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AN INFORMAL REPORT

Use of LAMPF for Isotope Production. Briefing to the AEC Division of Isotopes Development December 15, 1970

los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87544

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by

L. Rosen
M. E. Schillaci
B. J. Dropesky
H. A. O'Brien

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Presentation 1

GENERAL INTRODUCTORY REMARKS CONCERNING LAMPF AND ITS USE FOR ISOTOPE PRODUCTION

By

L. Rosen

We are pleased to have the opportunity this morning to demonstrate to you that the time has come when serious attention ought to be devoted to LAMPF as a source of valuable radioisotopes.

With me today are Dr. Mario Schillaci, who will discuss the production rates available from LAMPF, Dr. Bruce Dropesky, who will describe the proposed targeting system, hot cell and radiochemistry facilities available at LASL, and Dr. Hal O'Brien, who will tell you some of the markets now foreseeable for those radioisotopes which can be produced at LAMPF more abundantly than with any other production facility presently in existence or under construction.

Up to now, all of the effort devoted by LASL to explore the desirability and feasibility of an isotope production facility at LAMPF has been supported either by AMA or by the Division of Research. However, budgetary stringency in FY-72 will severely curtail this support and, in view of the obvious relation of this activity to DID missions and responsibilities, I would suggest that it is now appropriate for DID to pay some attention to LAMPF.

I realize, of course, that DID budgets are not enormous. I believe, in fact, that the national interest demands that they be substantially increased. The way to do this is by not only vigorously supporting those activities which are of proven value but also by exploring new activities which offer unusual promise, by exercising the kind of imagination, creativity, inventiveness and daring which has made our nuclear science and technology endeavor the envy of the world. Thus, with the support of the DID for this proposal, we can assist in obtaining additional funds for an isotope production facility at LAMPF.

As most of you know, we are constructing at Los Alamos, the world's most prolific pion-producing accelerator. It goes by the name of the Los Alamos Meson Physics Facility (LAMPF). As a corollary, it can be stated that the primary proton beam will carry more power than will the beam of any high-energy accelerator in the world now in operation or under construction. It can furthermore be stated that most of this beam power will be wasted if it is not used to produce isotopes. The actual production of isotopes will cost the nation nothing-it is a by-product of LAMPF operation. The reason we are here is to convince you that you ought to take the trouble not to engage in sowing the harvest, or even growing it - just reaping it. It is required that target facilities for isotope production be built, that techniques be developed for extracting the desired isotopes, for purifying them, for packaging them, and for selling them either for evaluation or use. Facilities for chemical processing already exist and are available at LASL. Thus, we are not requesting funds for a separation plant (which DID may wish to fund in the future, if warranted by demand), but solely for the irradiation facility. Last year, in testimony before the JCAE, I stated that this facility would cost less that $0.5M, and this estimate has not changed.

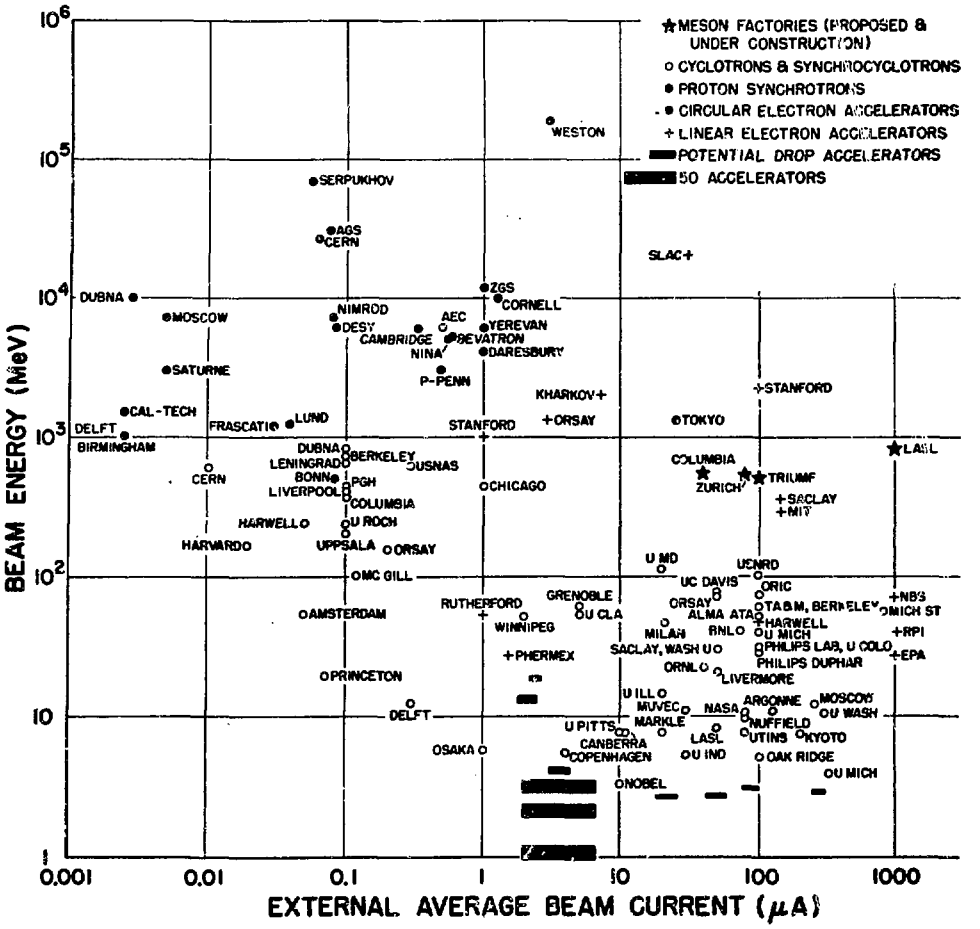

Fig. 1. World-wide accelerator picture.

In order to place the opportunity presented to you in proper context, I show you Fig. 1. It depicts the world-wide accelerator picture. The stars represent a new family of accelerators now under construction - they are called meson factories. The one in Switzerland and also the one in Canada will be operated to produce radioisotopes as a by-product. However, as you can see, LAMPF is the most powerful of the lot. It can produce not only more radioisotopes than all the others put together, but it can produce them further from the stability line, thus opening the possibility of using many more targets for the production of a given isotope. This is important for reasons of physical properties of the target, target material availability and purity of the desired isotope. I'm sure you gentlemen need not be told how foolish all of us will appear if several years from now the United States is buying proton-rich isotopes from Switzerland or Canada as a result of our proposal not being funded.

LAMPF is based on a proton linac of energy 800 MeV and average intensity 1 mA, resulting in a beam carrying 800 kW of power. The project is going very well. It is on schedule time-wise and budget-wise. Figure 2 shows the intermediate goals for LAMPF. Full energy beam is expected in the switchyard during July of 1972. We can start producing radioisotopes in January 1973. Some indication of our construction progress can be garnered from a quick perusal of Fig. 3. Figure 4 is an aerial view showing the Lab-Office Building, the Injector Building, the equipment gallery and the Operations Building. The blisters along the equipment aisle house the power supplies for the high-

INTERMEDIATE GOALS FOR LAMPF

EVENT DESCRIPTIONS	SCHEDULED DATE
START ASSEMBLY OF COCKCROFT-WALTON B	10/30/70
START CONST OF PH II EXP AREA	11/09/70
START CHECKOUT SEC B RF POW SYS (FL)	11/13/70
START CONST NUCLEAR CHEMISTRY WING	11/30/70
START OPR INJ A + TRNS AR W/CCS	12/15/70
START CONST OF PH III EXP AREA	1/15/71
START INSTLN OF SWITCHYARD BLA EQUIP	4/01/71
START INSTLN OF SWITCHYARD LX EQUIP	4/01/71
CONST OF AREA A BUILDING COMPLETED	4/06/71
START INTG MOD 3 & 4 W/CCS	4/15/71
*100 MEV BEAM DAY	7/01/71
START INTG OPRN MODS 1-12 FROM CCR	8/13/71
START OPRN CHECKOUT H-MINUS INJECTOR	10/01/71
*211 MEV BEAM DAY	10/01/71
CONST OF NUCLEAR CHEMISTRY WING COMP	11/30/71
COMPUTER ON LINE 24 HR/DAY	12/30/71
SECTOR G I & C COMPLETED	12/30/71
CONST OF AREA A SWITCHYARD CORR COMP	1/10/72
B.O.D. ON AREA C BUILDING	3/21/72
SWITCHYARD (BLA & LX) READY FOR BEAM	5/01/72
CONST EXP AREA PH III COMPLETED	5/18/72
PH II EXP AREA CONST COMPLETED	6/14/72
*STRT 800 MEV BM TESTS (BEAM DAY)+	6/30/72
STRT OPRNL CKOT H^- BM + H^- AND H^+ BM	10/01/72
BEAM AREA A FACILITY OPERATIONAL	12/29/72
HRS READY TO ACCEPT BEAM	1/03/73

Fig. 2. Intermediate goals for LAMPF

CONSTRUCTION COST STATUS FOR LAMPF AS OF OCTOBER 1, 1970

	Cost Incurred to Date	Outstanding Commitments	Estimate to Complete	Current Working Estimate	Anticipated Overrun (Underrun)
Design of Buildings and Accelerator	$ 5,270,000	1,004,000	300,000	6,574,000	
Buildings and Structures	12,587,000	1,545,000	6,287,000	20,419,000	5,279,000
Equipment Procurement and Installation	10,900,000	3,900,000	12,382,000	27,082,000	209,000
Utility, Roads, and Utilities	1,632,000			1,632,000	
Contingency			293,000	293,000	(5,409,000)
Totals	30,289,000	6,449,000	19,262,000	56,000,000	

Fig. 3. Construction cost status for LAMPF as of October 1, 1970

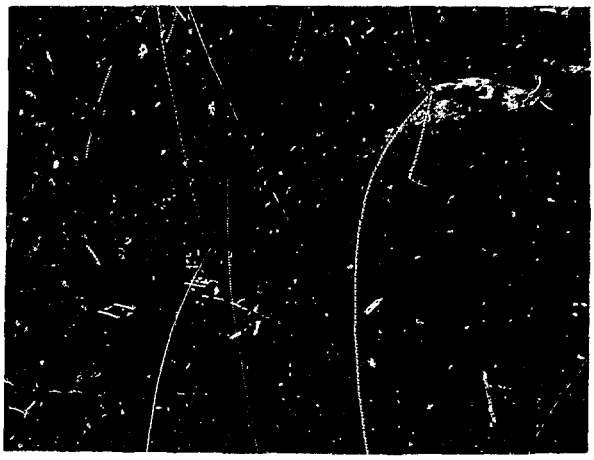

Fig. 4. Aerial view of LAMPF site.

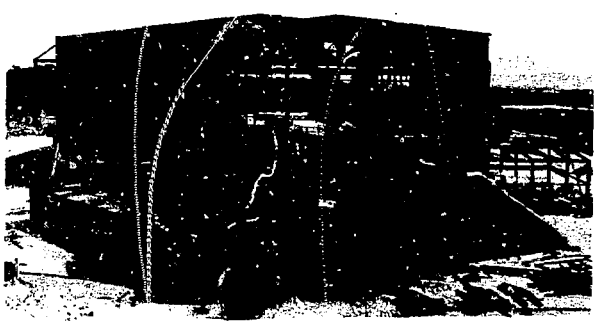

Fig. 5. Construction of main experimental area.

energy portion of the accelerator. Figure 5 depicts the main experimental area, looking west to east. In the foreground is the beam switchyard. To the south will be a pulsed neutron facility for weapons-related work. To the north will be a high-energy neutron facility and a high-resolution proton spectrometer for proton-nucleus reaction studies. Two hundred feet to the east will be a biomedical facility and immediately beyond that an isotope production facility. The accelerator comprises three stages shown in Fig. 6. There is the inevitable Cockroft-Walton, a drift-tube accelerator, and then a waveguide accelerator which takes the beam from 100 to 800 MeV. This latter was invented for the purpose at hand. It is already being installed in a half-mile tunnel, 25 feet underground (Fig. 7.) To prove to you that we are anxious to achieve practical applications in all of our activities, I show you Fig. 8. The accelerating structure used in the high-energy portion of the accelerator is already in use to serve humanity. Three companies, Varian, Arco and SHM Nuclear, have adapted the structures we developed for manufacture of reliable, inexpensive, high-energy X-ray machines. These use small electron linacs, 4 MeV and higher energy, to provide X-rays of better therapeutic properties than are available from cobalt or lower-energy machines. Already more than 50 of these machines have either been constructed or are in the process of construction.

Figure 9 is a schematic of the injector end of the accelerator. There will be three injectors: one for positive ions, one for negative ions, and

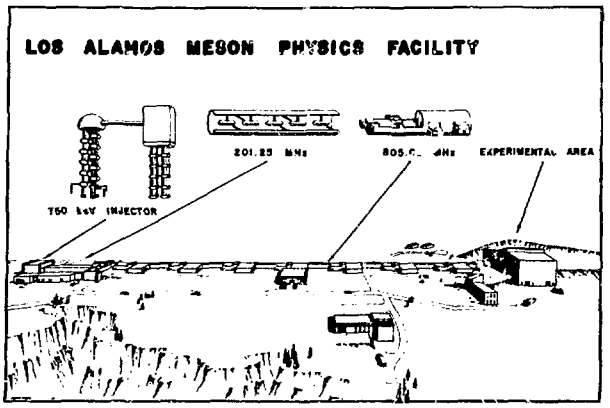

Fig. 6. The LAMPF accelerator system.

Fig. 7. Accelerator installation in beam channel.

one for polarized ions. Positive and negative ions will be accelerated simultaneously to be sorted out in the switchyard, thus doubling the duty factor of the facility.

Figure 10 is a schematic of the beam switchyard and experimental areas. It reveals the basic philosophy underlying the design of this facility - a capability to accommodate many uses simultaneously and with minimal interference of one with another.

Let's turn now to the need for an isotope production facility. Figure 11 illustrates the uses of radionuclides in medicine during 1966. There were, during that year, more than five million administrations involving more than three million patients. It is safe to say that these numbers are considerably higher today. It is also safe to say that many

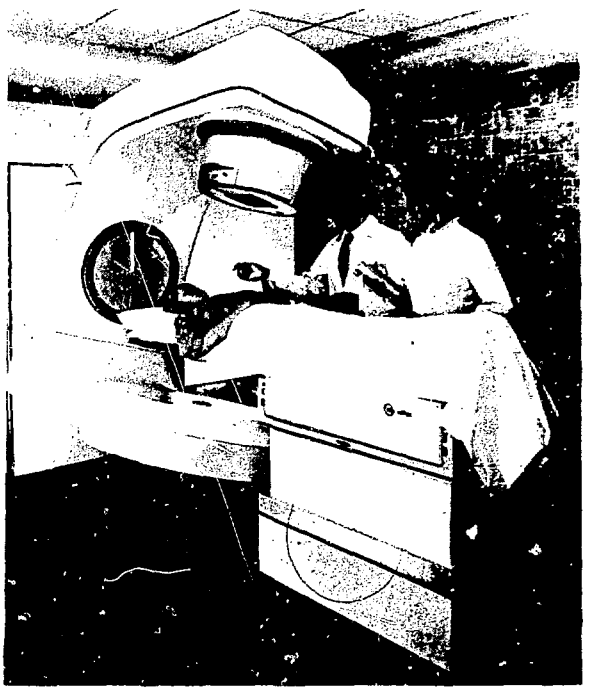

Fig. 8. High-energy X-ray machine used in treatment of cancers.

of the isotopes used have better and cheaper competitors which will be produced by LAMPF. It is also safe to predict that many of the proton-rich isotopes, producible by LAMPF, will be found to have exquisite applications in industry, agriculture, and especially in medicine.

A foreseeable practical application of LAMPF which is now receiving heavy attention has to do with the use of negative pions in radiotherapy. These particles offer promise of providing the best foreseeable radiation for the therapeutic treatment of certain classes of cancer, because of the possibility of achieving local deposition of high LET radiation under carefully controlled conditions and with visibility of the tissue volume where energy is being deposited. However, one must know where the tumor is and, for this knowledge, one needs recourse to radionuclides which concentrate in the tumor. This is now well recognized and a search is under way for tumor-seeking isotopes. The newly established LAMPF Biomedical Steering Committee has designated a subcommittee for

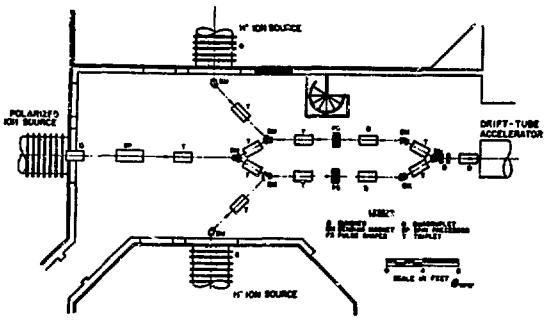

Fig. 9. Schematic of injector end of the accelerator.

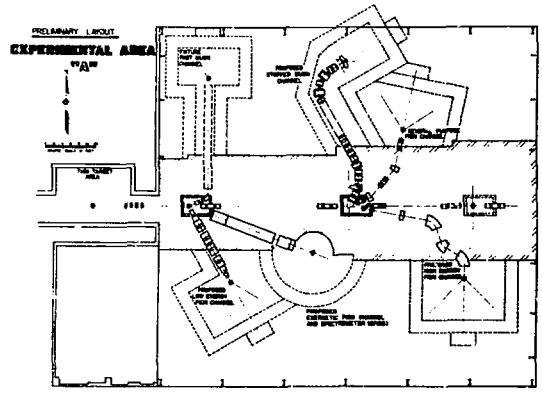

Fig. 10. Preliminary experimental area layout - (including switchyard).

PRINCIPAL NUCLEAR MEDICAL FUNCTION PROCEDURES PERFORMED DURING 1966

Radionuclide	Compound	Procedure	Physicians licensed for procedure	Physicians performing procedure	Patient administrations Number	Percent
^{131}I	Sodium iodide	Thyroid uptake	1,527	1,884	301,052	57.2
$^{131}I^{*}$	Labeled albumin	Blood volume determination	1,381	908	101,994	19.4
$^{131}I^{*}$	Sodium iodohippurate	Renal function	617	388	33,845	6.5
^{57}Co	Labeled vitamin B_{12}	Vitamin B_{12} absorption	952	582	28,996	4.7
^{51}Cr	Sodium chromate	Blood volume determination	1,064	437	22,468	4.3
^{60}Co	Labeled vitamin B_{12}	Vitamin B_{12} absorption	1,149	556	16,466	3.1
$^{131}I^{*}$	Labeled fats	Fat malabsorption	1,054	503	7,748	1.5
^{51}Cr	Sodium chromate	RBC survival	1,110	604	6,330	1.2
^{59}Fe	Chloride or citrate	Iron turnover	835	887	3,139	0.6
$^{131}I^{*}$	Labeled albumin	Cardiac output	687	79	1,503	0.3
$^{131}I^{*}$	Rose bengal	Hepatic function	33	23	1,808	0.2
Total for principal procedures listed above					520,357	98.8
Total for 39 other procedures with less than 1,000 administrations					6,119	1.2
Total for all function procedures					526,476	100.0

*Administrations using either ^{131}I or ^{125}I
Ref: Survey of the Use of Radionuclides in Medicine: Preliminary Report, U.S. Dept. of Health, Education and Welfare, NCRP 68-10, July 1968

Fig. 11. Principal nuclear medical function procedures performed during 1966.

Isotopes and Diagnostic Applications, (Fig. 12.) LAMPF can be an important source of these isotopes.

If radioisotope utilization continues to increase as we learn how to use those available and

BIOMEDICAL STEERING COMMITTEE

Chairman: Dr. Chaim Richman, Univ. of Texas at Dallas
Alternate: Dr. W. H. Langham, LASL
Assistant: Dr. D. E. Groce, JRB Associates

Subcommittee Chairmen:

Isotopes & Diagnostic Applications - J. D. Shoop, M. D., Univ. of New Mexico School of Medicine
Cellular Radiation Biology - Dr. P. Todd, Pennsylvania State University
Radiation Therapy - M. L. M. Boone, M.D., Univ. of Wisconsin Medical Center
Facility & Beam Line: P. R. Franke, Jr., and Dr. R. L. Hutson, LASL
Whole-Animal Radiation, Biology & Pathology - C. R. Kay, M.D., Univ. of New Mexico School of Medicine
Physical & Biological Dosimetry - Dr. M. R. Raju, Univ. of Texas at Dallas

Members at Large:

M. M. Kilgerman, M.D., Yale Univ. School of Medicine
R. E. Anderson, M.D., Univ. of New Mexico School of Medicine
Dr. C. A. Tobias, Univ. of California at Berkeley
J. R. Castro, M.D., M. D. Anderson Hospital
Dr. A. M. Koehler, Harvard University
David Hussey, M.D., M. D. Anderson Hospital

Fig. 12. Composition of Biomedical Steering Committee.

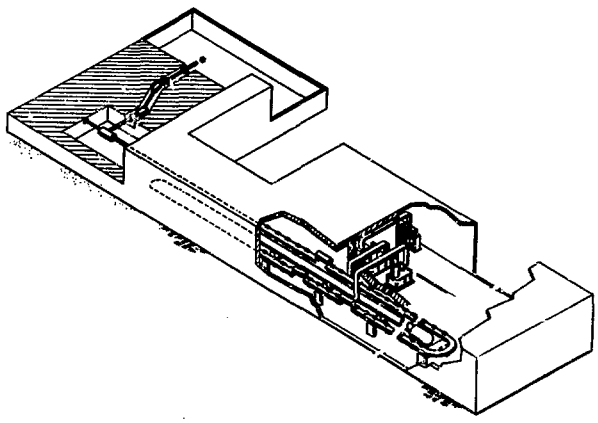

Fig. 13. Design of proposed medical proton linac.

find new uses for those yet to be produced in adequate quantities, we will need to look for production capabilities even larger than those which LAMPF can provide. We will also need a number of facilities to implement radiotherapy possibilities based on pions. These special-purpose facilities must, however, be far less expensive than LAMPF. We have given thought to this problem. Last spring, when I testified at hearings of the JCAE, I already could say that a straightforward extrapolation of LAMPF to a single-purpose, 500 MeV facility would decrease the cost to $10M. More recently, Dr. Darragh Nagle has given some serious, innovative

thought to this problem and has devised a scheme for cutting the above cost in half. His proposed accelerator is shown in Fig. 13. It starts with a 3 MeV pressurized injector, proceeds to a drift-tube, post-coupled linac at 400 MHz and into a side-coupled linac operating at 1200 MHz. By folding the line with three 180 degree bends one has a much more compact accelerator, more easily built and maintained.

I have repeatedly stated my conviction that both the Brookhaven and Los Alamos facilities should be supported. However, if because of budgetary restrictions or for other reasons only one of these facilities can be funded, the obvious choice must be LAMPF. The evidence presented here today should convince you that such a choice is necessary.

Presentation 2

COMPARISON OF LAMPF AND BLIP* FOR PRODUCING RADIOISOTOPES

By

M. E. Schillaci

In August, 1969, when we were first invited to brief representatives of the DID and other divisions of the AEC regarding the feasibility of utilizing LAMPF for the production of radioisotopes, we adopted a very noncompetitive approach in relation to the Brookhaven proposal. We believed then, as we do now, that both facilities merited funding. However, on that occasion and on several occasions since then,[1] representatives from Brookhaven have gone far beyond a straightforward presentation of the merits of the BLIP facility and have been rather agressive in comparing their facility with the proposed LAMPF Isotope Production Facility. In order to correct any misconceptions that may exist as a result of the previous unilateral comparison, we feel it is now necessary to present a detailed, quantitative comparison of these two facilities, and we have been encouraged to do so by a representative of the DID. Thus, although the tone of what follows in this section may appear to be unduly critical of the BLIP facility, it should be kept in mind that these remarks are in the nature of a reply to certain, specific claims made by Brookhaven representatives. In any case, every attempt has been made to remain fair and objective in this analysis.

The principal mechanism involved in the production of radioisotopes at both the Los Alamos Meson Facility (LAMPF) and the Brookhaven Linac Isotope Producer (BLIP) is the high energy proton-induced spallation reaction. In this reaction, many nucleons are knocked out of the nucleus, both singly and in clusters, resulting in a variety of possible nuclei. Some idea of the distribution of products that are obtained is provided by the example shown in Fig. 1. Here are shown mass-yield curves from a bismuth target bombarded at several proton energies. The 800 MeV curve has been sketched in to provide a qualitative comparison with the other curves which represent experimental data. The general features are: (1) a peak corresponding to spallation products near the target mass; (2) another peak (for higher mass targets) corresponding to fission products at intermediate masses; and (3) a rise at the low-mass end corresponding to fragmentation products. At lower energies, e.g., 200 MeV, the spallation peak is rather narrow. As the energy is increased, the spallation peak becomes broader and is somewhat decreased in height. Just how much broader and shorter this curve is at 800 MeV than at 200 MeV, for all possible targets, is a crucial question, for this determines the relative variety of the products and the magnitudes of their yields for these two facilities. We would now like to present a quantitative evaluation of LAMPF and BLIP in precisely these terms.

There are three factors which must be considered in any comparison of radioisotope yields:

1. beam intensity;
2. cross section dependence on energy;
3. thick-target attenuation of beam.

Other factors, such as processing facilities, personnel experience, etc., are of obvious importance and will be treated in the following presentation; however, such considerations are not relevant to a comparison of the production capacities of the two irradiation facilities. The first factor listed --

* Brookhaven Linac Isotope Producer.

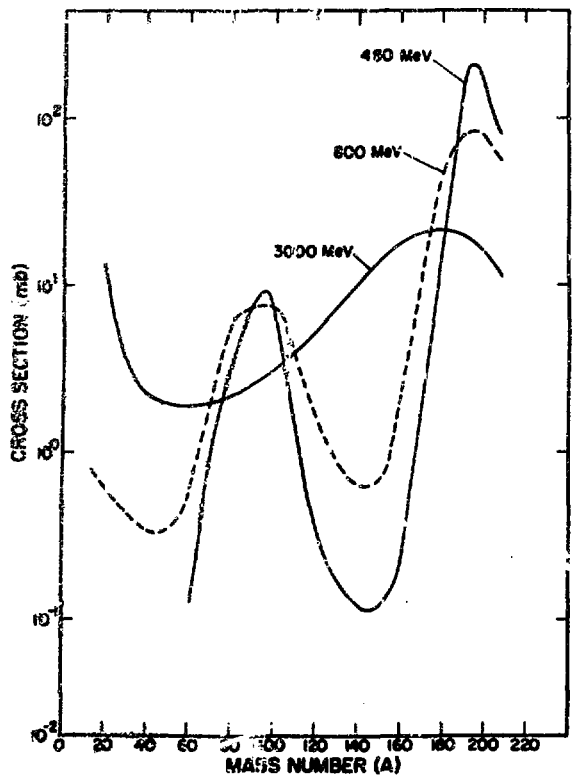

Fig. 1. Mass spectra of nuclides produced from a bismuth target by protons with energies of 480, 800, 3000 MeV.

beam intensity - - enters in an obvious way. The second factor - - cross section dependence on energy - - determines the relative yield for a given target-product combination at 200 MeV vs 800 MeV, assuming a target of fixed thickness. The third factor - - thick target attenuation of beam - - determines the penetration of the beam and, therefore, how much target material can be useful for producing radionuclides.

Let us now take up these separate points. The beam characteristics of the two facilities, which are of central importance to the discussion, are:

LAMPF: 700 ± 20 MeV, 400 ±100 μA

BLIP: 800 MeV, 180 μA.

The LAMPF figures are based on the assumption that all three pion producing targets in Target Area A are in the beam, plus the Bio-Med target which is also fairly massive. We wish to emphasize that these are realistic parameters for the Isotope Production Facility and not simply initial beam parameters. Thus, the beam intensity provides LAMPF with an advantage factor of (400 ± 100)/180= 2.2 ± 0.6.

Next, we must take up the factor dealing with cross sections. As stated in the presentations made to DID in August, 1969, by both BNL and LASL and from reports written at these institutions, both the Brookhaven group and ourselves have based our yield predictions on calculated, rather than experimental, cross sections. This has been necessitated by the paucity of data at the specified energies and has proved quite convenient for making rapid estimates of specific yields. Both our calculations are based on the same empirical relationship developed by Gösta Rudstam[2] about five years ago. This formula represents something like 1200 data points available at the time and was shown to be accurate to within a factor of two or three for almost all of the data. The accuracy tends to degenerate for low-mass targets because of the contribution of secondary particles, and for high-mass targets because of the importance of fission. In addition, this formula is meant to be used only for products that are three or more mass units removed from the target. Within these restrictions, it is felt that Rudstam's systematics are useful for estimating spallation cross sections for most target-product combinations. The mathematical details of the development of this formula can be found in Rudstam's paper[2] and is also discussed in a LAMPF report,[3] and, so, will not be discussed further here. We should emphasize that this same empirical approach forms the basis of both Brookhaven's and Los Alamos' yield predictions.

An abbreviated form of Rudstam's formula, which is taken from his paper, is given by

$$\sigma = G(A,Z,A_T)\,\frac{f_2(E)\,P(E)\,e^{-P(E)\,\Delta A}}{1 - 0.3/P(E)A_T}\,.$$

This formula gives the thin target cross section for the production of a nuclide with charge and mass numbers (Z,A) by an incident proton of energy E from a target nucleus with mass number A_T ($\Delta A = A_T - A$). Here, the function G does not depend on energy, whereas the functions f_2 and P depend on

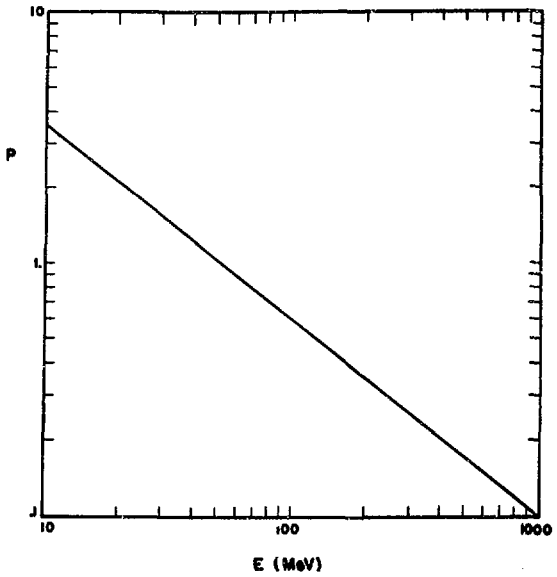

Fig. 2. The parameter P as a function of the irradiation energy.

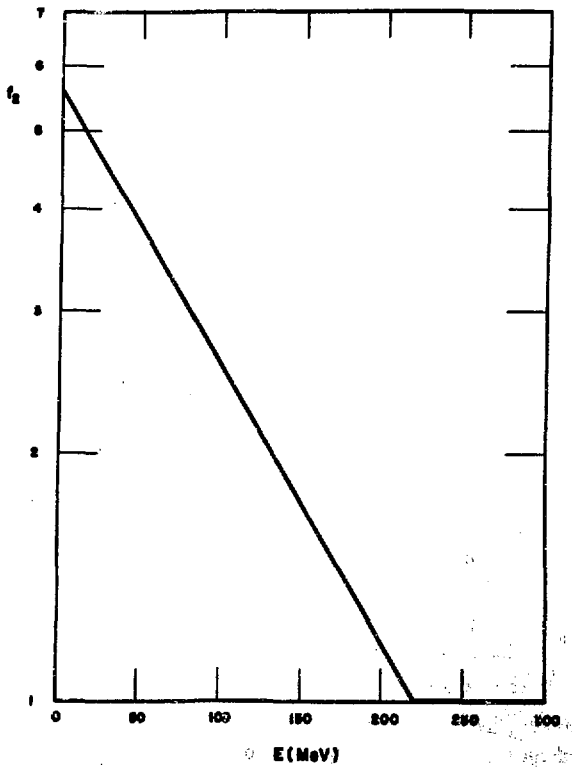

Fig. 3. The parameter f_2 as a function of the irradiation energy.

energy only. Rudstam shows in his paper that the optimum energy for producing a nuclide with a mass difference of ΔA from the target nucleus is given simply by

$$P(E_{opt}) \approx 1/\Delta A.$$

It turns out that 200 MeV is the optimum energy for making products with $\Delta A \approx 3$, and 700 MeV is optimum for products with $\Delta A \approx 8$. This can be seen from the curve for P vs E which is shown in Fig. 2. Also, the curve for f_2 vs E is shown in Fig. 3.

Let us now use Rudstam's formula to compare cross sections under both of these conditions -- viz., ΔA = 3 and ΔA = 8. First we assume that ΔA = 3, which is optimum for BLIP. Using the curves for P(E) and $f_2(E)$ at E = 200 MeV, we then obtain

$$\sigma_{opt}(200) = G(\Delta A=3)\frac{f_2(200)\,P(200)\,e^{-1}}{1 - 0.3/P(200)A_T}$$

$$\approx \frac{0.15\,G(\Delta A=3)}{1 - 0.86/A_T} \approx 0.15\,G(\Delta A=3).$$

For the production of this same nuclide (ΔA=3) at 700 MeV we obtain

$$\sigma(700) = G(\Delta A=3)\frac{f_2(700)\,P(700)\,e^{-P(700)/P(200)}}{1 - 0.3/P(700)A_T}$$

$$= \frac{0.090\,G(\Delta A=3)}{1 - 2.3/A_T} \approx 0.090\,G(\Delta A=3).$$

In forming the ratio of these cross sections, the function G, which does not depend on energy, is seen to cancel:

$$\left[\frac{\sigma(700)}{\sigma(200)}\right]_{opt\ BLIP} = \frac{0.090\,G(\Delta A=3)}{0.15\,G(\Delta A=3)} = 0.60.$$

This ratio, which gives the relative value of using LAMPF over using BLIP, with regard to the fundamental cross sections only, is optimized in favor of BLIP.

Next, we assume that $\Delta A=8$, which is optimum for LAMPF:

$$\sigma_{opt}(700) = G(\Delta A=8)\,\frac{f_2(700)\,P(700)\,e^{-1}}{1 - 0.3/P(700)A_T}$$

$$= \frac{0.048\,G(\Delta A=8)}{1 - 2.3/A_T} \approx 0.048\,G(\Delta A=8),$$

$$\sigma(200) = G(\Delta A=8)\,\frac{f_2(200)\,P(200)\,e^{-P(200)/P(700)}}{1 - 0.3/P(200)A_T}$$

$$= \frac{0.024\,G(\Delta A=8)}{1 - 0.86/A_T} \approx 0.024\,G(\Delta A=8).$$

Under these conditions, the relative advantage for LAMPF is given by

$$\left[\frac{\sigma(700)}{\sigma(200)}\right]_{opt\ LAMPF} = \frac{0.048\,G(\Delta A=8)}{0.024\,G(\Delta A=8)} = 2.0.$$

Summarizing these results, we see that regarding the cross section factor alone, LAMPF is at worst 60% that of BLIP, and at best two times that of BLIP.

The final factor to be dealt with involves the thick-target penetration. Now, a correct procedure would be to recalculate beam intensity, beam energy, and cross sections after every interval, say Δx, into the target. This is a rather complicated and lengthy procedure and is not really necessary for the desired accuracy. We, instead, chose a more empirical approach, utilizing existing thick-target data. In Fig. 2 is shown an example of such data obtained by Shedlovsky and Rayudu.[4] What is shown here is the effective cross section for producing various nuclides from a thick iron target as a function of depth. Such data were taken at 1 GeV and 3 GeV. The solid curves represent an attempt to fit the data with an empirical formula.[3] An important feature to note is that for products with relatively small ΔA, there is a buildup in the effective cross section, peaking at about 60 gm/cm^2 for the 1 GeV case. The effect is even more pronounced at 3 GeV. It is believed that this buildup is due primarily to secondary particles, in addition to the primary protons, producing such nuclides. If this is so, one would expect this effect to be less pronounced, if it exists at all, at 200 MeV.

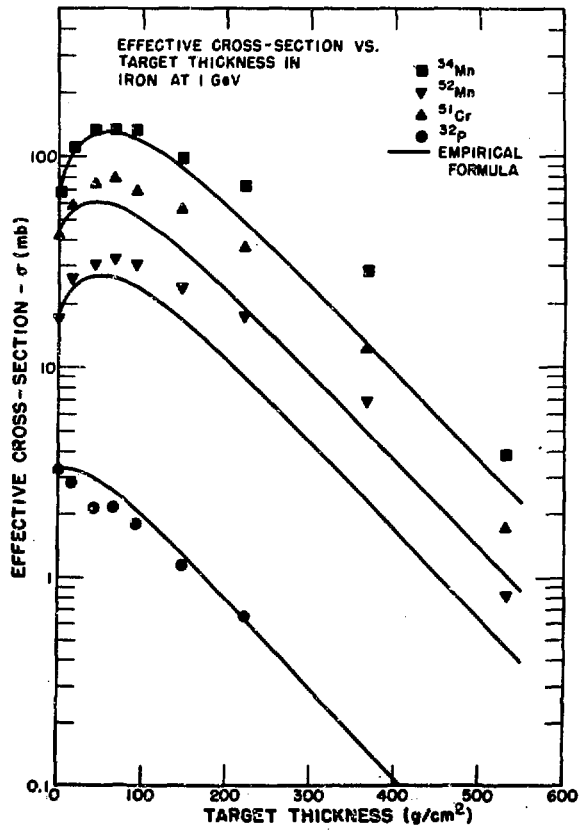

Fig. 4. Effective cross-section as a function of depth in an iron target at 1 GeV.

In recent weeks, several of us from Los Alamos completed a series of irradiations at the Space Radiation Effects Lab (SREL) 600-MeV proton synchrocyclotron in Newport News in cooperation with a group from Idaho Falls. One of the main objectives of these experiments was to elucidate such thick-target effects as a function of scattering material and size, and beam energy. Data were taken at 300 MeV and 600 MeV. In addition, thin-target data were obtained for several target materials, and these should provide a good check for our cross section calculations.

Getting back to the data shown in Fig. 4, it should also be noted that after the initial buildup, the effective cross section falls off exponentially. Since we are not sure how to handle this buildup effect at the lower energies, let us parametrize the data by ignoring this effect, and assume a simple exponential behavior of the effective cross section

with depth - - an approximation which is, if anything, generous to BLIP. The resulting parametrization of the data is given by

$$\sigma_{eff} = \sigma_o e^{-10x/E}.$$

Here, σ_o is the cross section at the front surface, which we assume to be given by our thin-target calculations. The depth, x, is in gm/cm^2 and the incident beam energy, E, is in MeV. Admittedly, the simple energy dependence assumed here is based on data taken at only two separate energies (1 and 3 GeV) plus the known trivial dependence at zero energy; however, our data taken at SREL at 600 MeV and 300 MeV should provide valuable additional information on the energy dependence.

The relevant factor in calculating the yield is the integral of σ_{eff} up to the thickness of the target (t = thickness, ρ = density). We thus obtain a thick-target factor given by

$$K(E) = \int_0^{\rho t} e^{-10x/E} dx = \frac{E}{10} (1 - e^{-10\ \rho t/E}).$$

What do we choose for the target thickness ρ t? A choice which, obviously, would be generous to BLIP would be to set ρ t equal to the range of 200 MeV protons in the target. As given in the Brookhaven report,[5] the range in ruthenium metal is about 35 gm/cm^2; and this is about right for copper and aluminium also, so let us use it. We thus end up with a thick-target factor advantage for LAMPF of

$$\frac{K(700)}{K(200)} = \frac{700\ (1 - e^{-0.50})}{200(1 - e^{-1.75})} = 1.7.$$

We can now put all three of the critical factors together for a yield comparison:

$$Y_E\ (\Delta A) \propto I\ \sigma_o(E,\Delta A)\ K(E),$$

$$R_{BLIP} = \frac{Y_{700}\ (\Delta A=3)}{Y_{200}\ (\Delta A=3)} = (2.2)(0.60)(1.7) = 2.2,$$

$$R_{LAMPF} = \frac{Y_{700}\ (\Delta A=8)}{Y_{200}\ (\Delta A=8)} = (2.2)(2.0)(1.7) = 7.5.$$

The subscripts BLIP and LAMPF for the yield ratio, R, correspond to ΔA=3 and ΔA=8, respectively. Thus, if one chooses a target which is equal to the range of protons available at BLIP, LAMPF will outproduce BLIP by a factor of about 2 to 7. We should point out here that the LAMPF beam will lose about 30% of its intensity and about 50 MeV in passing through such a target. Therefore, after passing through three such targets, the LAMPF beam will emerge with about 500 MeV energy and about 150 μA intensity, which is about equal to the total BLIP facility.

Although we feel that this analysis is certainly a fair one, perhaps a specific example taken from the Brookhaven report[5] would provide additional evidence. In Table III of the Brookhaven report, which is reproduced below as Table I, the yields from a ruthenium target at several energies are given for products with ΔA = 3, 6, 9. These calculations are based on the Rudstam formula, as was all of the preceeding analysis; however, the thick-target effects are treated rather differently here. Although the predictions at each energy are made assuming a target thickness equal to the range of protons at that energy, a comparison of the 200 MeV and 700 MeV results is made later in their report for targets of equal thickness. Thus, the total yield for ΔA = 3 at 700 MeV is given as 1.952 times that at 200 MeV; however, for a target of 35 gm/cm^2 thickness, the yield at 700 MeV is about 20% x 1.952 ≈ 40% that at 200 MeV. If one then multiplies by the beam intensity factor (400/180 = 2.2), one finds that the yield expected from only one such target at LAMPF is about 90% the total yield from BLIP. This comparison is for ΔA = 3, which is most favorable for BLIP, and, although it is considerably smaller than the figure of 2.2 obtained in our analysis, one is nevertheless led to the same inescapable conclusion - - viz., under no conditions will BLIP produce greater quantities of any isotope than LAMPF. This is particularly obvious when one remembers that at LAMPF, the beam will pass through <u>three</u> such targets and <u>then</u> emerge with the approximate potential for producing isotopes that is contained in the total BLIP facility.

Regarding the problem of radiopurity, one can see from Table I that the purity depends on the value of ΔA for the product. If the desired product has ΔA = 3, then the contamination for BLIP is about 40% that for LAMPF. However, if the desired product has ΔA = 9, then the contamination for BLIP is about

six times that for LAMPF. The advantage depends on the value of ΔA for the desired product. Clearly, one cannot expect to find a suitable target within three mass units of even most of the desired products.

Let us summarize our results for the total facility comparisons, taking the case of $\Delta A = 3$, which is most favorable for BLIP:

TABLE I[5]

EFFECT OF ENERGY ON THE RELATIVE ACTIVITY PRODUCED IN A RUTHENIUM TARGET FOR THREE ISOTOPES OF A GIVEN PRODUCT ELEMENT

(Normalized to the 200 MeV induced activity of mass 97)

Initial Energy (MeV)	I_f	A_T - A=3	A_T - A=6	A_T - A=9
100	0.949	0.280	0.035	0.005
200	0.809	1.000	0.236	0.064
225	0.778	1.097		
300	0.654	1.392		
400	0.512	1.646		
500	0.391	1.819		
600	0.293	1.914		
700	0.220	1.952		
800	0.160	1.942	1.127	0.695

[I_f is the fraction of original beam intensity remaining (after nuclear interactions) when the energy has been reduced to 50 MeV.]

$R_{BLIP}^{total} = 2.2\ [1 + .7 + (.7)^2] + 1 = 5.8$ (our calculations).

In this equation, the three critical factors resulted in a LAMPF advantage of 2.2 for a 35 gm/cm^2 target. Three such targets (with a 30% drop in beam intensity after each), plus remaining beam characteristics at least equal to the potential of BLIP, add up to give a total facility advantage for LAMPF of 5.8. If we choose $\Delta A = 8$, this advantage factor jumps to 20. Using the results given in Table I (from the Brookhaven report), together with the intensity factor of 2.2, one obtains a total facility advantage for LAMPF given by

$R_{BLIP}^{total} = 1.952\ (2.2) = 4.3$ (Brookhaven report).

Either 5.8 or 4.3 - - take your choice - - in either case the figure represents a minimum advantage for LAMPF. The maximum advantage factor for LAMPF is something like 20.

To put all of the preceeding analyses in more concrete terms, let us compare the yields of a representative list of radioisotopes for a one-year operation of both facilities. This is shown in Table II below. The scheduling assumed for BLIP is based on the assumption that the product of target thickness in gm/cm^2 and the irradiation time is proportionately the same as for LAMPF. Here, t_i is the irradiation time for a single target of a given material and (mult) is a multiplier which gives the number of such targets irradiated during the course of a one-year operation. For LAMPF we have assumed six 1-inch targets being irradiated simultaneously, while for BLIP we have assumed one target of 35 gm/cm^2 thickness being irradiated at any given time. We should emphasize that these are realistic yields in that we have assumed targets for each facility that can withstand the thermal and radiation environment. Furthermore, the loss in beam intensity in going through each LAMPF target is accounted for in an average way. The decay of activity during processing, packaging, and shipment has not been included in either example, however. The resulting product yields demonstrate concretely the clear superiority of LAMPF. For this particular example, the LAMPF yields range from 2 to 80 times the BLIP yields. Although the particular production schedules chosen for this example are certainly arbitrary, they are nevertheless fair and representative.

Finally, we would point out that many of the predicted yields advertised by Brookhaven are unrealistic. One glaring example is that of ^{123}I - - a most desirable product. As you may know, both LAMPF and BLIP plan to produce ^{123}I by first producing ^{123}Xe and allowing this to decay to ^{123}I. By such techniques, the purity of the desired product is greatly enhanced. While we plan to use a lanthanum target, Brookhaven uses in their calculations a CsI target, which has questionable stability under the intense radiation conditions to be encountered. The cross section for the production of

TABLE II

ONE YEAR PRODUCTION OF SELECTED RADIONUCLIDES

Product ($t_{1/2}$)	Target	LAMPF t_i (mult)	LAMPF Yield	BLIP t_i (mult)	BLIP Yield
^{26}Al (7.4x10^5 y)	Si	1mo(12)	130 μCi	0.054 yr	73 μCi
^{32}Si (~500 y)	V	1mo(12)	3.7 mCi	0.140 yr	46 μCi
^{42}Ar (33 y)			30 mCi		3.2 mCi
^{44}Ti (47 y)			120 mCi		20 mCi
^{67}Ga (78 h)	As	1wk(52)	5.9 kCi	1wk(6.864)	.81 kCi
^{68}Ge (280 d)			6.9 Ci		1.2 Ci
^{72}Se (8.4 d)			1.1 kCi		.43 kCi
^{83}Rb (83 d)	Zr	1mo(12)	310 Ci	0.150 yr	37 Ci
^{82}Sr (25 d)			390 Ci		35 Ci
^{88}Y (107 d)			350 Ci		150 Ci
^{123}I (13 h)	La	1wk(52)	27 kCi	1wk(7.384)	.64 kCi
^{128}Ba (2.42 d)			1.2 kCi		85 Ci
^{172}Hf (~5 y)	Ta	1mo(12)	15 Ci	0.382 yr	1.1 Ci

^{123}Xe from the ^{127}I in CsI is given experimentally as 8 mb.[6] Let us assume that the cross section for the production of ^{123}Xe from the ^{133}Cs in CsI is essentially the same as the measured cross section for the analogous reaction, ^{127}I(p,2p,9n) ^{117}Te, namely 50 mb.[7] Using a target of 35 gm/cm^2 thickness, we then obtain a yield of 87.4 Ci/day of ^{123}I for the BLIP facility. This figure is to be compared with an advertized[1] yield of 800 Ci/day! How can we reconcile this discrepancy? First of all, the Brookhaven group used calculated cross sections of 30 mb from Cs instead of 50 mb, and 220 mb from I instead of 8 mb. Secondly, they apparently used the density of pure Cs (1.96) instead of the partial density of Cs in CsI (2.31), and the density of pure I (4.93) instead of the partial density of I in CsI (2.20). If we "correct" our calculated yields by these factors we then obtain

$$\left.\begin{aligned}(\text{Cs}) \quad & 74.9 \times \frac{30}{50} \times \frac{1.96}{2.31} = 38 \\ (\text{I}) \quad & 12.5 \times \frac{220}{8} \times \frac{4.93}{2.20} = 770\end{aligned}\right\} 808 \text{ Ci/day},$$

apparently accounting for the noted discrepancy. We should point out that the daily yield of ^{123}I from a single lanthanum target can be as high as 150 Ci at LAMPF.

We have shown, by several different objective comparisons, that from the standpoint of yield alone, LAMPF is clearly superior to BLIP. This conclusion is based on our own independent analysis contained herein as well as from results contained in Brookhaven's report.[5]

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Presentation 3

PROPOSED TARGETING SYSTEM AND LASL HOT CELL AND RADIOCHEMISTRY FACILITIES

By

B. J. Dropesky

I would like to first describe the system we are proposing to use for target insertion, target cooling during irradiation, and target removal for an isotope production facility at LAMPF. It's necessary to appreciate the conditions expected to prevail at the main beam dump. With all the meson production targets in the beam we still expect 0.4 to 0.5 mA of ~700 MeV protons over a circular area about 6", down to possibly 4.5", in diameter to be impinging on the final target or beam stop. The rate at which these protons lose energy in passing through target material, i.e., dE/dx, varies from about 1 to 2 MeV per gram/cm^2 for high to low atomic number target elements. This means, for example, that if 1" thick targets of the metals, silicon, niobium, and tantalum, are placed in this proton beam, the energy deposited as heat would be about 5 kW, 14.4 kW, and 23.8 kW, respectively. Obviously, with such very substantial amounts of heat imparted to targets, provisions for cooling will be essential in order to prevent melting.

Water cooling of targets is the most efficient and practical means. Rim cooling has been investigated and found to be suitable only for very good conductors of heat such as copper, for example. For most target materials, face cooling - - that is, flowing a thin layer of water across the front and back faces of a target disk - - will be necessary. For the range of targets mentioned above, only 1 to 2 gallons/minute of water flow is required to remove the large amounts of deposited heat and maintain the targets at a moderate temperature. Figure 1 illustrates a thin walled target cell containing a 1" thick target of vanadium and 1/8" thick cooling water layers flowing over the front and back surfaces. Under the given irradiation conditions, 11.8 kW of heat will be deposited in the vanadium, water and stainless steel shell, and at 2 gpm of water flow, a reasonable temperature rise will be observed in the water and the central temperature in the target disk will be modest.

Of course, the use of water for cooling these targets presents some problems, namely the radiation dissociation of some of the water molecules into oxygen and hydrogen and also the production of radioactive nuclides, primarily by proton reactions on the oxygen nuclei. Because of this, the water layers should be kept as thin as possible. The water directly in the proton beam will become highly radioactive due to the production of 71-sec ^{14}O, 124-sec ^{15}O, 9.9-min ^{13}N, 20.5-min ^{11}C, 53-day ^{7}Be, and 12.3-yr ^{3}H. Therefore the water lines must be well shielded and the entire contaminated system must be an isolated closed loop. There are several such systems, including recombiners for the O_2 and H_2, to be provided at the LAMPF beam dump and our cooling lines would tie into one such system.

Figure 2 shows a version of a typical water-cooled target enclosure that would be coupled to the end of a horizontal stringer to be used to insert the target into the proton beam. Here the targets consist of 1/2" metal plates, which, for various

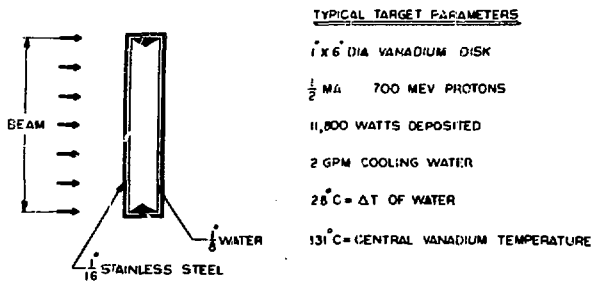

Fig. 1. Typical target parameters.

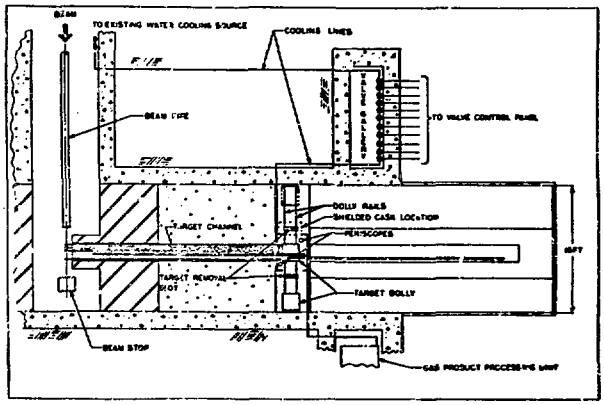

Fig. 3. Plan view of isotope production area.

may be preferable to using a single 1" thick target. The thickness and number of target disks could be varied considerably, depending upon the physical properties of the metal and on the desired rate of production of the useful radionuclides.

Figure 3 shows, somewhat schematically, an overall plan view of the proposed isotope production facility at the LAMPF beam dump. The residual proton beam, transported from the experimental area, leaves its vacuum pipe just ahead of our production station and enters a fairly massive water-cooled beam stop just behind the production station. Shown, in various positions are six ~26' long stringers of rectangular cross section (maybe 10" high by 2" to 3" wide) guided in channels through the 10' of steel and 15' of concrete that make up the shield wall. These stringers would probably be made of hollow steel or Dural channel through which the cooling water lines, gas lines, thermocouple cables, etc. would pass and then which would be filled with shielding material such as iron ore or steel shot. The number of channel slots for the target stringers was chosen to be six, for the full production facility, since such a number provides for maximum flexibility in choosing the optimum location for a given target-product combination.

Just outside the shield wall is a shielded trench where simple remote operations can be performed on the ends of the stringers; operations such as connecting and disconnecting water lines, coupling or uncoupling the target chambers, etc.. A simple dolly system operating in this trench will provide a means for conveying a highly radioactive target to a shielded cask for shipment to a hot cell for

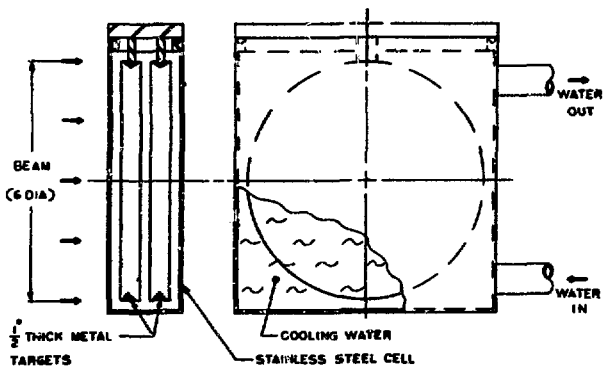

Fig. 2. Target enclosure.

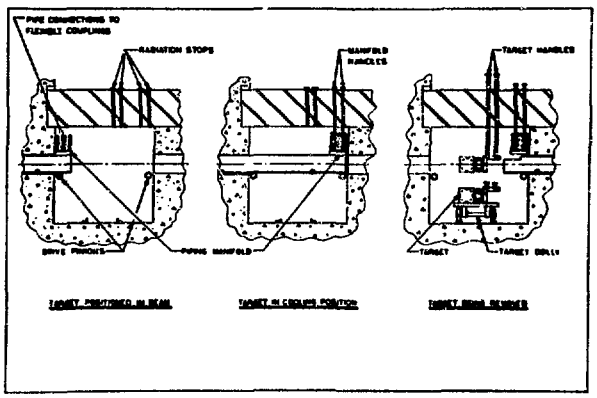

Fig. 4. Disconnection and removal-operations isotope production facility.

processing. Rather than providing costly lead glass windows through which to observe these remote operations, inexpensive periscopes are proposed for the job. A mating set of target stringer channels will be provided for the stringers to retract into when a target chamber is to be coupled to or uncoupled from a stringer. This set of channels will need only light shielding except for the trench end. Only buried cooling lines and a shielded valve gallery are shown on the recirculating water cooling system. We intend to develop at least one target cell, probably using cerium or lanthanum as target material, for producing 2-hr ^{123}Xe which will be swept with helium out to a processing unit where its decay product, 13-hr ^{123}I, would be separated.

Figure 4 is shown to illustrate the rather simple method proposed for performing remote operations on either end of the target stringer. The left view of a section through the trench shows a stringer all the way in so that a target is in the proton beam. A piping manifold, with flexible cooling water lines, is shown connected to the stringer. After the desired duration of target irradiation, the stringer would be retracted about 3 or 4 ft. to a cooling position and the cooling water temperature would be monitored until it shows that self-heating of the target by the β-decay process is negligible. Then working through a shield of about 18" of steel, and after the water lines have been blown out with air, an operator would unbolt the piping manifold with long uncoupling tools. The stringer would then be retracted completely until the target was accessible for uncoupling with another set of tools and lowered onto the dolly. We estimate that a single massive and long irradiated target may build up as much as 10,000 Curies of activity and that 18" of steel will provide adequate shielding for the operator above.

The lower sketch in Fig. 5 illustrates how an irradiated target chamber, after being moved on the dolly to just below a slot off to the side, in the steel shield, is picked up with a long set of tools and drawn up into the bottom loading cask. The upper sketch merely shows a target stringer in the irradiation position and also indicates how the outward movement of the Merrimac sliding door will result in preventing access to the trench. We anticipate no real problem here since the Merrimac door would be opened very infrequently and only when the accelerator is off and after our targets had been retracted to the cooling position or removed to the hot cells.

The proposed target enclosure system is obviously very versatile and should allow us to consider a wide variety of target schemes, for example, metal powders of high melting elements (for ease of dissolving), composite metal targets such as beads of poor heat conductor material in a matrix of good thermal conductor, many thin metal disks with very thin water films between them, and even low melting metals having low vapor pressures and high boiling points such that they can be allowed to melt and a sweep gas can be used to bring out gaseous products of interest.

I would like to digress for a moment to look at one aspect of the targeting system proposed for the Brookhaven Linac Isotope Producer (BLIP) as compared with the proposed LAMPF facility. This has to do with energy per unit volume of target materials for the two systems. To make the comparison I chose, arbitrarily, a 1 cm thick circular target of copper (8.92 gm/cm^2) since the dE/dx (rate of energy loss) for 200 MeV and 700 MeV protons in copper is readily available. If we take the targeting parameters that have been already indicated for BLIP and LAMPF, we arrive at the following table of comparisons:

	BLIP	LAMPF
Proton Energy	~ 200 MeV	~ 700 MeV
dE/dx (MeV/g/cm^2)	3.0	1.65
ΔE = (dE/dx)x 8.92 g/cm^2	26.8 MeV	14.7 MeV
Energy deposited = μA x MeV	180 x 26.8 = 4824 watts	500 x 14.7 = 7350 watts
Beam Diameter	2.0 cm	15.24 cm
Target volume in beam	3.14 cm^3	182 cm^3
Energy density	1536 watts/cm^3	40.4 watts/cm^3

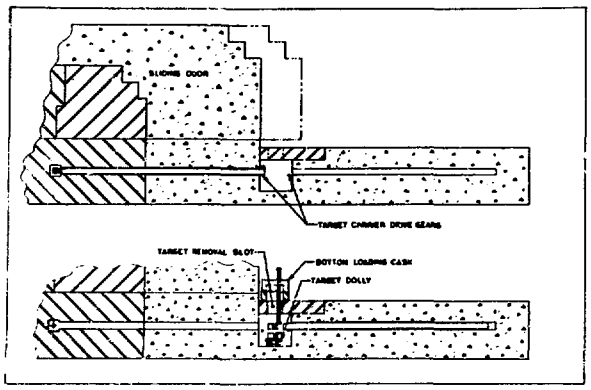

Fig. 5. Elevation views of target removal details.

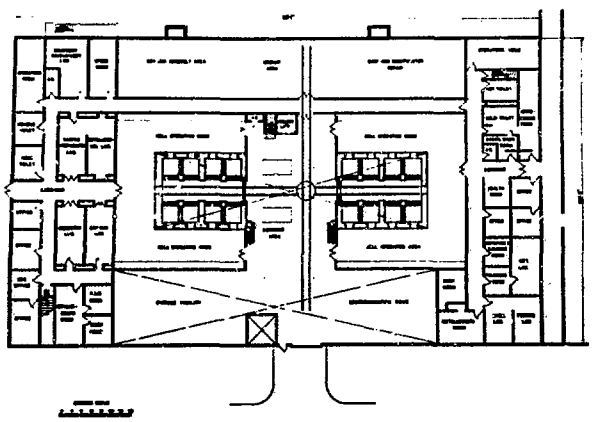

Fig. 6. Plan view of hot cells located in the LASL Chemistry-Metallurgy Building.

Fig. 7. Operating faces of a set of 4 hot cells.

This extremely high energy density for the BLIP case is about 38 times higher than the modest value in the LAMPF case and suggests that some difficult targeting problems exist for the former. For a target of copper, which is an excellent conductor of heat, the central temperature in the BLIP target will probably not exceed 100° C, with efficient water cooling of the front and back face. However, a target of lead, for example, which has ~ 1/10th the thermal conductivity of copper and a low melting point (327° C), would undoubtedly melt, internally, and thermal expansions would be severe. Of course, a BLIP target of 1 cm thickness would, as they have indicated, be made up of several thin "poker chip" disks with ~ 2 mm layers of cooling water flowing past the front and back face of each. However, this increase in the number of water layers in the proton beam results in a higher production of undesirable radioisotopes in the cooling water and a lower production of the desirable radionuclides in the target materials.

This relatively high energy density that will prevail under the BLIP targeting conditions also causes one to question the feasibility of using some of the target materials they have proposed for making various product isotopes. Some of these materials, such as phosphorus, iodine, and mercuric chloride have very low melting and boiling points, and ionic compounds, such as strontium bromide and cesium iodide, can be expected to undergo severe dissociation to their elements under intense proton bombardment. Of course, these targeting problems could be somewhat eased at BLIP by blowing up the beam diameter, which they will probably find necessary to do. This would somewhat defeat their stated objective of producing the desired radionuclides in as small a mass of target material as possible, presumably to minimize the scale of the chemical processing. Since we are proposing to recover for use only radioisotopes of elements other than the target element, and with no carriers of these elements added, then the specific activity of the recovered products would be the same whether we processed a 10 g target or a kG target. Anyway, even with our target cooling problems at LAMPF, which are much less severe than at BLIP, we are proposing to use only metal targets, at least initially. With the higher proton energy available we can afford to be

more selective of target materials that have favorable physical properties, even if they are further removed from the products we wish to make.

Now I would like to discuss the question of what facilities we have or will have and what chemical competence exists at LASL to process these highly irradiated targets. Regarding hot cell facilities, Fig. 6 shows a plan view of an array of elaborate hot cells located in the LASL Chemistry-Metallurgy Building, a few miles from LAMPF. These 16 hot cells are each capable of containing nominally 30,000 Curies (but actually 50-100,000 Curies) of gamma activity. The 8 cells to the left are presently committed to use for detailed testing of irradiated fuel elements from the Fast Flux Test Facility (center of the national program to develop fast flux breeder reactors). One of the other 8 cells is used for studies on irradiated plasma thermocouple devices. The remaining 7 hot cells are used intermittently for post-mortem operations and measurements on nuclear rocket engine (Project Rover) fuel elements after test runs in Nevada. Figure 7 is a photograph of the operating faces of a set of 4 of these hot cells showing the completeness of this facility.

Dr. R. D. Baker, head of the LASL Division operating this hot cell complex, has stated that, as far as he can foresee, some of the 7 cells for Rover work would be available, on an intermittent basis, for initial operations on the highly radioactive targets from LAMPF. In other words, those targets that are designed to produce large amounts of the long-lived products of interest and, therefore, would be in the proton beam for many weeks or months, could be disassembled, dissolved, and separated into different chemical fractions which then could be distributed to smaller cells for purification. It should be no problem scheduling such target processing when these cells are available.

Actually, there is a pair of large, multi-station 10,000 Curie hot cells now under construction in the so-called Merrimac Service Area of LAMPF. Shown to the left in Fig. 8 are three lead-windowed operating stations of the large hot cell to be used for servicing activated meson production targets, beam transport magnets, etc. To the right of the cell can be seen two windowed stations of the four that make up the nuclear chemistry hot cell. Two stations of this cell will be used primarily for the initial processing of proton irradiated targets involved in the nuclear chemistry basic research program. The other two stations would be available for the chemical processing of targets from the proposed isotope production facility.

In 1962 a hot cell wing was added to the LASL Radiochemistry laboratory. Figure 9 shows a layout of this wing with its 12 low level hot cells, each designed to handle 200 Curies of gamma activity, and the large dispensary bay capable of containing 2 thousand Curies of activity. The wing was built for radiochemical post-mortem diagnostic work on Rover Fuel elements and has been used predominantly for this project. However, as the Rover program has been trimmed back to less frequent tests of nuclear rocket engines in Nevada, these cells have become available for intermittent use on other projects. This facility would probably be ideal for an isotope production processing plant in which to dissolve irradiated targets, to distribute the chemical fractions to separate low level cells for purification, and to measure out and bottle the pure radioactive fractions for shipment. But, of course, no such commitment of this facility for this purpose is possible at this time. However, Dr. G. A. Cowan, head of the Radiochemistry group, has said that intermittent use of some of these hot cell facilities for the isotope production project is clearly feasible.

The next few figures are included to illustrate the completeness of this Radiochemistry Hot Cell Facility. Figure 10 shows the corridor, between the 12 low level cells, through which shielded casks containing highly radioactive fuel elements are transported to the dispensary bay for removal of the "hot" items. In Fig. 11 can be seen the operating face of the dispensary bay, and through the lead glass window can be seen the tracks of the small electric train used to transport items from the bay to any of the low level cells. A standard containment box, in which all chemical operations are conducted, is shown in Fig. 12 in one of the low level cells. Figure 13 illustrates the full scale use of a set of the low level hot cells during an operation on Rover fuel elements. Finally, Fig. 14 shows the indispensable, little U and Pu electric railroad which is completely remote controlled.

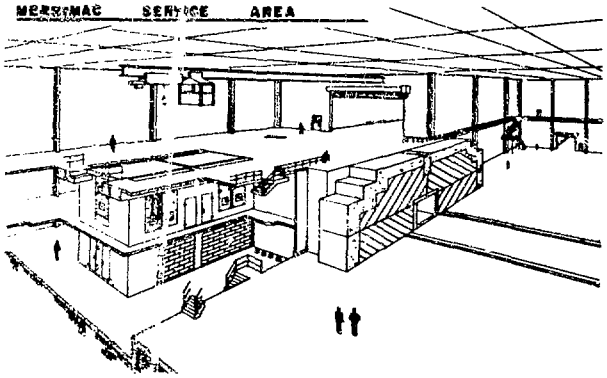

Fig. 8. Merrimac service area.

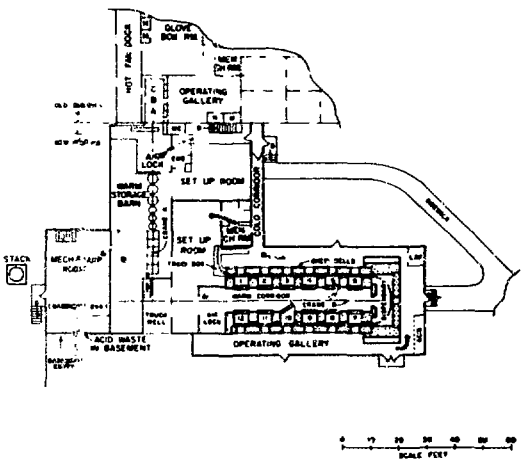

Fig. 9. Layout of the Hot Cell Wing of the LASL Radiochemistry Laboratory.

At this point I wish to emphasize another aspect of the business of chemical processing of irradiated targets and recovering, in pure form, the spallation radionuclides of value. In the LASL Radiochemistry group exists a rather formidable amount of radiochemical expertise and experience. This group has done all the radiochemical diagnostic measurements on LASL's nuclear devices (atmospheric and underground tests) and the nuclear rocket engine fuel elements. I think the book of "Collected Radiochemical Procedures" (LA-1721, 3rd Ed.) put out by this Radiochemistry group illustrates my point. Figure 15 shows the table of contents of

Fig. 10. Corridor between the 12 low level cells.

this book and lists separation and purification procedures for 45 individual elements, plus the 14 rare earth elements and the 14 actinide elements, or a total of 73 elements covered; a rather large fraction of the known elements. I do not mean to imply that this group of about 30 chemists and support personnel could shift from the important work presently being done to work on an isotope production program, but it is obvious that the existing expertise would be available to any group set up at LASL to develop an isotope production capability.

Before finishing, I should like to mention something about the measurements being made relative to the target irradiations obtained at SREL. These experiments, which were briefly discussed in the previous presentation and will be outlined in more detail in the next presentation, call for precision measurements of all the gamma-emitting spallation nuclides produced in a wide variety of

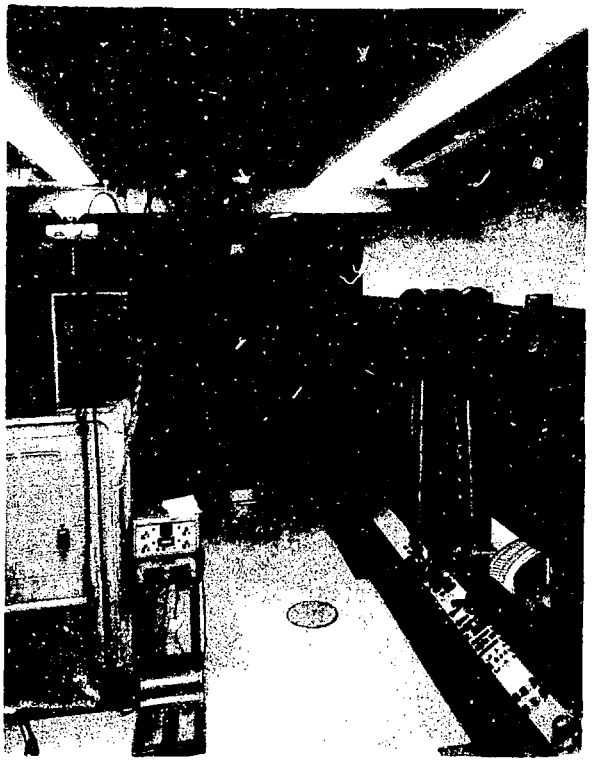

Fig. 11. Operating face of Dispensary Bay.

Fig. 12. A standard containment box in one of the low level cells.

target elements. The target foils, of vanadium, arsenic, niobium, molybdenum, tantalum, lead, and bismuth, most of which were bombarded with 600 MeV protons, but some of which were irradiated at 300 MeV, were quickly returned to the LASL Radiochemistry lab for the measurements. The number of protons each target was exposed to was determined by the conventional method of activation of aluminum and carbon beam-monitor foils. High resolution Ge (Li) gamma-ray spectrometry, followed by computer stripping of the gamma spectra, has been used to identify and to measure the yields of most of the spallation-produced radionuclides in the low and medium atomic number targets. Radiochemistry was required, prior to gamma spectrometry, for the high atomic number targets; for example polonium, bismuth, lead, thallium, mercury, and gold elemental fractions were separated from the bismuth targets. Some of the heavily irradiated targets will be used for investigating various isotope generator systems involving some of the potentially useful long-lived spallation nuclides, such as 5-y ^{172}Hf which decays to 6.7-d ^{172}Lu and 280-d ^{68}Ge which decays to 68-m ^{68}Ga. Measurements are also being made of the release, as a function of temperature, of xenon activity (namely, 36-d ^{127}Xe) from irradiated targets of various cesium and lanthanum compounds, in connection with a proposal to produce ^{123}Xe, sweep it from the target with helium, and collect the decay product, ^{123}I.

In summary, I would say that we have developed a feasible system for introducing targets into the LAMPF proton beam, for cooling them during irradiation, and for removing them to shielded containers for transport to hot cells. Clearly, we have substantial hot cell facilities at LASL for handling and processing the irradiated targets, at least on an intermittent basis and, depending upon future program schedules, possibly on a very regular basis. We have all the chemical and hot cell expertise

Fig. 13. Full scale use of a set of low level cells.

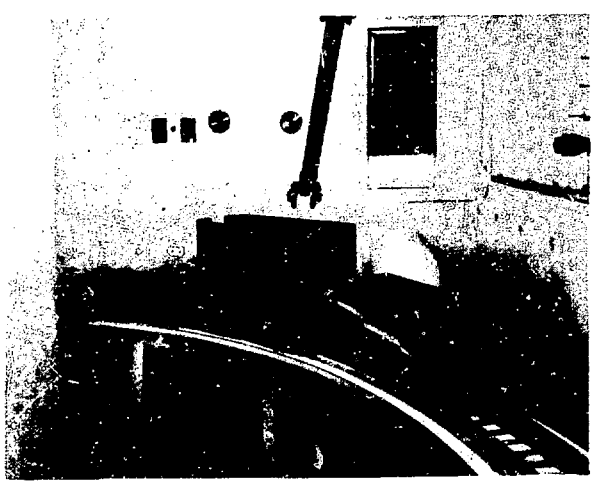

Fig. 14. Remotely controlled electric railroad.

CONTENTS

Fig. 15. Table of Contents of "Collected Radiochemical Procedures" (LA-1721, 3rd Ed.)

needed for developing the procedures and carrying out the chemical separations and purifications of the desired radioactive products from the targets. From the bombardments at SREL we expect to learn a great deal that will be of direct benefit in planning for isotope production at LAMPF.

Presentation 4

RADIONUCLIDES FROM LAMPF: YIELDS AND APPLICATIONS

by

H. A. O'Brien

I. INTRODUCTION

In the present section, specific radionuclides are discussed, including their decay properties, probable areas of application, and, in selected cases, projected demands. As pointed out previously, the spallation process results in the formation of both neutron-deficient and neutron-excess nuclides. Obviously only a few of the possible products can be discussed here. However, the brevity of this list should not convey the idea of a lack of interest in other products; but, instead, this list should be considered as a representative sample of the range of products that can be prepared in LAMPF.

The "probable areas of application" were determined from a consideration of the past or present uses of other isotopes of the same element, and from the fact that the products listed here represent major improvements in terms of availability, decay characteristics, and/or isotopic purity. In many instances, areas of application were identified through direct communication with potential users.

To arrive at projected demands for specific products necessarily entails a certain degree of speculation, and in many cases cannot be predicted *A Priori*.

A case in point is that of ^{99m}Tc, which, when first introduced as a possible brain scanning agent in 1964, was of unknown potential value. Yet within two years of its introduction, it surpassed, in number of patient administrations, all other isotopes used in brain scanning.[1] The widespread use of this nuclide in medicine today is attributable primarily to the new tagging procedures that were developed since its introduction, and even the most farsighted scientist involved in its early development failed to adequately predict the current prominence of this nuclide in medicine.

In those cases where projected demands for the nuclide in question are made, the annual demand is estimated on the basis of: 1) current use of other isotopes of the same element; 2) present doses of that nuclide in clinical evaluation studies; and/or 3) projections of scientists in the field of application. Care was taken to insure that these estimates are conservative. The important conclusion to be drawn from these projected annual needs is that the LAMPF production capability is amply justified in these cases.

II. RADIONUCLIDES FOR MEDICAL APPLICATIONS

A representative list of nuclides that are expected to be useful in medicine is presented in Table I. The yields given in this table and throughout this communication are based on the assumed conditions: 750 MeV protons; 1/2 mA beam current; a 1 in. thick target.

In those cases where the primary radionuclide product discussed herein is obtained directly from spallation reactions and not from an isotope generator system, it is well recognized that such a product will contain an admixture of isotopes of that element. The degree of isotopic "contamination" can be manipulated by varying the bombardment time and the subsequent decay time. However, the successful use of each of these products in the application stated must be demonstrated experimentally.

A. Gallium - 67

Investigations of the possible medical applications of gallium radionuclides were begun in 1950 using low specific activity ^{72}Ga. Between 1951 and 1953, ^{67}Ga was studied, but the recognition of its

utility was hampered by poor instrumentation. These studies were discontinued until 1961 when ^{68}Ga became available. Then in 1968, ^{67}Ga was again studied, but this time with a rectilinear scanner.[2] Although the original aim of these studies was to develop a new bone scanning agent, ^{67}Ga was discovered to localize in a variety of lymphomas, a truly spectacular breakthrough in locating such tumors. Since that time, clinical trials have confirmed the value of ^{67}Ga for this application.[3,4,5]

This nuclide decays entirely by electron capture with a total of 10 gamma photons associated with its decay. Of these gamma photons, three are prominent [0.296 MeV (22%), 0.184 MeV (24%), and 0.093 MeV (40%)][5] --- all within the energy range of conventional scanners. The 78-hr half life is convenient for shipping, but not too long to preclude its use on a routine basis. Since other radioisotopes of gallium have much shorter physical half lives, ^{67}Ga can be produced via spallation reactions in a radiochemically pure form.

The most significant advantage of ^{67}Ga-citrate is its selective concentration in soft tissue tumors, when normal binding sites are blocked with small amounts of stable scandium. Heretofore, no soft tissue tumor scanning agent was found to be as effective. As a result it is anticipated that this agent will become the diagnostic agent of choice for a variety of suspected cancers: a) breast; b) lymphomas; c) head and neck; and d) cervix and uterine, in which the soft tissues are involved to a major extent. Having the means for locating non-palpable, deep-seated tumors will allow for a more judicious choice of the mode of treatment, such as tailoring the radiation therapy depending

TABLE I

RADIONUCLIDES FROM LAMPF OF INTEREST IN MEDICINE

(Assumed: 750 MeV; 1/2 mA; 1 in. Thick Target)

Product	Daughter	Target	Production Yield	Comments
^{67}Ga(78 h)	--	As	68 Ci/d	Soft tissue tumor scanning agent.
^{68}Ge(280 d)	^{68}Ga(68 m)	As	320 mCi/wk	Short-lived positron emitter for multiple applications.
^{72}Se(8.4 d)	^{72}As(26 h)	Nb	8.7 Ci/wk	A new arsenic brain scanning agent.
^{82}Sr(25 d)	^{82}Rb(75 s)	Nb	26 Ci/wk	Short-lived Rb for rapid dynamic flow studies.
^{83}Rb(83 d)	^{83m}Kr(1.9 h)	Nb	15 Ci/wk	Rb for muscular dystrophy studies.
^{123}Xe(2 h)	^{123}I(13 h)	La	27 Ci/d	Lower iodine radiation dose to patients (about 1% dose of ^{131}I)
^{172}Hf(5 y)	^{172}Lu(6.7 d)	Ta	3 Ci/mo	General purpose rare earth tracer.
^{44}Ti(47 y)	^{44}Sc(4 h)	V	25 mCi/mo	Potential bone and soft tissue scanning agent.
^{42}Ar(33 y)	^{42}K(12 h)	V	6 mCi/mo	Established useful K nuclide.

on the degree of tissue involvement and the depth of the site. In the cases of breast cancer, the use of this diagnostic agent may well mean the difference between a radical resection of the breast and proximal lymph glands or merely the removal of the tumor mass from the breast followed by radiation therapy to the thoracic region.

A recent publication[6] estimated that the expected number of new cancer patients in these four categories in 1970 will total about 150,000. Clinical trials indicate that a 2 mCi dose of ^{67}Ga is adequate for most diagnostic purposes, with a minimum of two doses per patient.[7] Hence, the potential annual market for ^{67}Ga in cancer diagnoses alone is projected to be 600 Ci/yr. Allowing for processing and decay losses, the annual production of ^{67}Ga will have to be in the range of 900-1200 Ci/yr to satisfy this demand. This quantity is well within the annual production capability of one of our six target stations.

B. Gallium - 68

Up to the present time, positron emitting nuclides have been infrequently used in nuclear medicine, partly because of a lack of suitable instrumentation to detect coincidence gamma photons and partly because of the lack of generally available, inexpensive nuclides that decay by positron emission. The Anger positron camera[8] now provides the capability of imaging coincidence events as well as of selecting a plane of best focus, permitting radioisotope tomography with positron emitters.

Among the positron-emitting nuclides, ^{68}Ga possesses almost ideal properties. It is produced from the decay of 280-d ^{68}Ge, and has a half life of 68 min. Moreover, 88% of the decay of ^{68}Ga is by positron emission with few non-annihilation gamma photons. With the development of new compound tagging procedures in recent years, this nuclide may well become a valuable diagnostic tool, the development of which will be dependent on its general availability. No projections of annual use have been made for this nuclide; however, the ^{68}Ge production rate of 320 mCi/wk in LAMPF is expected to be adequate.

A recent study using ^{68}Ga-labeled iron hydroxide particles in conjunction with an Anger positron camera suggests that perfusion tomograms will have great value in accurately determining the size and location of perfusion abnormalities in the lung.[9]

C. Arsenic - 72

The availability of positron-emitting, 26-h ^{72}As should stimulate the development of brain tumor localization by coincidence counting techniques, especially since it can be "milked" from its 8.4-d parent, ^{72}Se. Hence, the daughter would be available with a useful shelf life equal to that of its parent. In comparison with 18-d ^{74}As, the ^{72}As has a shorter half life and emits about three times as many positrons per 100 decays, resulting in greater detection efficiency. Also, 32% of the decay of ^{74}As is by negatron emission, while ^{72}As decays entirely by electron capture (< 30%) and positron emission, resulting in a lower total body dose to the patient. The rate of ^{72}Se production in LAMPF is greater than 8 Curies per week.

D. Rubidium - 82

A very short lived ($T_{1/2}$ = 75s) isotope, ^{82}Rb would be of value in repeated, rapid dynamic studies, such as cardiac output measurements. It can be "milked" from a 25-day parent (^{82}Sr) that is producible in high yield (26 Curies per week). This rubidium isotope decays 96% by positron emission to stable ^{82}Kr, and could be followed in the body either by coincidence counting or by observing a single 511 keV gamma photon.

E. Rubidium - 83

At first glance it appears as though the ^{83m}Kr daughter of ^{83}Rb possesses ideal decay properties for nuclear medicine applications. It decays entirely by isomeric transition to stable ^{83}Kr, and has a relatively short 1.9-hr half life. However, the most energetic gamma photon emitted is only 32 keV, which is highly converted. On the other hand, ^{83}Rb has been useful in muscular dystrophy studies.[10] Although it possesses a longer physical half life (83 days) than 33-d ^{84}Rb, ^{83}Rb is more desirable because of its pure electron capture decay.

F. Iodine - 123

The most important isotopes in medicine today are those of iodine, as witnessed by the fact that, of the 5 million administrations of radioisotopes to patients in 1966, over 3.3 million (66.5%) were of iodine isotopes.[1] Because of this widespread usage in diagnostic applications, there is a great demand for ^{123}I as it would reduce the radiation dose to the patient to 1% that of ^{131}I. This dose reduction is of extreme importance in pediatric and maternity cases.

The present amounts of ^{131}I administered per patient for diagnostic purposes depend on the organ being studied (e.g., 35 μCi for thyroid, 150 μCi for liver, and 300 μCi for kidney).[11] A conservative estimate of the average dose of ^{131}I is 50 μCi per patient.[11] Further, it is assumed that the average amount of ^{123}I given per patient would be 5 times that of ^{131}I to obtain better scan images. This quantity times the number of iodine administrations in 1966 (see above) results in a projected annual demand for 803 Curies of ^{123}I. This projection does not include an estimate of the growth of patient administrations since 1966 nor of the amount that would be used in pediatric and maternity cases. To satisfy this projected demand, the annual production of ^{123}I would have to be between 3 and 4 kilocuries per year.

During the past few years, ^{99m}Tc has become more prominent in medicine and, in certain cases, has replaced ^{131}I as the isotope of choice. However, only iodine is recognized by the medical profession as giving an accurate thyroid function. In addition, it remains the element of choice for liver and kidney scans, and in cisternography, as a number of ^{99m}Tc-labeled compounds are observed to rapidly disintegrate upon injection into humans.[7]

Although ^{123}I has been made in rather large quantities (a few hundred millicurie batches) at Oak Ridge, the use of that product has been compromised due to the presence of traces of ^{124}I contamination. At LAMPF it is proposed to isolate the 2-hr ^{123}Xe from a lanthanum target during bombardment and subsequently recover the ^{123}I from the xenon. No ^{124}I contamination would result as the ^{124}Xe is stable. However, 60-d ^{125}I would be present in the product as a result of the decay of 17-h ^{125}Xe. By limiting the bombardment time and the time allowed for the decay of 2-h ^{123}Xe, an ^{123}I product with as little as 0.7% ^{125}I could be produced. This product would be acceptable to the medical profession,[11] and would be a great improvement over what can be produced at the present time. Because of the widespread use anticipated for this nuclide, it is imperative that it be made readily available on a routine basis, which can best be achieved by the continuous recovery system proposed at LAMPF.

G. Lutetium - 172

Evaluation of the medical potential of the rare earth elements (Z = 57 to 71) can be carried out utilizing a ^{172}Hf - ^{172}Lu generator. The parent (^{172}Hf) can be produced in LAMPF from a tantalum target, and, with a half life of 5 years, would provide a long-lasting supply of ^{172}Lu ($T_{1/2}$ = 6.7 days) for clinical evaluation. Since lutetium is a rare earth element and hafnium is not, a practical generator is feasible.

H. Scandium - 44

Reported to be a bone seeker,[12] ^{44}Sc would provide localization of metastatic bone involvement earlier than is possible utilizing radiographic techniques. This nuclide decays 95% by positron emission and 5% by electron capture, giving rise to a 1.15 MeV gamma photon per disintegration in addition to the 511 keV annihilation gammas. Scandium-44 is the daughter of 47-yr ^{44}Ti, which can be made in the LAMPF facility at the rate of 25 millicuries per month. The long half life of the parent is a significant economic attraction, and the half life, imaging characteristics, and chemical properties of the scandium daughter appear attractive as an all-purpose bone scanning agent. In addition, it has been suggested by Woodard that scandium may be of value for localizing soft tissue tumors.[13]

I. Argon - 42

The 33-y ^{42}Ar, which can be produced in LAMPF at the rate of 6 millicuries per month, decays by beta emission to 12-h ^{42}K and, hence, would be valuable to medical institutions as a continuous supply of this short-lived nuclide. The medical uses of ^{42}K are well established,[14] and include, among others, muscular dystrophy, blood flow, and myocardial[15] studies. Some medical people, who have

TABLE II

PROPOSED NUCLIDES PRODUCED BY SPALLATION REACTIONS AT LAMPF FOR USE IN MEDICINE

Product	Target	Reaction	Half-Life	Possible Medical Use
^{48}Cr	^{51}V	(p,4n)	23 hr.	Substitute for ^{51}Cr in blood pool and red cell evaluation.
^{52}Fe	^{59}Co	(p,2p6n)	8.3 hr	Substitute for ^{59}Fe in rapid iron metabolism studies.
^{83}Sr	^{93}Nb	(p,4p7n)	33 hr.	Bone Imaging substitute for ^{85}Sr.
^{129}Cs	^{139}La	(p,3p8n)	32 hr.	Cardiac muscle imaging (? evaluation of central tumor necrosis).
^{194}Hg	^{203}Tl ^{205}Tl	(p,2p8n) (p,2p10n)	700 d	Generator for carrier-free, 39-hr ^{194}Au (applications as ^{198}Au substitute, diagnosis).

recently been experiencing difficulties in obtaining ^{42}K as a pharmaceutical, are excited about the possibility of obtaining the 33-y parent, ^{42}Ar, as this would insure a regular, long-term supply of ^{42}K essentially on demand.[16]

A number of other nuclides produced by high energy proton reactions at LAMPF may be of value in medicine. Table 2 provides information on the physical characteristics, method of preparation, and the possible medical uses of some of these nuclides.

III. RADIONUCLIDES OF INTEREST IN THE PHYSICAL SCIENCES

One of the unique advantages in using 600-800 MeV protons for isotope production is the wide distribution of spallation reaction products that is obtained. As a result it is possible to prepare a number of useful products simultaneously in a single target, as illustrated in Tables I and II above. This high proton energy combined with the large beam intensity available in the LAMPF enables up to 6, 1-in. targets to be bombarded simultaneously, resulting in the preparation of numerous radionuclides, many of which are relatively far from the beta stability line.

Aluminum-26 ($T_{1/2}$ = 7 x 10^5 y) is the only nuclide of aluminum with a half life sufficiently long for use in studies of any extended time, and, as a result, would be useful in chemical studies of aluminosilicates, organometallic compounds, and geochemistry. In addition, it would be of value in solid state physics, metallurgy, and oceanographic studies. Although this nuclide is currently available, the product has an extremely low specific activity and sells for $43,000 per μCi (before November 1968, the price was $1,000,000 per μCi).[17] This nuclide decays 85% by positron emission and 15% by electron capture, giving rise to 1.81 MeV (100%) and 1.12 MeV (4%) gamma photons, in addition to the annihilation photons. With the LAMPF production facility, ^{26}Al can be made at the rate of 26 μCi per month, and the specific activity would be about 50% of the maximum value because of the simultaneous production of ^{27}Al. This represents an increase in specific activity of about 30,000 times that of the current ^{26}Al product. As a result, it is anticipated that the demand for this new product will substantially increase.

Another long-lived nuclide that has no shorter-lived isotope suitable for extended studies in the

TABLE III

LAMPF RADIONUCLIDE PRODUCTS OF INTEREST FOR (γ,n) SOURCES

Source ($T_{1/2}$)	Target	Yield (Ci)†	n/sec/Ci x 10^{-6}††	Neutron Energy (keV)
^{124}Sb (60 d)	--	--	2.8	23
^{88}Y (107 d)	Zr or Nb	250	2.9	151
^{206}Bi (6.24 d)	Pb	645 (t_1 = 15.3 d)	1.8	48
^{205}Bi (15.3 d)	Pb	180 (t_1 = 15.3 d)	1.5	89 (peak)
^{56}Co (77.3 d)	Cu	87	1.2 (D_2O)	185,514
^{124}I (4.2 d)	La	94	0.73	21 (peak)
^{156}Eu (15.2 d)	Tb	160	0.38	188-463 (7)
^{156}Tb (5.4 d)	Ho	370	0.080	164
^{48}V (16 d)	Cr	340	0.040	511
^{106m}Ag (8.3 d)	Sb	61**	0.087	146
^{119m}Te (4.7 d)	Sb	430**	0.053	377

† Irradiation conditions assumed: E = 750 MeV; I = 1/2 mA; $t_1 = T_{1/2}$

†† Beryllium is used as the target material except where noted otherwise. The geometry is assumed that of a sphere 1 cm thickness.

** These yields should be multiplied by the fraction of the isomer that is produced.

physical sciences is ^{32}Si ($T_{1/2} \cong 500$ y). Although this nuclide is presently unavailable, requests for it have been received from:

a) The Chemistry Department, Univ. of Calif. at Davis;

b) The Physics Department, Univ. of Minnesota;

c) IBM's Thomas J. Watson Research Center, Solid State Physics Section;

while inquiries as to its availability have been made by:

d) The Metallurgy Department, Case-Western Reserve;

e) The Biology Department, Washington State Univ.

Even with an estimated production rate of 740 μCi per month in LAMPF, the current demand, based on the above requests, for over 10 mCi cannot be met in less than one year.

One of the potential areas of application for radionuclides from LAMPF that has been looked into during the past year is that of photo-neutron sources, which are useful in the nuclear safeguards program, forensic science, field activation analysis, oil-well logging, etc. Some of the possible alternatives[18] to the presently used ^{124}Sb-Be source are given in Table III, which also contains pertinent information on ^{124}Sb for comparison. Of the nuclides listed in Table III, the most promising is ^{88}Y, which has a half life almost twice that of ^{124}Sb. The neutron output per unit activity of the ^{88}Y product affords greater flexibility in source geometry than does the ^{124}Sb. These advantages coupled with the high yield of ^{88}Y in the LAMPF indicate that the ^{88}Y-Be source will be economically superior. Present estimates indicate the need for about sixty ^{88}Y-Be sources (15 Ci/source) during the first year of operation.[19]

During August 1970, an informal meeting was held with physicists representing the AEC's Nuclear Cross Sections Advisory Committee to discuss the capabilities of the Isotope Production Facility at LAMPF for preparing, in tens of milligram quantities, radionuclide targets for use in nuclear physics

TABLE IV

LAMPF RADIONUCLIDE PRODUCTS OF INTEREST FOR NUCLEAR PHYSICS TARGETS*

Product ($T_{1/2}$)	LAMPF Target	Yield	
		Activity (Ci)	Mass (mg)
^{83}Rb (83d)	Nb	67	3.5
^{84}Rb (33d)		64	1.4
^{87}Y (80h)	Nb	840	1.8
^{88}Y (107d)		87	6.2
^{88}Zr (85d)	Nb	200	12.0
^{89}Zr (78.4h)		1300	2.9
^{91}Nb (10^7y?)		7.0×10^{-6}	18.0
^{92}Nb (10^7y)	Mo	6.0×10^{-6}	15.0
^{93m}Nb (3.7y)		11†	11.0†
^{148}Eu (54d)		120	5.1
^{149}Eu (106d)	Tb	130	11.0
^{150}Eu (5y)		12	18.0
^{167}Tm (9.6d)	Hf	820	7.0
^{168}Tm (86d)		260	20.0
^{173}Lu (1.4y)	Hf	110	53.0
^{174}Lu (3.56y)		44	52.0

* Irradiation conditions assumed: E = 750 MeV; I = 1/2 mA; t_i = 1 mo.

† These yields should be multiplied by the fraction of the isomer that is produced.

cross section measurements. These studies are part of on-going programs being supported by the AEC's Divisions of Research and Military Applications. As a result of this meeting, a list of targets of interest, together with both the activities and masses for each nuclide, was compiled and is presented in Table IV. Note that the calculated yields are based on a one-month irradiation. The yields and distribution of isotopic masses appear adequate, in general, for the proposed studies. It is anticipated that requests for such targets would occur with a frequency of one per year. To meet this demand would probably require two-thirds of the Isotope Production Facility at LAMPF for one month each year.

In this section and the one immediately preceding, conservative estimates of the annual demand have been made for ^{67}Ga, ^{123}I, ^{32}Si, and ^{88}Y. It is informative to compare the annual production rate for

TABLE V

COMPARISON OF PROJECTED ANNUAL DEMANDS FOR

^{67}Ga, ^{32}Si, ^{123}I, AND ^{88}Y

WITH ANNUAL PRODUCTION IN LAMPF AND BLIP*

Nuclide	Projected Annual Demand †	Annual Production	
		LAMPF††	BLIP
^{67}Ga	1,200 Ci	5,900 Ci	1,530 Ci
^{32}Si	10 mCi	3.7 mCi	0.082 mCi
^{123}I	4,000 Ci	27,000 Ci	1,130 Ci
^{88}Y	900 Ci	350 Ci	250 Ci

* Assume one LAMPF target station per year per nuclide, and the entire BLIP facility per 1/4 year per nuclide.

† Based on estimates contained in this communication.

†† Estimates from Table II, Presentation 2, this communication.

these four nuclides at LAMPF and BLIP on the basis of these projections. This comparison is presented in Table V, assuming the use of one LAMPF target station per year per nuclide, and the entire BLIP facility for 1/4 year per nuclide. Several important conclusions can be drawn from Table V. First, there apparently will exist a national requirement for large quantities of selected nuclides that can be satisfied through production from proton-induced spallation reactions. Second, the Isotope Production Facility at LAMPF is clearly necessary to produce radionuclides via spallation reactions, since the BLIP facility obviously is unable to satisfy the annual demand for all four nuclides in this example. Third, the annual time required at LAMPF to produce sufficient quantities of ^{67}Ga and ^{123}I can be reduced by factors of 4 and 6, respectively, thereby providing added target space, in addition to the two remaining target stations not considered in this example, for producing greater quantities of ^{32}Si and ^{88}Y as well as the other radionuclide products discussed in this communication.

IV. CURRENT RESEARCH IN SPALLATION REACTIONS

Although present plans indicate that the Isotope Production Facility at LAMPF will not be operational until January, 1973, the paucity of data on spallation reactions in the LAMPF energy range has necessitated the initiation of a modest research effort at the Los Alamos Scientific Laboratory to insure the full utilization of this facility with the least possible delay. The major objectives of these research studies are the following. (1) Measure spallation reaction cross sections in the 500-750 MeV proton energy range to enable the selection of the optimum target for each specific radionuclide product or combination of products. Other factors that relate to this question are the thick-target buildup effect, the effect of scattering material Z, and the effect of varying target diameter on the cross section. (2) Develop chemical flow charts for recovering several radionuclide products from a common target, using targets bombarded with low-intensity, 600-MeV protons as tracer materials. (3) Where possible during these early studies, recover usable quantities of future LAMPF products to initiate applications development through cooperative research investigations with interested colleagues in the proposed field of application.

The first irradiations in this program, being carried out on a collaborative basis with the Physics Section of Idaho Nuclear Corporation, utilized the 300- and 600-MeV protons available at the Space

Radiation Effects Laboratory (SREL) in Virginia. These studies are being sponsored jointly by the AEC's Divisions of Research and Military Applications, and by NASA, which operates the SREL facility and, in addition, partially funds the Idaho Nuclear Corporation group. However, the availability of these funds cannot long continue, and it is imperative to obtain support for this work on a continuing basis.

The primary goals of these initial irradiations are the following: a) measure thin target cross sections for a variety of targets well spaced in the periodic table to verify the Rudstam systematics, which forms the basis for estimating all spallation reaction yields, and to determine the optimum target for each specific product; b) measure the effective cross sections as a function of depth for the thick target case; c) study the effect of varying the Z of the scattering material on the effective cross section; d) determine the effect of varying the diameter of the scattering material on the thick target buildup effect;[20] and e) measure the magnitude of the buildup effect in thick targets at 600 and 300 MeV.

In addition to the above stated objectives, the irradiations performed at the SREL facility will provide activated targets having product distributions similar to those expected from the LAMPF facility. These targets will be employed in radiochemical studies to develop the chemical flow charts to be used in the processing of the targets from LAMPF. Due to the variety of elements formed in targets subjected to bombardment with 700-800 MeV protons, as discussed in a previous section of this communication, the complexity of these chemical recovery studies cannot be overemphasized. For this reason it is mandatory that these studies be initiated well in advance of the LAMPF facility, and on a continuing basis, to insure the earliest availability of radionuclide products from Los Alamos,

Also, targets of V, As, Nb, and Ta were subjected to extended bombardments with 600 MeV protons in the internal beam of the SREL machine. The intent behind these irradiations was to build up sufficient quantities of longer-lived products in these targets to permit their recoveries and use in the initiation of limited applications development.

V. RADIONUCLIDE APPLICATIONS DEVELOPMENT

As shown above, many of the radionuclides from the Isotope Production Facility at LAMPF will be utilized in existing AEC-funded programs at Los Alamos and other laboratories throughout the nation. In addition to these basic and applied research applications in the physical sciences, a new program in the area of medical radiotherapy holds a promise for greatly stimulating research in the use of radionuclides, both as radiochemicals and tagged pharmaceutical compounds, as agents for locating all known types of cancer tumors in humans. The purpose of this new program is the evaluation of the potential of negative pions as a new modality for cancer therapy.[21,22]

Since the LAMPF will produce the most intense flux of negative pions in the world, the pre-clinical investigations and eventual therapeutic trials will, in all likelihood, be centered in Los Alamos. In anticipation of this program, the National Cancer Institute has awarded a two-year planning grant to the University of New Mexico School of Medicine in order to expedite, in maximal fashion through early planning, the introduction of negative pions as a therapeutic modality in the treatment of tumors.

Of major importance to the effective utilization of negative pion beams in tumor therapy is the need to accurately define the tumor limits prior to treatment. The successful use of radionuclides to locate a variety of human carcinomas and lymphomas in the past holds promise that, with an intensive research effort, other radionuclide agents will be found to localize in a wider variety of tumors. The importance of the contribution of radioisotopes to the overall negative pion therapy research effort was formally recognized during the recent organization of the LAMPF Biomedical Users Group, with the establishment of an Isotopes and Diagnostic Applications Subcommittee. Table VI indicates the national interest that exists in this program, which will be centered in Los Alamos. Hence, a sizeable local demand for medical radioisotopes, both for research and diagnostic uses, will develop in the foreseeable future.

It is anticipated that financial support for research studies of tumor localization with radioisotopes will come from a variety of federal and

TABLE VI

ORGANIZATION OF THE LAMPF BIOMEDICAL USERS

Steering Committee

Chairman:	Chaim Richman (U. of Texas at Dallas)
Alternate Chairman:	Wright Langham (LASL)
Ass't. to Chairman:	Dave Groce (JRB Associates)
Members at Large:	Morton Kligerman (Yale University)
	R. E. Anderson (UNM School of Medicine)
	Cornelius Tobias (Donner Laboratory)
	Andrew Koehler (Harvard Cyclotron)
	David Hussey (M. D. Anderson Hospital)
	Joseph Castro (M. D. Anderson Hospital)

Subcommittees:

1. Isotopes and Diagnostic Applications
   - Chairman: J. D. Shoop (UNM School of Medicine)
   - Alternate: H. O'Brien (LASL)
2. Physical and Biological Dosimetry
   - Chairman: M. R. Raju (LRL - Berkeley)
   - Alternate: P. Dean (LASL)
3. Cellular Radiobiology
   - Chairman: Paul Todd (Penn State University)
   - Alternate: Don Peterson (LASL)
4. Radiation Therapy
   - Chairman: Max Boone (University of Wisconsin)
5. Facility and Beam Line
   - Co-Chairmen: Paul Franke (LASL) and Dick Hutson (LASL)
6. Whole Animal Radiation Biology and Pathology
   - Chairman: Charles Key (UNM School of Medicine)

private institutions. The National Cancer Institute is already supporting the planning grant mentioned above. In addition, the American Cancer Society recently informed the University of New Mexico School of Medicine that a grant request to support investigations of tumor localization with radioisotopes would receive favorable consideration.[11]

Other groups, some of which were mentioned previously, around the country have expressed an interest in the Isotope Production Facility at LAMPF. A list of a few of these institutions is presented in Table VII. It will be noted from this table that most are medical institutions. One in particular is worth additional comment. It is well known that the University of Miami School of Medicine has obtained private funding to purchase and operate a multi-particle medical cyclotron. Why then are they interested in obtaining radionuclides from Los Alamos? The answer is simply economics. Greater economy is achieved by purchasing hard-to-produce, longer-lived nuclides than to develop costly target systems themselves and run their machine many hours to produce small amounts.[23] This course of action, however, would have to be taken if these nuclides continue to remain generally unavailable.

TABLE VII

INSTITUTIONS EXPRESSING INTEREST IN THE ISOTOPE PRODUCTION FACILITY AT LAMPF

Upstate Medical Center	Syracuse, N. Y.
Sloan-Kettering Instutute for Cancer Research	New York, N. Y.
Baylor College of Medicine	Houston, Texas
M. D. Anderson Tumor Institute	Houston, Texas
Johns Hopkins Medical Institutions	Baltimore, Md.
Southwestern Medical School	Dallas, Texas
Mayo Clinic	Rochester, Minn.
University of Miami School of Medicine	Miami, Fla.
University of New Mexico School of Medicine	Albuquerque, N. M.
Vanderbilt University School of Medicine	Nashville, Tenn.
Medical Div., Oak Ridge Assoc. Universities	Oak Ridge, Tenn.
University of Michigan	Ann Arbor, Mich.
Cincinnati General Hospital	Cincinnati, Ohio
University of Colorado School of Medicine	Denver, Colo.

VI. SUMMARY

The information contained in this communication clearly demonstrates a national interest in, and need for, new radionuclide products in quantities that can be made available from the Isotope Production Facility at LAMPF. It is evident from a comparison with the BLIP facility that the superior production capability of the LAMPF facility will be required to satisfy projected annual demands for selected nuclides.

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