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THE RADIOISOTOPE PROGRAM AT LAMPF

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H. A. O'Brien, Jr.

Los Alamos Scientific Laboratory
Los Alamos, New Mexico 87544

I. Introduction

Accompanying the rapid proliferation of the uses of radioactive isotopes in research, diagnostic, and therapeutic applications in medicine has been a continuing search for new radionuclide preparations that are more amenable to the problems in this field. In addition to special requirements of radioactive half-lives and decay properties, increased attention has been given to radionuclides in an isotopically pure state (commonly called carrier-free radioisotopes) in order to permit in vivo studies without disturbing cellular chemistry, and to study the metabolic behavior of otherwise toxic trace elements in the body.

For the past several years we have been investigating proton-induced spallation reactions as a means of augmenting existing techniques for making radioisotopes. This work was initiated in anticipation of the Clinton P. Anderson Meson Physics Facility (LAMPF), a linear accelerator that will accelerate both H^+ and H^- ions to an energy of 800 MeV and have an average beam current of one milliampere[1]. The results of our studies clearly indicate that useful quantities of heretofore rare nuclides could be prepared by this techniques[2,3,4].

This report summarizes the progress of the work to date and the near-future plans for the radioisotope program at Los Alamos.

II. LAMPF Accelerator and Isotopes Production Facility

Many experiments, both in nuclear physics and nuclear chemistry, are planned to utilize the primary proton beam and also other secondary beams

of pions, muons, and neutrons, which are produced when the primary proton beam interacts with a suitable target. The experimental targets will share the primary and secondary beams simultaneously; even so, a sizable fraction of the primary proton beam is unused. It is estimated that between 30% to 50% of the initial beam intensity will reach the main beam stop.

Various target materials can be inserted into this excess beam near the beam-stop area to produce a broad spectrum of radionuclides, primarily neutron-deficient nuclides, through a complex series of nuclear interactions called spallation reactions. Extensive investigations of spallation reactions have been performed by personnel from this Laboratory at the Space Radiation Effects Laboratory in Virginia using the 600-MeV proton synchrocyclotron[4,5]. The spallation reaction data from these investigations have provided a firm basis for predicting radioisotope yields for the conditions at LAMPF.

A schematic view of the LAMPF accelerator (Figure 1) shows the Cockroft-Walton injector, the drift-tube accelerator, and the wave-guide accelerator. At the end of the accelerator is the beam switchyard (Figure 2), through which portions of the beam can be deflected to the high-resolution mass spectrometer (Area C), the nucleon-nucleon physics lab (Area B), and to the National Defense Neutron Research Area. The major portion of the proton beam will proceed through Area A, containing three major target cells, and continue on through the biomedical research facility, and, finally, terminate in the main beam stop area. The Isotope Production Facility will be located immediately in front of the main beam stop.

The Isotope Production Facility, which represents an addition of about 400 sq. ft. to the LAMPF main beam stop structure, is seen in Figure 3, which is a simplified line drawing of two views of this facility and the beam-stop area. Functionally, the Isotope Production Facility is designed to enable an operator to: 1) remotely attach a target-containing chamber to the end of a stringer (26 ft. long x 8 in. high x 2 in. wide); 2) drive the stringer through a slot in the stringer

housing in the shielding so that the target is in position to intercept the proton beam; 3) carry out the irradiation with adequate water cooling of the target; and, after an appropriate irradiation period, 4) retract the stringer, remove the target chamber, and transfer it to a shielded cask for transport to hot cell facilities located elsewhere in the laboratory. The facility will have an initial set of four independently-operable stringers, but contains, in addition, provisions for future expansion to twelve target stringers. It is expected the proton beam will reach this facility soon after January 1, 1974.

III. Initial Radioisotope Program

In preparation for the start-up of the Isotope Production Facility and in accord with the current interest in ^{82}Sr, ^{123}I, ^{125}I, and ^{127}Xe for nuclear medical applications, initial emphasis has been placed on the measurements of the yields of these nuclides or their precursors from several potential target materials and on the chemical recovery of these isotopes following irradiation[6]. The data presented in Table I are selected results of yield measurements obtained over a range of incident proton energies for several targets, and include results from irradiations performed both at LAMPF and at the Space Radiation Effects Laboratory in Virginia. The 590-MeV yield data was used to calculate projected isotope yields at LAMPF that will be discussed later.

The ^{82}Sr yields from zirconium, niobium, and molybdenum targets are essentially equivalent. Subsequently, molybdenum was chosen for use in the preparation of ^{82}Sr on the basis of anticipated ease of chemical procedures leading to the isolation of a strontium fraction. Using aged, proton-irradiated molybdenum targets, we have developed a five-step chemical procedure for the strontium recovery as shown in Figure 4[7]. The spallation-product chemical yield results from several experimental determinations are presented in Table II. These results demonstrate a large percentage strontium recovery and excellent decontamination. Plans are in progress to test this procedure on the hundred-gram scale.

TABLE I.

CUMULATIVE YIELDS OF SELECTED ISOTOPES OBTAINED FROM IRRADIATION OF LANTHANUM-COPPER, ZIRCONIUM, NIOBIUM, AND MOLYBDENUM WITH 100- TO 590-MeV PROTONS

Target	Isotope (Half-Life)	Yield, mb					
		100 MeV	211 MeV	302 MeV	496 MeV	590 MeV[a]	590 MeV
La 74 at. % - Cu 26 at. %	^{123}I (13.2 h)	N. D.	12 ± 2	37 ± 7	52 ± 9	57 ± 9	57 ± 6[b]
	^{123}Xe (2.1 h)	N. D.	9 ± 1	27 ± 5	33 ± 5	36 ± 5	45 ± 6[b]
	^{125}Xe (17.0 h)	N. D.	26 ± 4	55 ± 11	53 ± 9	57 ± 9	43 ± 9[b]
	^{127}Xe (36.4 d)	< 5	48 ± 10	60 ± 18	68 ± 13	53 ± 11	
	^{127}Cs (6.25 h)	--	44 ± 7	66 ± 13	47 ± 7	45 ± 7	59[c]
	^{129}Cs (32.1 h)	34 ± 7	73 ± 15	108 ± 32	78 ± 15	68 ± 14	35[c]
Zr	^{82}Sr (25 d)					19 ± 4	
	^{88}Y (107 d)					23 ± 3[d]	
Nb	^{82}Sr (25 d)					22 ± 4	
	^{88}Y (107 d)					19 ± 3[d]	
Mo	^{82}Sr (25 d)					15 ± 3	
	^{88}Y (107 d)					9 ± 1[d]	

N.D. = NOT DETECTED

a. From irradiations performed at the Space Radiation Effects Laboratory

b. Data from reference 8

c. Data from reference 9

d. Independent yield

TABLE II.

DISTRIBUTION OF RADIOISOTOPES DURING RADIOCHEMICAL SEPARATION OF STRONTIUM FROM PROTON-IRRADIATED MOLYBDENUM METAL TARGET

Step	Exper. Fraction	Overall Chemical Yield (%)
Original Solution	--	As, Se, Tc, Zr, Sr, Rb, Nb, Y, Zn, Co, Be (all 100%)
$PbMoO_4$	Filtrate	As (93%), Se (99%), Tc (89%), Zr (2%), Rb (98%), Nb (7%), Y (1%), Zn (66%), Co (97%)
	Precipitate	As (7%), Se (1%), Tc (11%), Zr (98%), Sr (100%), Rb (2%), Nb (93%), Y (99%), Zn (34%), Co (3%), Be (100%)
HDEHP	Organic	Zr (97%), Sr (3%), Nb (89%), Y (99%), Be (100%)
	Aqueous	As (7%), Se (1%), Tc (11%), Zr (1%), Sr (97%), Rb (2%), Nb (4%), Zn (34%), Co (3%)
Sulfide Pptn.	Precipitate	As (5%), Tc (11%), Zr (1%), Sr (1%), Nb (4%), Zn (9%), Co (3%)
	Filtrate	As (2%), Se (1%), Sr (96%), Rb (2%), Zn (25%)
HZO	Exchanger	As (2%), Se (1%), Sr (6%), Rb (1%), Zn (25%)
	Eluent	Sr (90%), Rb (<1%)

In addition, we will soon initiate experiments on the development of a Sr-Rb generator system, which would permit, following rapid separation, the use of the 75-sec ^{82}Rb in biological systems.

To prepare ^{123}I with a minimum of radiocontaminants via medium-energy proton-induced spallation reactions, the xenon fraction containing its 2.1-h parent, ^{123}Xe, must be separated from the target soon after the end of bombardment, collected and allowed to decay to 13.2-h ^{123}I for a suitable time, and then removed from the collection vessel. Using lanthanum (74%)-copper (26%) eutectic targets, we have examined the rate of xenon release under a variety of conditions and have determined that at 1400°C over 98% of the xenon can be removed in 15 min by cryopumping (pressure $<1 \times 10^{-4}$ torr). This process is being scaled for operation in hot cells and installation of a 100-kW motor generator plus ancillary equipment to power the needed heavy-duty furnace is proceeding. This equipment is expected to be operational by March, 1974.

Several methods of preparing ^{43}K from proton-induced spallation reactions at LAMPF have been examined. The cross sections for the formation of ^{42}K and ^{43}K from vanadium bombarded with 590-MeV protons are measured to be 8.9 mb and 3.8 mb, respectively. These values were used to calculate the ^{42}K and ^{43}K yield data in Table III, which illustrates that substantial quantities of ^{43}K, with a ^{42}K content of 10-12% or less, could be made by this technique.

Alternatively, an emanating vanadium target might be used with an on-line, continuous, gas-sweeping system designed to collect the 5.4-min ^{43}Ar in a cold trap and subsequently to recover the ^{43}K daughter. Since all the other radioisotopes of argon, except ^{42}Ar, decay to stable products and ^{42}Ar has a 33-year half-life, the only radiocontaminant would be a small amount of ^{42}K. The yields of ^{43}K and ^{42}K, assuming 100% emanating efficiency and argon recovery, are shown in Table IV. Our measured ^{42}Ar cross section of 0.61 mb and an assumed cross section of 0.15 mb for the ^{43}Ar were used in these calculations.

A study to measure the yields of cesium and barium nuclides from lanthanum irradiated with 590-MeV protons has not been completed; however,

TABLE III.

YIELDS OF ^{43}K AND ^{42}K FROM VANADIUM

(Assume: E_p = 600 MeV; ϕ = 0.5 mA; 1" thick target)

		Irradiation Time			
		1d	2d	3d	4d
EOB	43 Act (Ci)	45.4	66.8	76.9	81.7
	42 Act (Ci)	149	188	198	200
	43 Act (Ci)	1.07	1.58	1.81	1.93
EOB+5d	42 Act (Ci)	0.178	0.225	0.237	0.239
	% 42/43	16.6	14.2	13.1	12.4
	43 Act (Ci)	0.506	0.745	0.858	0.911
EOB+6d	42 Act (Ci)	0.0463	0.0585	0.0616	0.0622
	% 42/43	9.2	7.9	7.2	6.8
	43 Act (Ci)	0.239	0.352	0.405	0.431
EOB+7d	42 Act (Ci)	0.0121	0.0152	0.0160	0.0162
	% 42/43	5.1	4.3	4.0	3.8

TABLE IV.

YIELDS OF ^{43}K AND ^{42}K FROM DECAY OF ^{43}Ar AND ^{42}Ar PRODUCED IN VANADIUM

(Assume: E_p = 600 MeV; ϕ = 0.5 mA; 1" thick target)

Irradiation Time	^{43}K (Ci)	^{42}K (m Ci)	^{42}K in ^{43}K (%)
1d	1.20	0.24	0.020
2d	1.78	0.69	0.039
3d	2.04	1.2	0.059
4d	2.16	1.7	0.079

it should be possible to prepare a ^{129}Cs product, with a radionuclidic purity sufficient for medical applications, from the decay of 2.2-h ^{129}Ba. By allowing some five hours to elapse between the end of a lanthanum target irradiation and the chemical recovery of the barium fraction therefrom, many of the short-lived barium nuclides and their precursors would be eliminated, leaving only 1.62-h ^{126}Ba, 2.46-d ^{128}Ba, 2.1-h ^{129m}Ba, 2.2-h ^{129}Ba, and 11.7-d ^{131}Ba to decay to radioactive cesium nuclides. Both 1.64-min ^{126}Cs and 3.8-min ^{128}Cs rapidly decay to stable xenon nuclides. Thus, the 9.69-d ^{131}Cs, which decays to stable ^{131}Xe solely by electron capture without the emission of a gamma ray, would be present as a radiocontaminant; and the amount present could be regulated to some extent by varying the time allowed for the growth of ^{129}Cs before separating the cesium from the barium.

A lack of available information on the decay scheme of ^{129}Ba has hampered our efforts to measure the cross section for its formation from lanthanum. An assumed cross section of 40 mb was used to calculate ^{129}Ba yields, and subsequent ^{129}Cs yields, from lanthanum.

The proton beam is expected to reach the main beam stop in early January, 1974, with an initial intensity of 1 μA or less. During the next six months, the beam intensity should increase to 10 μA, and will gradually be increased thereafter until it reaches 100 μA by January, 1975. In anticipation of this schedule, we have prepared two tables showing expected yields for ^{82}Sr, ^{123}I, ^{125}I, ^{129}Cs, and ^{127}Xe from LAMPF assuming beam intensities of 10 μA and 500 μA, and varying irradiation and processing periods. The yields shown in Table V, which should be available sometime between January and July, 1974, would provide adequate quantities of these nuclides for preliminary evaluation. At full LAMPF operation, the yields shown in Table VI ought to be realized.

IV. Summary

At this point in time, we can summarize the status of the Los Alamos Radioisotope Program as follows. On the basis of our cross section and yield measurements, we can project, with reasonable assurance, yields of

TABLE V.

NEAR-TERM ISOTOPE YIELDS FROM LAMPF

(Jan. - July 1974)

(Assume: Target 1.5 in. thick; E_p = 600 MeV; ϕ = 10 μA)

Target	Isotope	Bomb't Time	Yield (mCi)	Comments
La	13-h ^{123}I	2 h	120	2 h proc. + 2 h decay
La	36-d ^{127}Xe	48 h	315	Allow 114 h decay for ^{125}I recovery
La	32-h ^{129}Cs	4.4 h	63	5 h proc. + 9.1 h decay of ^{129}Ba parent
Mo	25-d ^{82}Sr	48 h	332	
La	60-d ^{125}I	34 h	70	4 h proc. + 110 h decay
La	60-d ^{125}I	48 h	81	4 h proc. + 110 h decay

TABLE VI.

LONG-RANGE ISOTOPE YIELDS FROM LAMPF

(Target: 1.5 in. thick; Ep = 600 MeV; Ø = 0.5 mA)

Target	Isotope	Bomb't Time	Yield Ci	Comments
La	13-h ^{123}I	2 h	6.0	2 h proc. + 2 h decay
La	36-d ^{127}Xe	36 d	228 Ci	209 Ci after 110 h decay for ^{125}I recovery
La	32-h ^{129}Cs	4.4 h	3.0 Ci	5 h proc. + 9.1 h decay of ^{129}Ba parent
Mo	25-d ^{82}Sr	25 d	154 Ci	
La	60-d ^{125}I	36 d	4.7 Ci	Recovered from ^{127}Xe run 4 h sep'n + 110 h decay
La	60-d ^{125}I	2 h	0.34 Ci	Recovered from ^{123}I run 4 h sep'n + 110 h decay
La	60-d ^{125}I	34 h	3.8 Ci	Optimize for ^{125}I

^{82}Sr, ^{123}I, ^{125}I, ^{129}Cs, and ^{127}Xe from LAMPF. The development of recovery procedures for each of these isotopes, except ^{129}Cs, has been completed to the extent that our attention is now being focused on scaling these procedures to hot cell operations.

Although the basic Isotope Production Facility addition to the beam stop is complete, a major effort will be required for the installation and check-out of the mechanical, instrumentation, and control systems in the facility prior to January, 1974.

The first useful quantities of radioisotopes from LAMPF should be available sometime during the first half of calendar-year 1974.

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FIGURE CAPTIONS

Figure 1. A Schematic View of LAMPF

Figure 2. A Schematic View of the LAMPF Experimental Area

Figure 3. Sectioned Plan and Elevation Views of the LAMPF Beam Stop Area and Isotope Production Facility

Figure 4. Molybdenum Target Chemical Separation Procedure

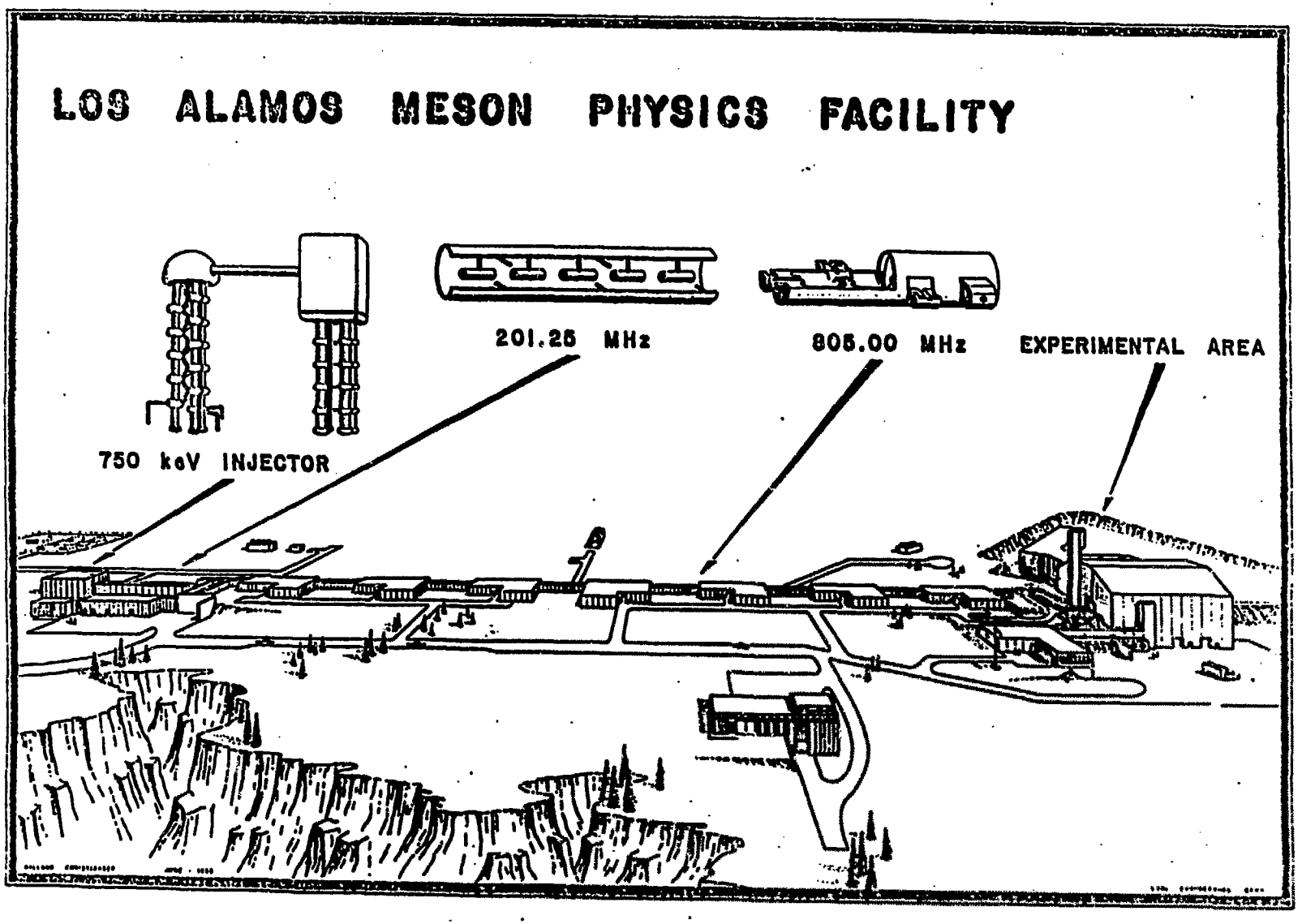

LOS ALAMOS MESON PHYSICS FACILITY
201.25 MHz
805.00 MHz
EXPERIMENTAL AREA
750 keV INJECTOR

Figure 1.

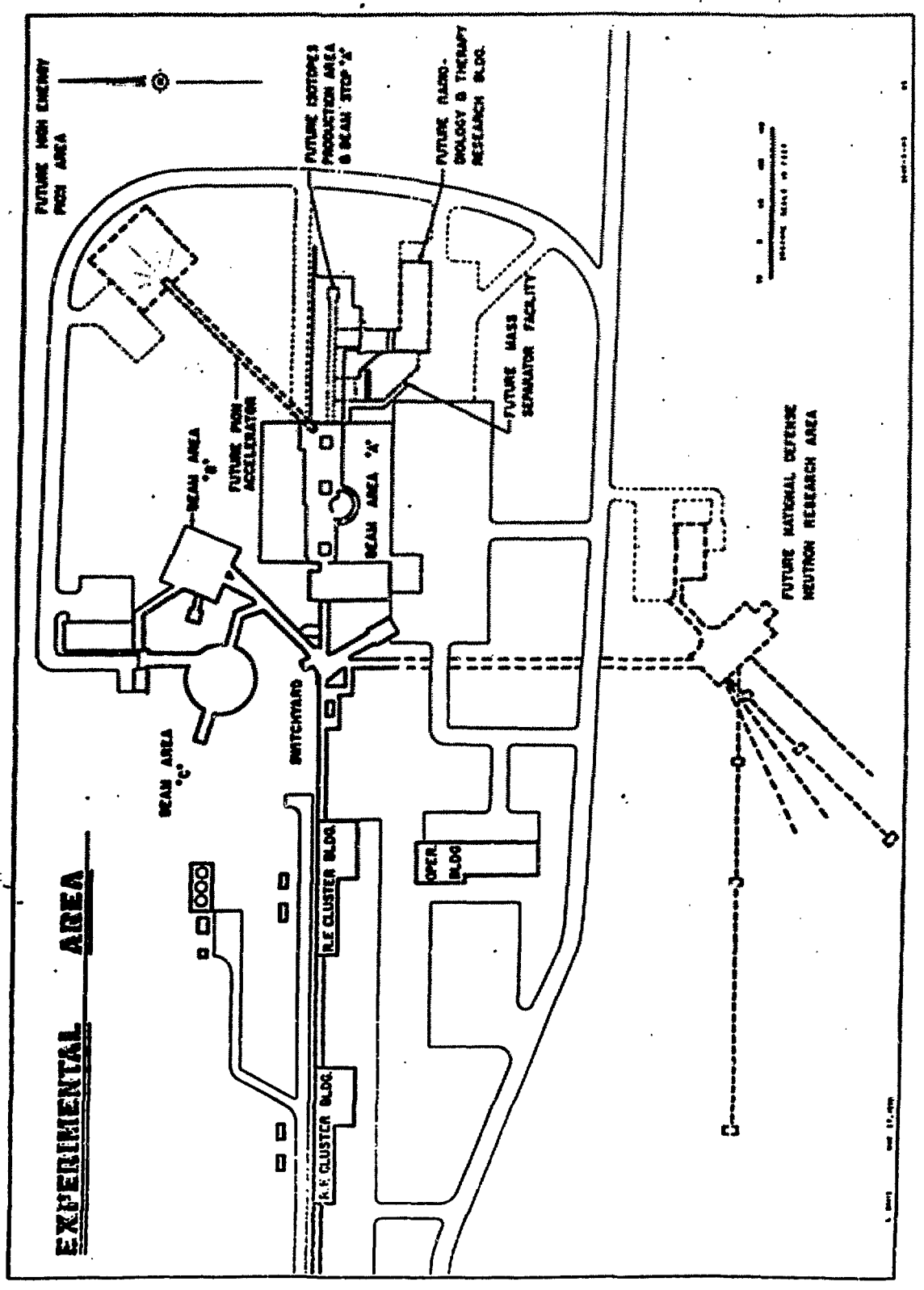

EXPERIMENTAL AREA
FUTURE HIGH ENERGY PION AREA
FUTURE ISOTOPES PRODUCTION AREA & BEAM STOP "A"
FUTURE RADIO-BIOLOGY & THERAPY RESEARCH BLDG.
FUTURE MASS SEPARATOR FACILITY
FUTURE PION ACCELERATOR
BEAM AREA "B"
BEAM AREA "A"
BEAM AREA "C"
SWITCHYARD
L.E. CLUSTER BLDG.
H.E. CLUSTER BLDG.
OPER. BLDG.
FUTURE NATIONAL DEFENSE NEUTRON RESEARCH AREA

Figure 2.

1. DISSOLUTION

$$\text{Mo Target} \xrightarrow{30\%\ H_2O_2} H_2MoO_4$$

2. PRECIPITATION

$$H_2MoO_4 \xrightarrow[Pb^{2+}]{\text{basic}} PbMoO_4\downarrow$$

3. SOLVENT EXTRACTION

$$PbMoO_4 \xrightarrow[50\%\ \text{HDEHP in toluene}]{0.1\ M\ HCl} \begin{cases} \xrightarrow{\text{organic}} Y \\ \xrightarrow{\text{aqueous}} Sr \end{cases}$$

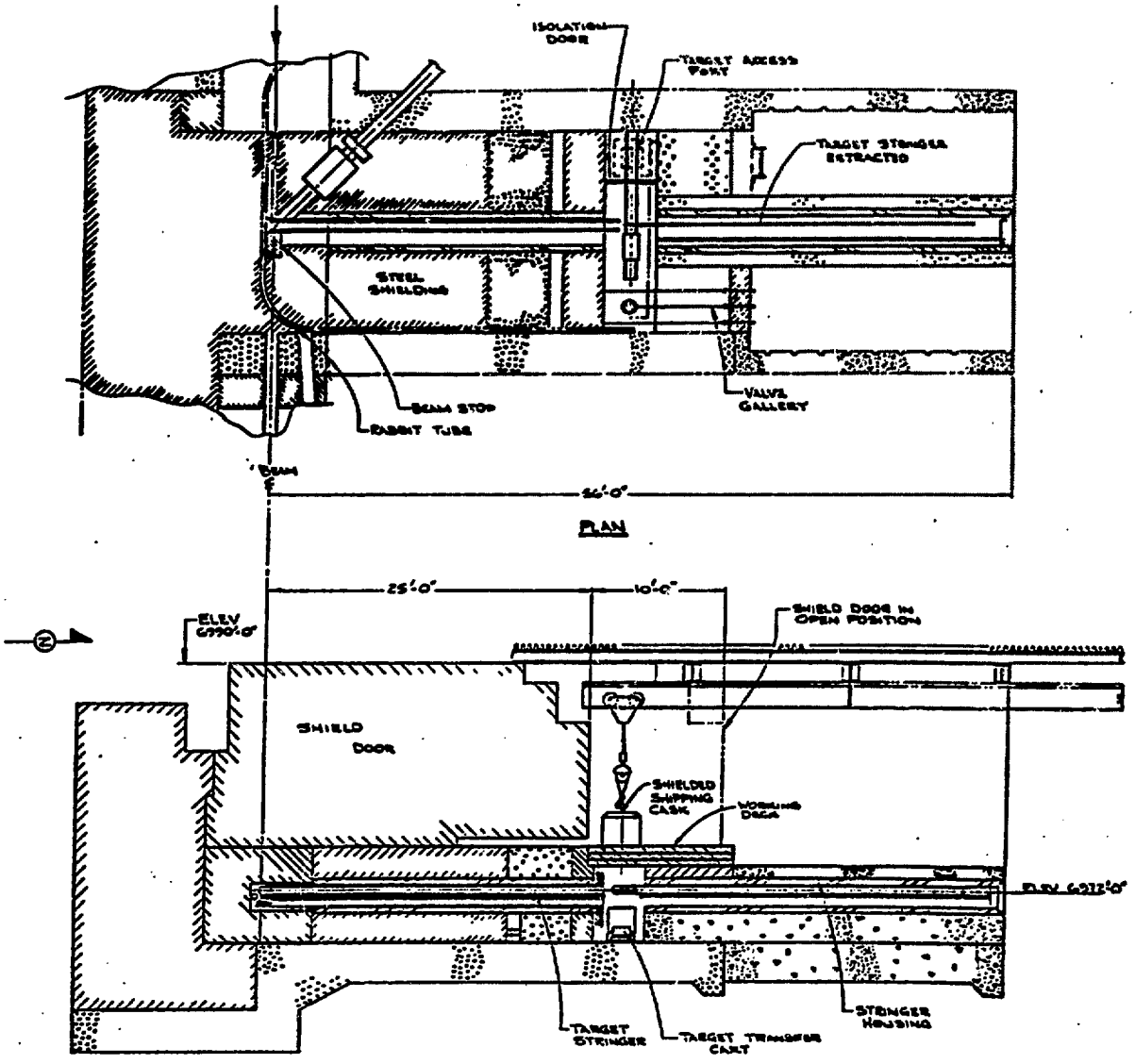
4. SULFIDE PRECIPITATION

$$\text{Aqueous Solution} \xrightarrow[H_2S]{\text{basic}} PbS\downarrow + MoS_4^{=} \xrightarrow{\text{acidic}} MoS_3\downarrow$$

5. ION EXCHANGE

$$\text{Sulfide Filtrate (+ Sr carrier)} \xrightarrow[pH = 5]{HZO} Sr$$

Figure 4.



ISOTOPE PRODUCTION FACILITY

Figure 3.