were performed on each target to recover elemental fractions of interest, and analyses of the various gamma photon spectra were performed using the GAMANAL code. The results are shown in Table 4.

The target utilized for ^{68}Ge production at LAMPF is RbBr, and a number of additional medically-interesting nuclides are also produced in this target when irradiated with medium energy protons (13). In this case, three pressed pellets of RbBr were irradiated simultaneously along with Al monitor foils. Following irradiation, two of the pellets were dissolved in HCl, aliquots withdrawn and mounted for counting, and the remainder of the solutions were chemically processed to isolate the Ge, As, and Se fractions. The third pellet was chemically processed to isolate the Cu and Ga fractions to permit independent determinations of the yields of ^{67}Cu and ^{67}Ga. The cross section results are presented in Table 5.

Vanadium targets are of interest as a potential source of ^{43}K, which also contains ^{42}K as an isotopic impurity (14), as well as a source of ^{44}Ti, which would serve as the parent in the ^{44}Ti-^{44}Sc biomedical generator system (15). Also this target would serve as a source of ^{32}Si that would be useful in a number of geologic and nuclear studies (16). The data in Table 6 are the results of our measurements (17).

One of the more important targets we have studied at LAMPF is molybdenum, for it is from this that the following useful nuclides are derived: ^{88}Zr, ^{88}Y, ^{83}Rb, ^{82}Sr, ^{77}Br, and ^{72}Se. Isolation of the Zr/Y fraction would produce a source material for an ^{88}Y-Be photoneutron source that would produce a monoenergetic source of 151 keV neutrons (18). Alternatively, an initial separation of the Zr and Y followed by the decay of ^{88}Zr would yield a source of isotopically pure ^{88}Y. ^{82}Sr ($T_{1/2}$ = 25.55 days) is the parent of 1.273-min. ^{82}Rb (19), which has been shown important in brain and heart studies employing positron emission tomographic (PET) imaging (20,21). Employing a relatively short irradiation time of five to six days, ^{77}Br can be made via spallation of Mo and a relatively high specific activity of 150,000 Ci/gm has been achieved (22). This material has proven to be useful in radiohalogen labeling research of estrogen derivatives (23) The cross section results (24) obtained for the spallation of molybdenum by 800 MeV protons are given in Table 7.

5. Spallation Yields at 800 MeV

Because of the combination of the LAMPF proton energy and large beam intensity (an intensity of 1.2 milliamperes has been achieved), it is possible for us to irradiate thick targets (about 2 cm thick) and to recover unusually large amounts of radioactive isotopes. In this section, I will briefly summarize a number of achievements that have been realized in this program.

Silicon-32 is the longest lived radioisotope of that element and is of interest to the astrophysical community as well as in other physical sciences. This nuclide was produced by irradiating 320 grams of V metal in the IPF during an intermittent bombardment over a period of three years (16), after which it was determined that about 1.5 mCi of ^{32}Si (possibly the world's supply) was made.

The decay properties of ^{67}Cu are suitable to make this nuclide attractive for applications in medical imaging and in internal radiation therapy, and extensive research is in progress on linking this nuclide to monoclonal antibodies to achieve tumor imaging and therapy (25). A target containing 79.2 g of ZnO was bombarded at a proton beam current of 423 microamperes for 93.4 hours, and the observed yield was 3.11 Ci of ^{67}Cu(26).

Space limitations will not allow even brief coverage of a number of other radioisotopes developed in this program. These include ^{7}Be, ^{22}Na, ^{26}Al, ^{44}Ti, ^{52}Fe (9,27), ^{68}Ge (28,29), ^{77}Br (9,30), ^{83}Rb, ^{82}Sr (31), ^{88}Y, ^{88}Zr, ^{109}Cd (27), ^{123}I (9,32), ^{127}Xe (29), and ^{172}Hf. The interested reader should refer to the reference citations for additional details.

One additional example will serve to emphasize the enormous production capacity at LAMPF. A tantalum target weighing a total of 770 g was irradiated over a four month period, with the irradiation terminating on November 18, 1980. During this period there were about 70 beam days with a proton current of approximately 350 microamperes. The estimated total proton fluence was ~10^{22} protons. The yields obtained, corrected to EOB, are shown in Table 8 and graphically illustrate the substantial resource capability of the LAMPF accelerator.

6. Acknowledgements

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Figure Captions

Figure 1   Effective spallation cross sections for niobium foils located in thick iron array (12" x 12" x 20") irradiated with 590 MeV protons.

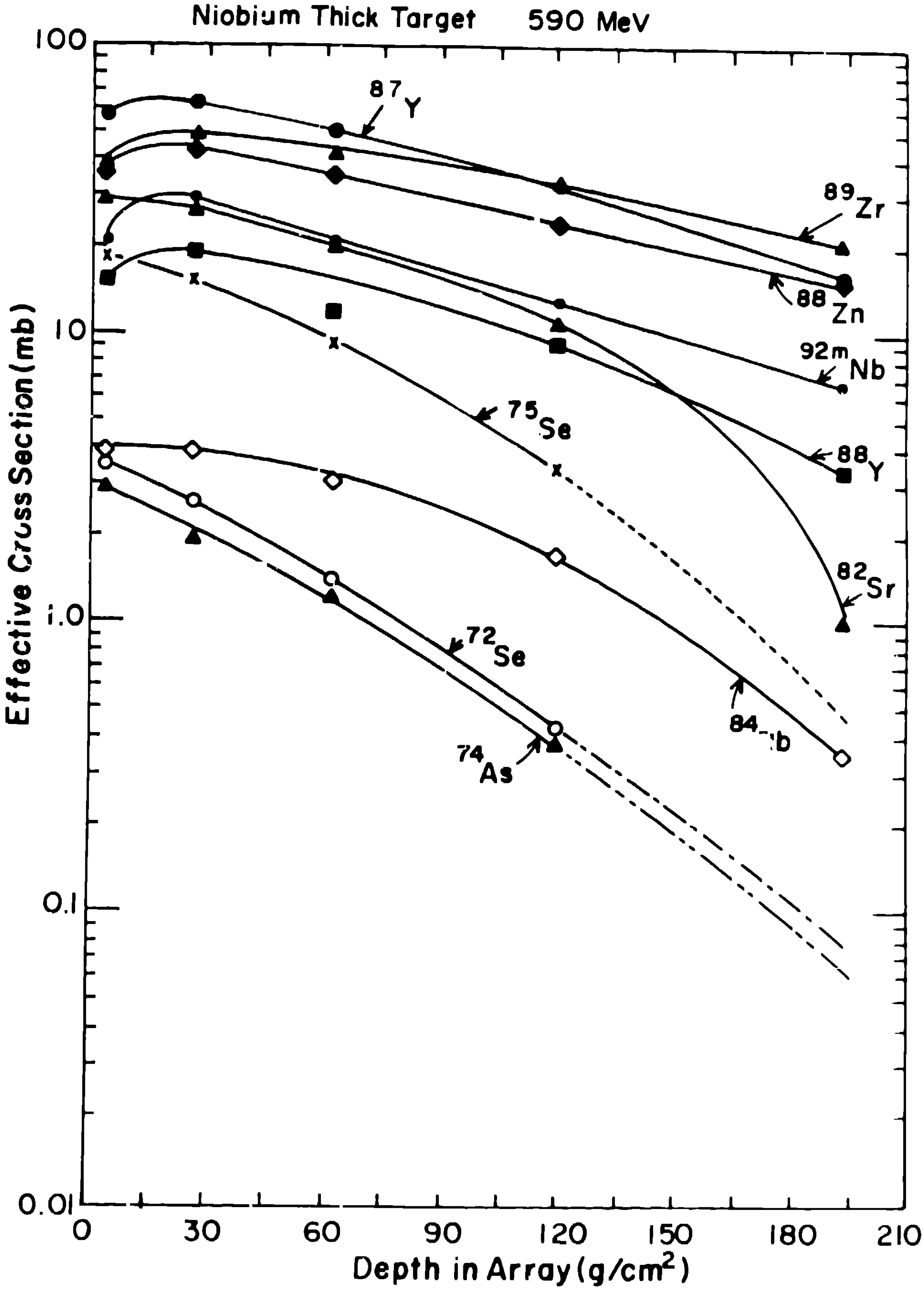
Niobium Thick Target 590 MeV
Effective Cross Section (mb)
Depth in Array (g/cm²)
100
10
1.0
0.1
0.01
0
30
60
90
120
150
180
210
87Y
89Zr
88Zn
92mNb
88Y
75Se
82Sr
72Se
84Rb
74As

Table 1. Spallation Yields From 590 MeV Protons on Titanium

Nuclide		Cross Section (mb) This Work	Cross Section (mb) Brodzinski, et al (2)
16-d	^{48}V	1.22 ± 0.40	1.02 ± 0.35
47-y	^{44}Ti	1.42 ± 0.40	0.62 ± 0.21
3.9-h	^{43}Sc	6.94 ± 1.00	-
3.9-h	^{44}Sc	16.8 ± 2.5	-
2.4-d	^{44m}Sc	7.30 ± 1.00	4.6 ± 1.5
34-d	^{46}Sc	29.5 ± 3.5	17.5 ± 5.6
3.4-d	^{47}Sc	22.9 ± 3.5	17.4 ± 5.6
1.8-d	^{48}Sc	2.8 ± 0.5	1.62 ± 0.52
4.5-d	^{47}Ca	0.27 ± 0.09	-
12.4-h	^{42}K	9.2 ± 1.2	6.0 ± 1.9
22.4-h	^{43}K	3.7 ± 0.5	2.11 ± 0.68
1.8-h	^{41}Ar	1.60 ± 0.60	
21-h	^{28}Mg	0.25 ± 0.08	0.100 ± 0.034
2.6-y	^{22}Na	0.86 ± 0.15	0.64 ± 0.21
15-h	^{24}Na	1.46 ± 0.21	0.87 ± 0.28
53-d	^{7}Be	2.4 ± 0.4	0.91 ± 0.32

Table 2. Spallation Yields From 590 MeV Protons on Iron

Nuclide		Cross Section (mb)		
		This Work	Brodzinski, et al (2)	Cline, et al (3)
18-h	^{55}Co	1.03	-	-
77-d	^{56}Co	1.1	3.3 ± 1.1	1.25 ± 0.20
271-d	^{57}Co	0.24	-	-
71-d	^{58}Co	0.005	-	-
8-h	^{52}Fe	2.05	-	0.80 ± 0.09
5.6-d	^{52}Mn	11.7	6.5 ± 2.0	37.9 ± 4.2
312-d	^{54}Mn	37.3	20.8 ± 6.7	55.1 ± 5.0
2.6-h	^{56}Mn	1.17	-	1.34 ± 0.25
23-h	^{48}Cr	0.93	0.55 ± 0.17	1.37 ± 0.30
28-d	^{51}Cr	51.3	29.6 ± 9.2	64.9 ± 7.0
16-d	^{48}V	22.7	13.7 ± 4.3	1.37 ± 0.15
3.9-h	^{43}Sc	4.56	-	10.4 ± 1.5
3.9-h	^{44}Sc	8.25	-	12.8 ± 1.4
2.4-d	^{44m}Sc	9.1	5.2 ± 1.6	9.00 ± 0.95
84-d	^{46}Sc	9.3	5.4 ± 1.8	11.7 ± 1.5
3.4-d	^{47}Sc	2.78	1.90 ± 0.58	3.26 ± 0.50
1.8-d	^{48}Sc	0.57	0.236 ± 0.076	1.05 ± 0.40
12 h	^{42}K	4.25	2.49 ± 0.78	4.82 ± 0.75
22 h	^{43}K	1.03	0.70 ± 0.23	1.21 ± 0.25
1.8 h	^{41}Ar	0.67		0.62 ± 0.07
2.6 y	^{22}Na	0.37	0.260 ± 0.097	
15 h	^{24}Na		2.66 ± 0.085	0.42 ± 0.05
53 d	^{7}Be	2.19		

Table 3. Spallation Yields From 590 MeV Protons on Copper

Nuclide		Cross Section (mb)	
		This Work	Cline, et al (3)
9.3-h	^{62}Zn	0.60	0.81 ± 1.0
3.3-h	^{61}Cu	17.5	20.7 ± 2.1
36-h	^{57}Ni	1.01	1.78 ± 0.24
5.3-y	^{60}Co	8.8	31.7 ± 6.0
72-d	^{58}Co	28.9	55.0 ± 5.5
271-d	^{57}Co	33.1	32.3 ± 3.0
77-d	^{56}Co	8.68	14.8 ± 1.5
18-h	^{55}Co	3.45	4.0 ± 0.5
45-d	^{59}Fe	1.30	2.54 ± 0.25
8.2-h	^{52}Fe	0.70	0.25 ± 0.03
2.6-h	^{56}Mn	3.62	5.5 ± 0.7
312 d	^{54}Mn	20.0	31.9 ± 3.2
5.7-d	^{52}Mn	9.09	12.0 ± 1.2
28 d	^{51}Cr	27.4	29.0 ± 3.0
23 h	^{48}Cr	0.52	0.51 ± 0.08
16-d	^{48}V	10.6	14.35 ± 2.00
1.8 d	^{48}Sc	0.60	0.30 ± 0.08
3.4 d	^{47}Sc	2.66	1.44 ± 0.25
84 d	^{46}Sc	4.78	3.2 ± 0.4
2.4 d	^{44m}Sc	4.23	1.8 ± 0.2
3.9 h	^{44}Sc	2.89	3.90 ± 0.40
3.9 h	^{43}Sc	3.97	0.95 ± 0.20
22 h	^{43}K	0.67	0.68 ± 0.10
12 h	^{42}K	2.29	1.52 ± 0.20

Table 4. Selected Spallation Cross Sections From 800 MeV Protons on Nickel, Arsenic, Bismuth, and Lead

Target	Chemical Yield	Isotope	σ (mb)	Type
Ni	0.9592	^{52}Fe	1.54 ± 0.13	C.Y.
		^{59}Fe	0.306 ± 0.048	C.Y.
	0.8577	^{43}Sc	7.02 ± 0.83	C.Y
		^{44m}Sc	7.34 ± 0.65	I.Y.
		^{46}Sc	5.69 ± 0.49	I.Y.
		^{47}Sc	1.49 ± 0.13	I.Y.+ C.Y.
		^{48}Sc	0.249 ± 0.022	I.Y.
As	0.4393	^{61}Cu	7.01 ± 0.68	C.Y.
		^{64}Cu	15.4 ± 1.6	I.Y.
		^{67}Cu	1.51 ± 0.13	C.Y.
	0.1237	^{66}Ga	11.6 ± 1.0	C.Y.
		^{67}Ga	28.4 ± 2.4	C.Y.
		^{72}Ga	3.16 ± 0.27	I.Y.+ C.Y.
Bi	0.2276	^{200}Tl	83.8 ± 8.8	I.Y.+ C.Y.
		^{201}Tl	58.5 ± 6.7	C.Y.
		^{202}Tl	6.56 ± 0.69	C.Y.
Pb	0.4934	^{200}Tl	67.8 ± 7.1	I.Y.+ C.Y.
		^{201}Tl	54.7 ± 5.9	C.Y.
		^{202}Tl	18.2 ± 1.9	C.Y.

I.Y. = Independent Yield

C.Y. = Cumulative Yield

Table 5. Spallation Cross Sections From 800 MeV Protons on RbBr

Nuclide	Cross Section (mb)	Type
^{88}Y	0.3 ± 0.2	I.Y.
^{85}Sr	4.3 ± 0.9	C.Y.
^{83}Sr	3.0 ± 0.5	C.Y.
^{82}Sr	2.1 ± 0.2	C.Y.
^{86}Rb	19 ± 1	C.Y.
^{84}Rb	50 ± 2	C.Y.
^{83}Rb	42 ± 1	C.Y.
^{82m}Rb	12 ± 2	I.Y.
^{81}Rb	14 ± 1	C.Y.
^{82}Br	4.9 ± 0.4	C.Y.
^{80m}Br	23 ± 2	I.Y.
^{77}Br	49 ± 2	C.Y.
^{75}Br	12 ± 6	C.Y.
^{75}Se	67 ± 2	C.Y.
^{73}Se	22 ± 1	C.Y.
^{72}Se	12 ± 0.4	C.Y.
^{76}As	14 ± 4	I.Y.
^{74}As	29 ± 0.9	I.Y.
^{71}As	27 ± 1	C.Y.
^{69}Ge	35 ± 2	C.Y.
^{68}Ge	19 ± 1	C.Y.
^{67}Ga	41 ± 1	C.Y.
^{65}Zn	39 ± 2	C.Y.
^{62}Zn	2 ± 1	C.Y.
^{67}Cu	1.6 ± 0.08	C.Y.
^{64}Cu	16 ± 0.9	I.Y.
^{61}Cu	7.8 ± 0.6	C.Y.
^{60}Co	9.1 ± 0.7	C.Y.
^{58}Co	18 ± 0.8	C.Y.
^{57}Co	13 ± 0.5	C.Y.
^{59}Fe	1.8 ± 0.1	C.Y.
^{56}Mn	3.0 ± 0.2	C.Y.
^{54}Mn	11 ± 0.4	I.Y.
^{52}Mn	2.7 ± 0.1	I.Y.
^{51}Cr	7.9 ± 0.5	C.Y.
^{48}V	2.3 ± 0.09	C.Y.
^{46}Sc	2.2 ± 0.1	C.Y.
^{44m}Sc	0.8 ± 0.2	I.Y.
^{44}Sc	0.6 ± 0.1	I.Y.
^{7}Be	4.1 ± 0.7	C.Y.

I.Y. – Independent yield
C.Y. – Cumulative yield

Table 6. Spallation Cross Sections From 800 MeV Protons on Vanadium

Nuclide	Cross Section (mb)	Type
^{52}Mn	0.015 ± 0.004*	C.Y.
^{51}Cr	2.2 ± 0.3	C.Y.
^{48}V	13 ± 0.4	C.Y.
^{48}Sc	6.9 ± 0.07	I.Y.
^{47}Sc	16 ± 0.7	C.Y.
^{46}Sc	25 ± 0.9	I.Y.
^{47}Ca	0.56 ± 0.02	C.Y.
^{43}K	5.3 ± 0.3	C.Y.
^{42}K	12 ± 0.4	I.Y.
^{28}Mg	0.38 ± 0.01	C.Y.
^{24}Na	2.3 ± 0.03	C.Y.
^{22}Na	1.1 ± 0.04	C.Y.
^{7}Be	2.7 ± 0.1	C.Y.

I.Y. - Independent yield
C.Y. - Cumulative yield
* Uncertainties are minimum experimental errors

Table 7. Spallation Cross Sections From 800 MeV Protons on Molybdenum

Nuclide	Cross Section (mb)	Type
^{96}Tc	1.8 ± 0.2	C.Y.
^{95m}Tc	0.56 ± 0.04	C.Y.
^{99}Mo	8.8 ± 0.2	C.Y.
^{92m}Nb	9.7 ± 0.1	I.Y.
^{91m}Nb	7.1 ± 0.6	C.Y.
^{89}Zr	51 ± 8	C.Y.
^{88}Zr	43 ± 0.2	C.Y.
^{87}Y	59 ± 1	C.Y.
^{85}Sr	48 ± 0.7	C.Y.
^{82}Sr	23 ± 1	C.Y.
^{86}Rb	1.4 ± 0.01	C.Y.
^{84}Rb	5.7 ± 0.05	C.Y.
^{83}Rb	46 ± 0.4	C.Y.
^{77}Br	27 ± 0.9	C.Y.
^{75}Se	26 ± 0.3	C.Y.
^{74}As	4.6 ± 0.04	I.Y.
^{71}As	12 ± 0.7	C.Y.
^{65}Zn	7.6 ± 0.2	C.Y.
^{58}Co	2.2 ± 0.1	C.Y.
^{57}Co	1.2 ± 0.1	C.Y.
^{56}Co	0.44 ± 0.02	C.Y.
^{59}Fe	0.18 ± 0.01	C.Y.
^{52}Mn	0.31 ± 0.01	C.Y.
^{51}Cr	0.75 ± 0.08	C.Y.
^{48}V	0.26 ± 0.01	C.Y.
^{46}Sc	0.21 ± 0.01	C.Y.

I.Y. – Independent yield
C.Y. – Cumulative yield

Table 8. Spallation Yields From 800 MeV Protons on Tantalum

Isotope	$t_{1/2}$	Curies	mgrams
^{172}Hf	1.87y	6.7	6.0
^{175}Hf	70d	58	5.4
$^{179m2}Hf$	25d	8.5	0.3
$^{178m2}Hf$	31y		0.04 (est.)
^{173}Lu	1.37y	10	6.6
^{174}Lu	3.3y	0.11	0.16
^{174m}Lu	142d	0.19	0.04
$^{177m\text{-}177}Lu$	161d(6.7d)	0.047	0.01
^{169}Yb	32d	28	1.2
^{170}Tm	129d	0.04	0.01
^{168}Tm	93d	0.38	0.05
^{167}Tm	9.3d	15	0.17
^{146}Gd	48d	2.4	0.13
^{148}Gd	75y	0.009	0.28
^{150}Gd	2×10^{6}y		
^{151}Gd	120d	1.5	0.21
^{153}Gd	242d	0.9	0.25
^{147}Eu	24d	1.81	0.049
^{148}Eu	55d	0.084	5.1×10^{-3}
^{149}Eu	93d	2.25	0.24
^{150}Eu	36y	2.1×10^{-4}	3.1×10^{-3}
^{145}Sm	340d	0.33	0.12
^{146}Sm	7×10^{7}y		
^{143}Pm	265d	0.10	0.03
^{144}Pm	363d	2.4×10^{-3}	9.5×10^{-4}
^{139}Ce	138d	0.032	4.7×10^{-3}