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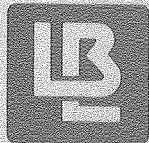https://escholarship.org/uc/item/2tc3155k

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THE FEASIBILITY OF PRODUCING ^{11}C, ^{13}N, ^{15}O, and ^{18}F
WITH HEAVY-ION BEAMS

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DISCLAIMER

The Feasibility of Producing ^{11}C, ^{13}N, ^{15}O, and ^{18}F

with Heavy-Ion Beams[*]

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Abstract

The feasibility of producing the short-lived tracers ^{11}C, ^{13}N, ^{15}O, and ^{18}F with heavy-ion beams was investigated. Production cross sections for 15-MeV p, 75-MeV ^{3}He and 300-MeV ^{12}C beams incident on H_2O targets were calculated using the particle evaporation code ALICE (3). These calculations predicted substantial thick target yields of all four activities with the heavier beams. Experiments were performed with 15-MeV p and 75-MeV ^{3}He beams to check the accuracy of the theoretical calculations. Absolute product yields were measured with NaI well counters, and chemical composition was determined by GC and HPLC.

For several years we have been developing techniques for the production, purification and utilization of the short-lived tracer ^{13}N ($t_{\frac{1}{2}}$ = 9.96 min) in our studies of biological nitrogen transformations ([1],[2]). Sources of ^{13}N were typically produced by the $^{16}O(p,^{4}He)^{13}N$ reaction using 15-MeV protons from the Michigan State University Cyclotron to bombard natural H_2O targets. This beam energy was chosen to optimize the production of ^{13}N without significant contamination from ^{11}C and ^{15}O which are produced at higher beam energies. Small amounts of ^{18}F were produced from the $^{18}O(p,n)^{18}F$ reaction; however, this did not interfere with our experiments. The sources varied in chemical purity containing typically about 85% $^{13}NO_3$ with the remainder $^{13}NH_4^+$, $^{13}NO_2^-$ and small amounts of labeled N_2O and N_2 (< 2%). Purified sources of all these species were routinely obtained by fast chemical techniques, gas chromatography (GC) and high pressure liquid chromatography (HPLC).

With the closing of the low energy accelerator at Michigan State University and the subsequent construction of a new heavy-ion facility there, it was desirable to investigate the feasibility of producing ^{13}N with the less optimal heavy-ion beams. Although it was anticipated that the heavy-ion reactions would produce substantial quantities of ^{11}C, ^{15}O and ^{18}F, we hoped that simple chemical techniques, which we had already developed, would be adequate to purify the sources. Thus, if sufficient quantities of ^{13}N were produced by heavy-ion reactions it would be possible to continue our tracer studies with these beams.

Two methods were chosen to investigate the usefulness of ^{13}N production by heavy-ion beams. First we performed theoretical calculations of ^{13}N yields with 15-MeV p, 75-MeV ^{3}He, and 300-MeV ^{12}C beams on water

targets (typical MSU energies) using the particle evaporation code ALICE ($\underline{3}$). Then we experimentally tested the accuracy of these calculations by producing ^{13}N activity with a 75-MeV ^3He beam from the Michigan State University Cyclotron. This beam should produce similar reactions and experimental conditions to those expected for heavier ions of the same energy to mass ratio. Sources produced by the ^3He beam were analyzed by decay half-life, HPLC and GC to determine absolute yields and chemical composition. Comparisons between this analysis of ^3He induced products, previous 15-MeV p experimental results and the ALICE calculations were then made to determine both the usefulness of the calculations and the feasibility of producing experimentally useful short-lived isotopes with heavy-ion bombardments.

ALICE Calculations

Cross sections for the production of ^{11}C, ^{13}N, ^{15}O and ^{18}F were calculated for 15-MeV p, 75-MeV ^3He, and 300-MeV ^{12}C beams incident on natural H_2O targets using the particle evaporation code ALICE ($\underline{3}$). This program was used to calculate the total $p + n + \alpha$ evaporation cross sections for all partial waves assuming the parabolic model ($\underline{4}$) to determine the compound nucleus cross section and using the n,p, and α binding energies of Wapstra ($\underline{5}$). The thick target yields for the isotopes were then calculated by integration using the ALICE cross sections and the particle range tables in Lederer et al. ($\underline{6}$). These yields are shown in Table 1 where they are expressed in the units mCi/μA per 10 min bombardment and corrected for decay during irradiation. For ^3He and ^{12}C the calculations assume a particle beam current and not a charge current. In Figures 1-3 we have shown the calculated thick target yields of ^{11}C, ^{13}N, ^{15}O and ^{18}F as a function of incident beam energy. Here the yields are expressed in the units mCi/μA per second. To

calculate the yield for a given bombardment interval one should use the relation

$$Y = Y° \frac{\lambda t}{1-e^{-\lambda t}}$$

where

$\lambda = \ln 2 / t_{\frac{1}{2}}$ $Y° = $ yield from Figures 1-3

$t = $ bombardment interval $Y = $ yield per μA beam current for a bombardment interval t.

$t_{\frac{1}{2}} = $ half-life

Experimental Results

The procedures for bombardment and analysis which we discuss below are described in greater detail in Reference 1. After both the proton and ^{3}He bombardments, samples were counted in calibrated NaI well detectors to establish absolute source intensity and time-decay characteristics. Sources were simultaneously analyzed by HPLC (with a NaI coincidence detector) and by GC (with a proportional counter detector) to determine their chemical composition. After chemical purification, the sources were again analyzed to determine purity. All absolute activites discussed below are normalized to a zero time at the end of the bombardment.

15-MeV proton bombardment

Sources produced by 15-MeV protons were shown by a two component fit half-life analysis of the multiscaled well counter data to contain about 1% ^{18}F (by activity) and no detectable ^{11}C or ^{15}O. The total absolute activity recovered after this bombardment is indicated in Table 1 and represents 84% of the calculated yield. This agreement is especially striking when we consider that the bombardment system was not gas tight and some ^{13}N gases were probably lost. A typical HPLC separation for a raw source of ^{13}N is shown in Figure

4. The chemical composition varied with beam conditions but we typically found 75-90% NO_3^-, 5-10% NO_2^- and 0.5-25% NH_4^+. Various separation techniques, described in Reference 1, were utilized to produce samples of > 99% purity in the desired chemical species as confirmed by HPLC or GC analysis. Activity from ^{18}F did not elute from the Partisil 10-SAX strong anion exchange column used in HPLC and hence could be easily removed from the samples. Analysis showed that about 90% of the gas retained in the source water was N_2, the remainder being N_2O. An unknown amount of ^{13}N gas was lost during bombardment and handling. No measurable increase in the activity of the air surrounding the bombardment system was observed leading us to believe that the amount of radioactive gas produced by the proton beam on water was small. The amount of gaseous activity found in the water was small and could be effectively removed by evaporation to dryness or by sparging the sample with helium.

75-MeV ^{3}He bombardment

The sources produced with ^{3}He were handled similarly to those from proton irradiations except that only a 2 min bombardment at 100 particle nA was performed. The yields were then normalized to mCi/μA per 10 min for comparison with the other data. The decay data from the NaI well counter for the raw source were analyzed by a four component half-life fit to determine the relative amounts of activity present. The stripped decay curve is shown in Figure 5, and the resultant analysis is given in Table 1. Again the ^{13}N and ^{18}F production rates agreed qualitatively with the calculations, with larger amounts of activity being produced than were predicted. However, no ^{11}C was observed and only 25% of the expected ^{15}O was recovered. An HPLC analysis of the raw source is shown in Figure 6a. In addition to NO_3^-, NO_2^- and NH_4^+ we observed two previously undetected peaks. The first new peak,

occurring near the solvent front, is apparently $^{15}O-H_2O_2$ (personal communication Joe Chasko) and the second peak elutes at the position expected for the ^{13}N gases N_2O, N_2 or NO. A further analysis of the HPLC spectrum peak areas, after correction for decay, indicates that the amount of $^{15}O-H_2O_2$ produced represents 41% of the total activity observed by HPLC ([7]). This is the same quantity of ^{15}O that was observed by half-life analysis (correcting for ^{18}F which remained on the HPLC column). GC analysis of the raw source gases indicates the presence of one major and one minor component. The elution characteristics of these gases from a Porapak Q column correspond to an identification of the major component as $^{13}N-N_2O$ and the minor component as $^{13}N-N_2$ and/or $^{13}N-NO$.

The 0.7 ml raw source was acidified by the addition of 0.1 ml of 0.01 N HCl. The head space gas of the acidified source was sampled and analyzed by GC for the presence of $^{11}C-CO_2$; none was detected. The sample was then evaporated to dryness with a flash evaporator and reconstituted to 1 ml with H_2O, after which 0.1 ml of 0.01 N NaOH was added and the sample was again evaporated to dryness. Water was then added and the treated source was counted in the NaI well detector. Half-life analysis of the multiscaled decay curve indicated only ^{13}N and ^{18}F activity (Figure 7) and the relative proportion of these two isotopes was nearly the same as in the original source. The HPLC analysis of the treated source is shown in Figure 6b. Only $^{13}N-NO_3^-$ was observed in this sample which is consistent with the removal of virtually all other activities; however, too much time elapsed before completion of the second analysis (about 30 min) to verify that all of the ^{15}O was removed by this treatment. After purification 50% of the ^{13}N activity remained (all as $^{13}N-NO_3^-$). This represents the fraction that existed as NO_3^- in the original source;

thus, apparently all NH_4^+, NO_2^- and N-gases were removed by the treatment.

Discussion

We have demonstrated that useful sources of ^{13}N can be generated by 75-MeV ^3He beams. For both 15-MeV p and 75-MeV ^3He, the ALICE calculations were successful in predicting ^{13}N and ^{18}F production rates. ^{11}C and ^{15}O yields appear to be significantly overestimated by ALICE; however, this result may reflect losses from the bombardment system which was not designed to be gas tight. One might expect ^{11}C to be produced mostly as CO_2 and/or CH_4, both of which could be subject to loss from the thermally hot target. ^{15}O may be produced primarily as H_2O_2, H_2O and O_2, so that much of the O_2 and some of the H_2O may be lost. Conversely, ^{13}N will be produced largely as the ionic forms NO_3^-, NO_2^- and NH_4^+ with only portions of the minor gaseous products N_2O, N_2 and NO being lost. This is consistent with our experience from low energy protons where only small amounts of the gases were observed. Finally, ^{18}F may be produced almost entirely as F^- and should not be lost from solution. This is consistent with the fact that no detectable loss of ^{18}F occurs during evaporation to dryness.

These results lead us to believe that usable sources of ^{13}N, ^{15}O and ^{18}F can be produced with heavy ions. Although ^{11}C was not observed here, nuclear reaction theory strongly predicts substantial production of this species. Various fast chemical techniques such as separation by HPLC or GC can provide pure sources of the different ^{13}N labeled chemical species. Similarly the use of gas-tight target systems, variation of target materials, and innovative separation techniques such as differential trapping should allow enhanced production of the other short-lived tracers. In addition, the yields of these activities can be qualitatively predicted by the computer

code ALICE. In Table 1 we have shown our calculations for a 300-MeV ^{12}C beam. Although the yields are estimated to be somewhat lower for ^{12}C than for ^{3}He, substantial quantities of activity are expected (comparable to 15-MeV p). If our experience with ^{3}He can be used as a guideline, these estimates may be somewhat conservative for ^{13}N and ^{18}F production. For ion beams heavier than ^{12}C the predicted yields will be further increased by reactions of the beam with H in the target.

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Table 1: Calculated and experimental isotope yields for various beams on H_2O targets

			Yield (mCi/μA per 10 min)[1]				
			15-Mev p		75-MeV ^{3}He		300-MeV ^{12}C
Isotope	$t\frac{1}{2}$ (m)	E_β(MeV)	Experiment	Theory[2]	Experiment	Theory[2]	Theory[2]
^{13}N	9.96	1.20	20	31	96	81	26
^{11}C	20.4	0.96	0	0	< 1[3]	51	8.2
^{15}O	2.03	1.73	0	0	67	384	109
^{18}F	109.8	0.63	0.2	0.1	12	5.5	4.2

[1]Assuming beam stops in the target.

[2]Calculated using the ALICE compound nucleur particle evaporation code (Reference 3).

[3]None observed.

Figure Captions

Fig. 1. Calculated isotope yields from proton bombardments with incident beam energy. No ^{11}C is predicted.

Fig. 2. Calculated isotope yields from ^3He bombardments with incident beam energy.

Fig. 3. Calculated isotope yields from ^{12}C bombardments with incident beam energy.

Fig. 4. Typical HPLC determination of the chemical composition of a source produced by proton bombardment. ^{18}F, while produced by the conditions of bombardment, does not elute from the HPLC column.

Fig. 5. Time decay curves of raw source from ^3He bombardment. The observed decay of the source (- data) and the fit (0 fit) resulting from the summation of the decay patterns of the individual isotopes present (^{15}O, ^{13}N and ^{18}F) are indicated.

Fig. 6a). Chemical composition of raw source, as determined by HPLC separation.

6b). Chemical composition of purified source, as determined by HPLC separation.

Fig. 7. Time decay curve of purified source. The observed decay of the source (- data) and the fit (0 fit) resulting from the summation of the decay patterns of the individual isotopes present (^{13}N and ^{18}F) are indicated.
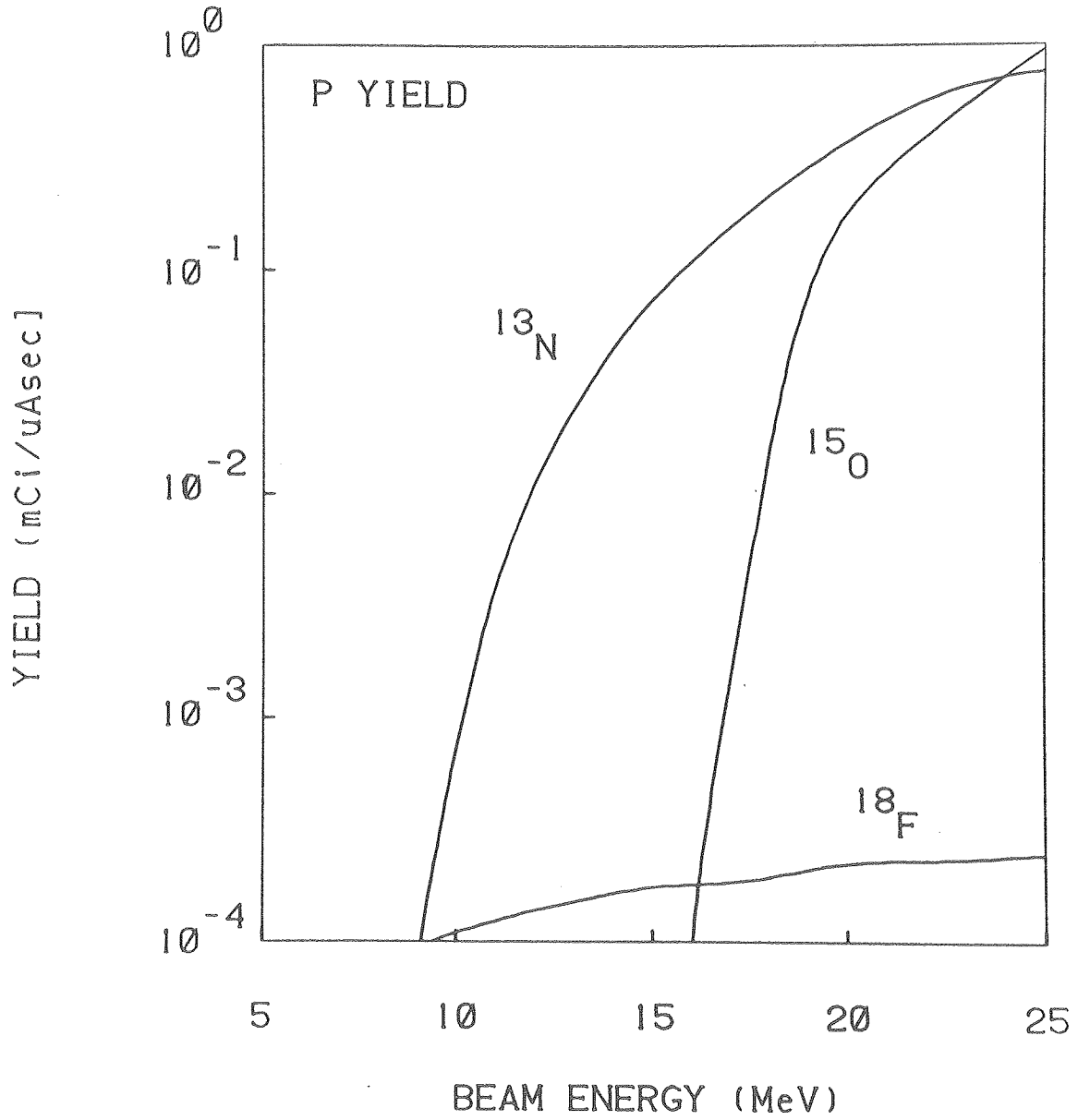
Fig. 1

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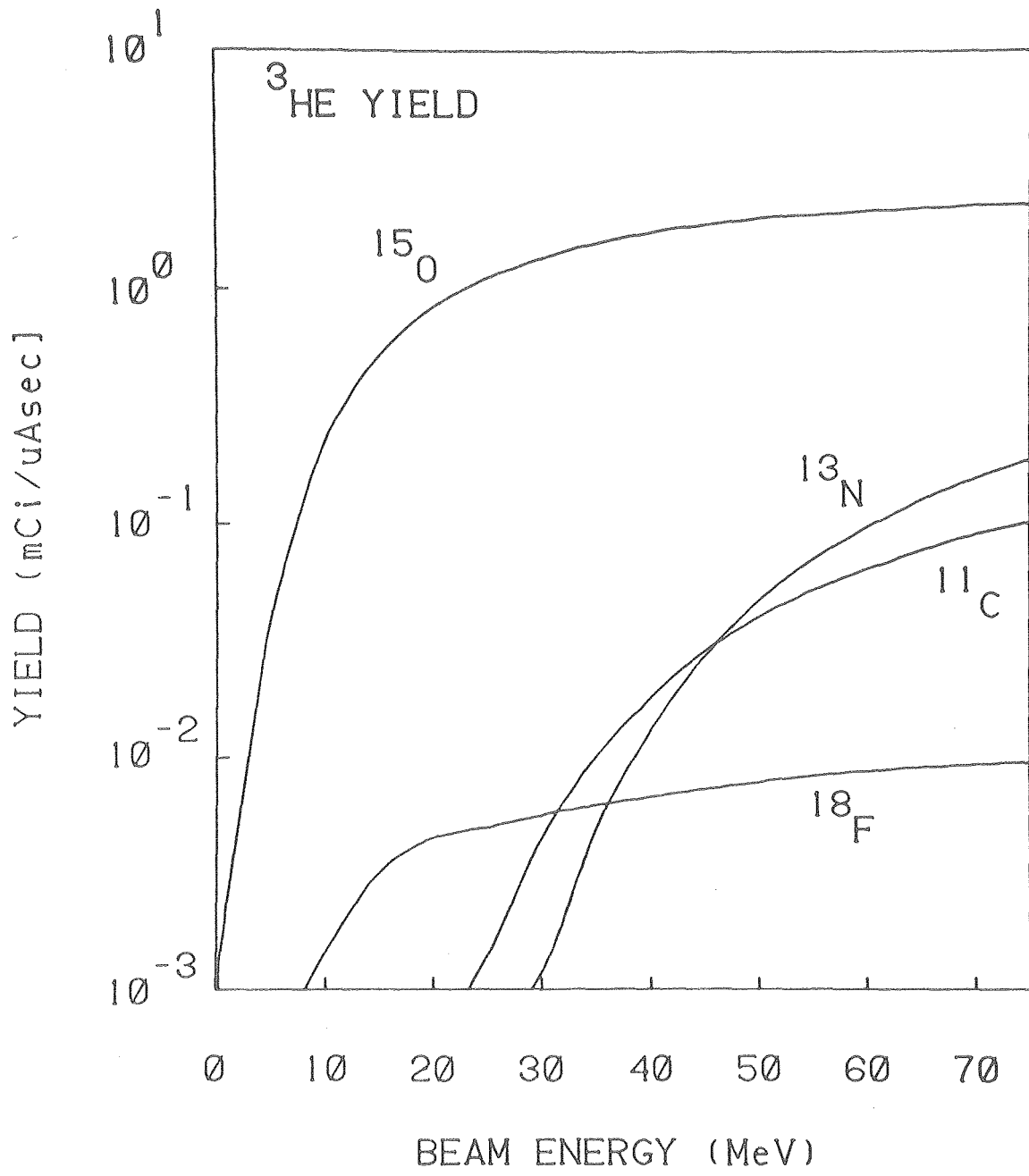
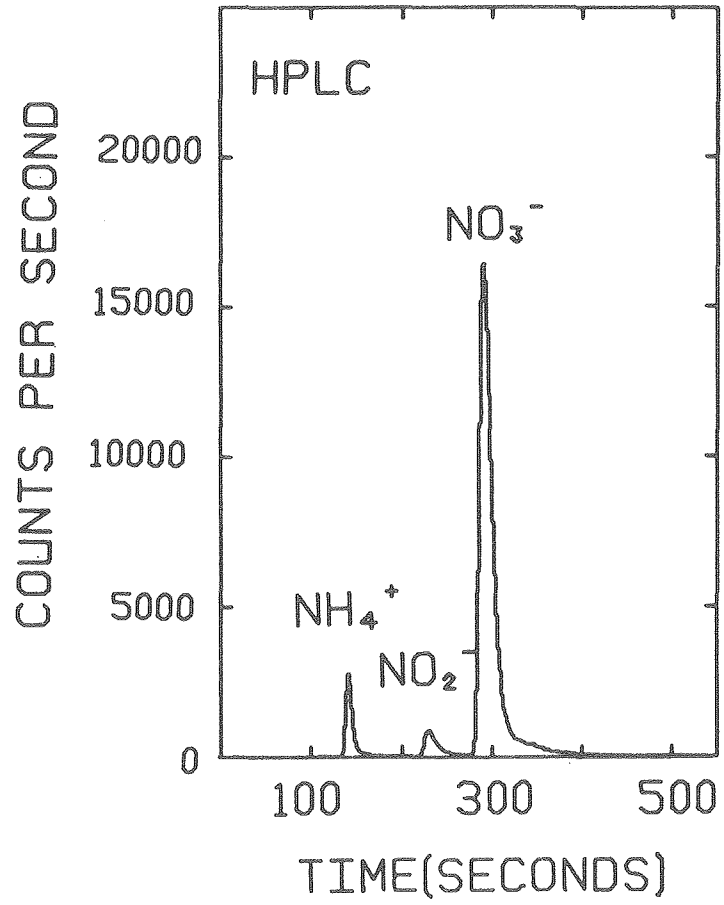Fig. 2
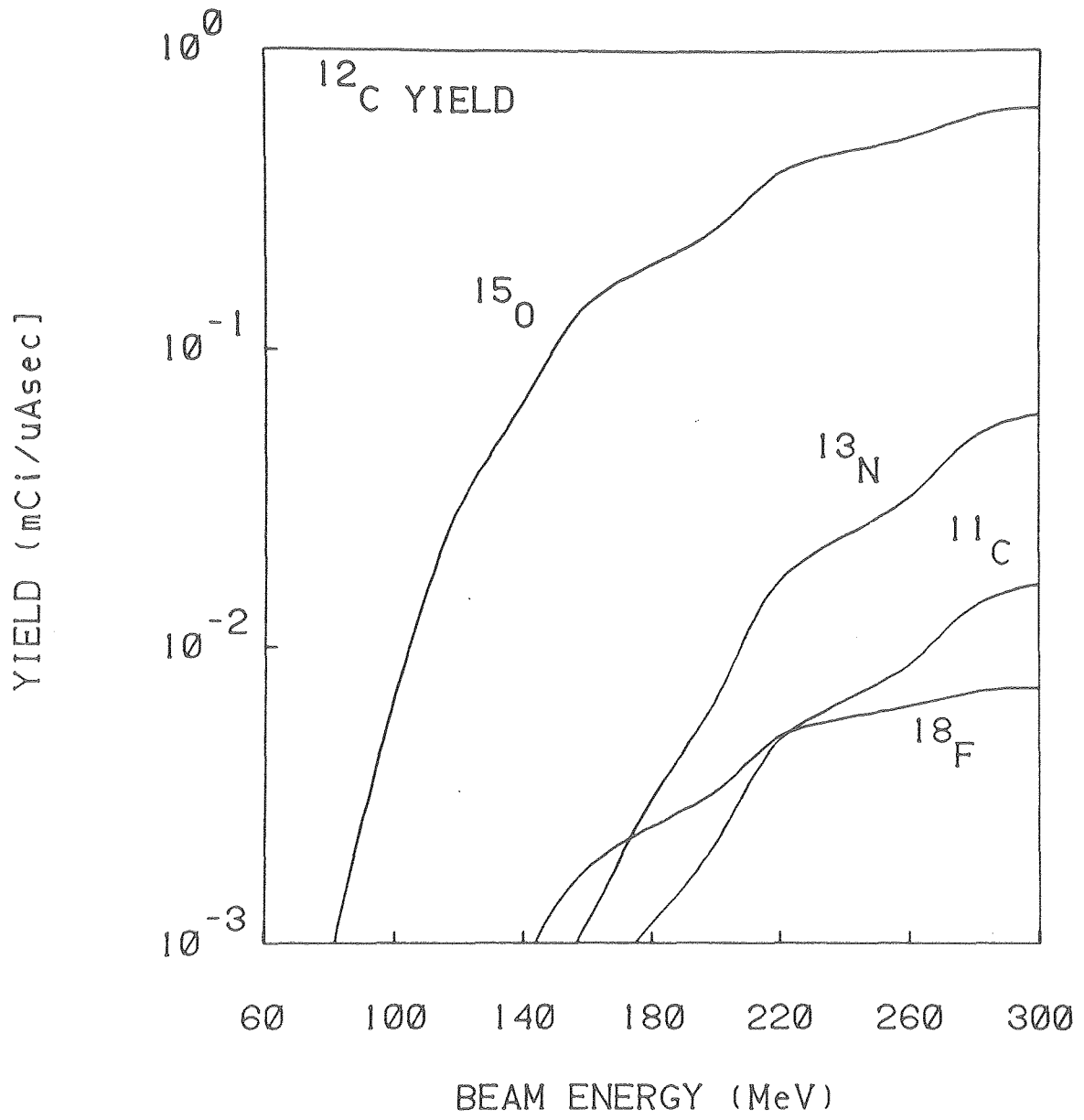
Fig. 3

HPLC SEPARATION OF ^{13}N IONS

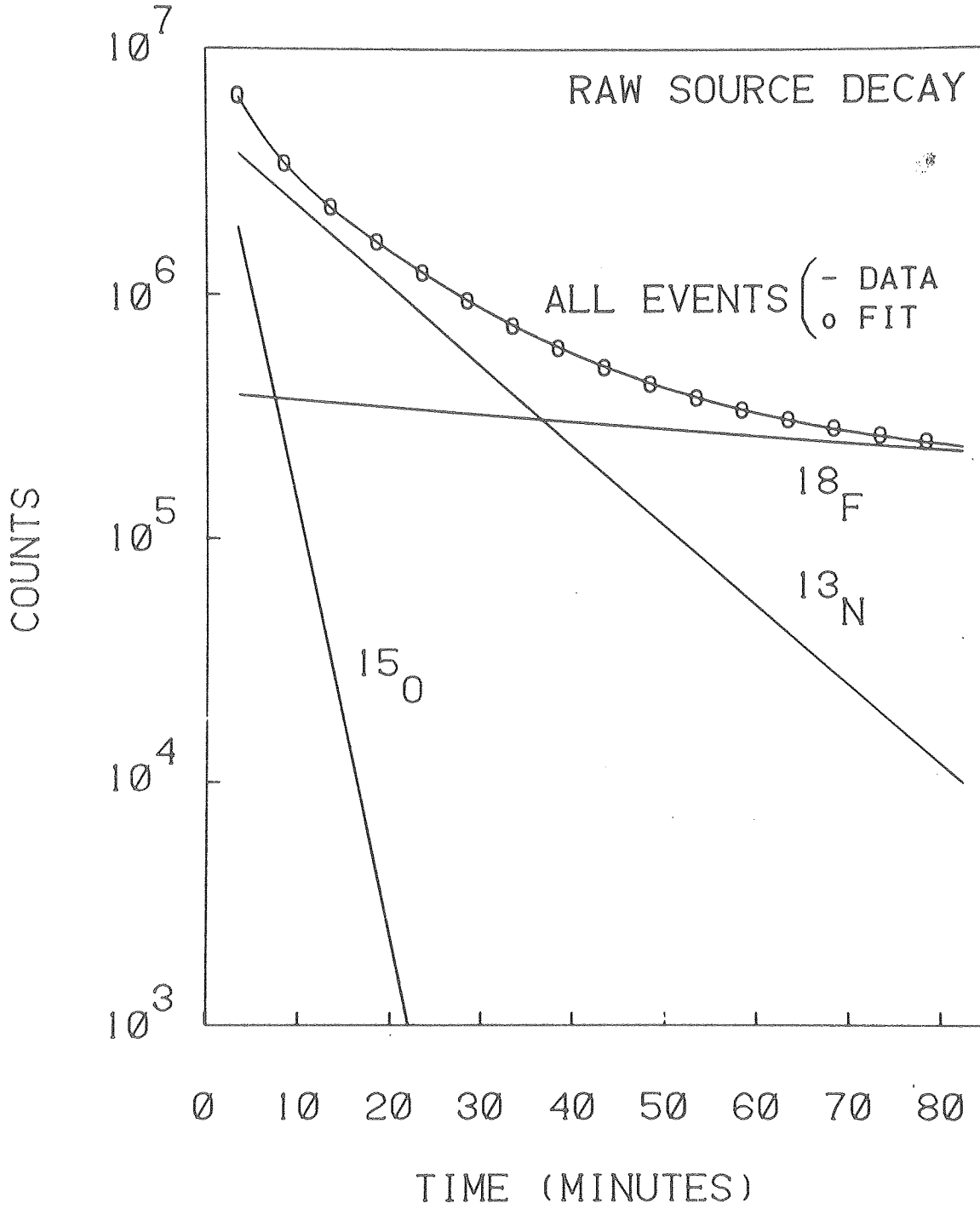Fig. 4
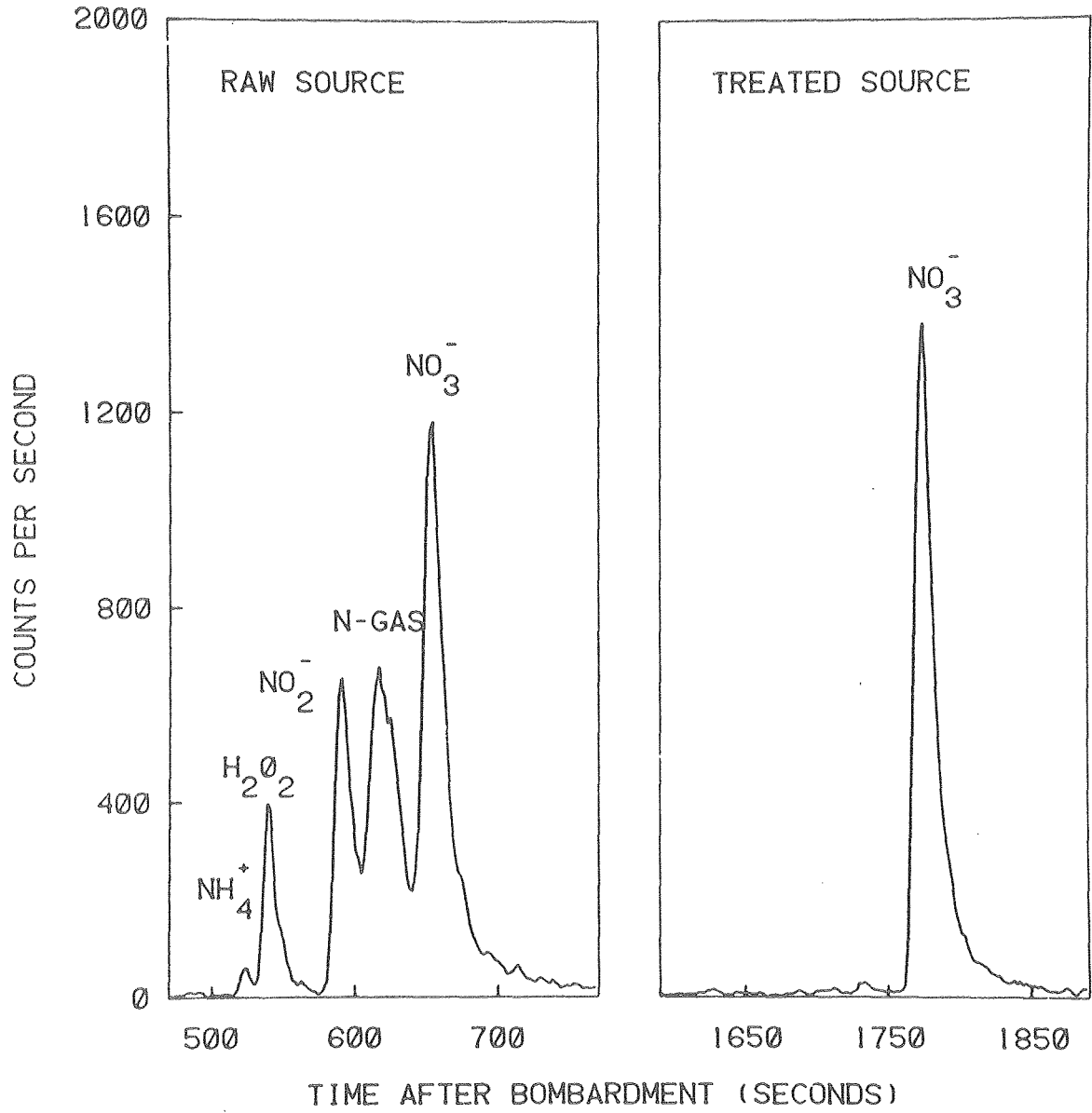
Fig. 5

Fig. 6

XBL 804-9287

Fig. 7