

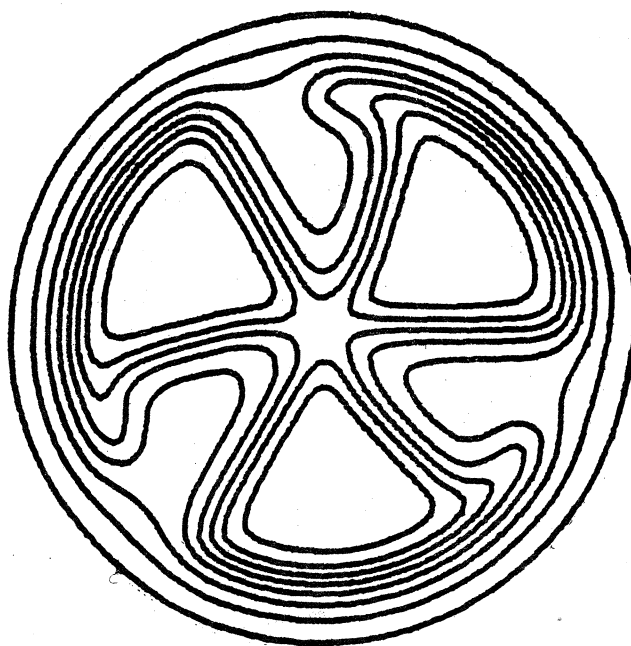
MICHIGAN STATE UNIVERSITY

CYCLOTRON LABORATORY

A BATCH PROCESS FOR THE PRODUCTION OF

^{13}N -LABELED NITROGEN GAS

SAM M. AUSTIN, AARON GALONSKY, J. BORTINS
and C. PETER WOLK



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Sam. M. Austin, Aaron Galonsky, and J. Bortins*

Cyclotron Laboratory, Physics Department[†]
Michigan State University, East Lansing, Michigan 48824

and

C. Peter Wolk

MSU/AEC Plant Research Laboratory[‡]
Michigan State University, East Lansing, Michigan 48824

ABSTRACT

A technique has been developed for generating ^{13}N ^{14}N gas with specific activity ~ 20 mCi/ml. The ^{13}N is produced, nearly free of other radioactivities, in the $^{13}\text{C}(p,n)^{13}\text{N}$ reaction using 17 mg amorphous carbon targets enriched to about 97% ^{13}C and a proton bombarding energy of 11 MeV. The irradiated material is converted to $^{13}\text{N}^{14}\text{N}$ gas by automated Dumas combustion and compressed into 1 ml vials, the entire procedure taking less than 15 minutes. A number of biological tests have shown that the $^{13}\text{N}^{14}\text{N}$ gas is essentially free of other labeled compounds of nitrogen. The advantages and disadvantages of other ^{13}N -producing nuclear reactions are discussed.

* Present address: School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota, 55414.

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1. INTRODUCTION

Available techniques¹⁻⁷ for the production of ^{13}N -labeled nitrogen gas ($^{13}\text{N}^{14}\text{N}$ and $^{13}\text{N}^{13}\text{N}$) involve continuous-flow processes in which the radioactivity is either produced in a flowing target gas² or is swept from a solid target by a carrier gas.³⁻⁷ Such techniques have yielded labeled gases with rather low specific activities, typically less than 1 mCi/ml. Activities of this magnitude, or less, are satisfactory for many purposes. However, in some experiments (e.g. autoradiographic studies⁸ of the sites of fixed ^{13}N) the nature of the analytic procedure may limit the amount of biological material. In other experiments (e.g. studies⁹ of the early metabolism of fixed ^{13}N) the fixation time may be limited so as to study the time dependence of the process. Larger specific activities are often required to obtain usable amounts of fixed labeled nitrogen in such cases.

The batch process described in this paper was developed to satisfy our requirement of high specific activity, high purity ^{13}N -labeled nitrogen gas for use in studies of the fixation and metabolism of nitrogen by certain blue-green algae.^{8,9} Briefly, the procedure is as follows. A ^{13}C target is bombarded with 11 MeV protons and ^{13}N is produced by the $^{13}\text{C}(p,n)^{13}\text{N}$ reaction. The irradiated material is removed from the sample holder, is subjected to automated Dumas combustion^{10,11} and the resulting gas is compressed into a 1 ml vial. The gas is found to be essentially free of radioactive constituents other than $^{13}\text{N}^{14}\text{N}$ or $^{13}\text{N}^{13}\text{N}$ and to have a total radioactivity of up to 20 mCi for a 1 μA bombarding current.

Activities of this magnitude have been obtained by one other technique,³ but the procedure required very high beam power (60 μ A of 14 MeV deuterons) and can therefore be carried out only at a few rather specialized installations. With the present technique, it is possible to achieve comparable activities in any laboratory where a proton accelerator with an energy of at least 7 MeV and a beam current of about 1 or 2 μ A is available.

2. COMPARISON OF REACTIONS LEADING TO ^{13}N

The $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$, $^{16}\text{O}(\text{p},\alpha)^{13}\text{N}$, $^{14}\text{N}(\text{p},\text{pn})^{13}\text{N}$ and $^{12}\text{C}(\text{p},\gamma)^{13}\text{N}$ reactions have all been used previously for the production of labeled compounds of nitrogen.^{1-7,12} In order to compare these reactions with the $^{13}\text{C}(\text{p},\text{n})^{13}\text{N}$ reaction we have calculated the yields expected from bombardment of a $\frac{1}{2}$ inch thick target of the pure isotope. Cross sections were taken from the literature¹³ and the variation of stopping power with energy was taken into account. Further details of the calculations are given in ref. 14. Some of the results are shown in Fig. 1.

For a variety of reasons the choice among these reactions reduces to one between $^{13}\text{C}(\text{p},\text{n})^{13}\text{N}$ and $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$. The yield of the $^{12}\text{C}(\text{p},\gamma)^{13}\text{N}$ reaction is much smaller than that of the other reactions and no compensating advantages are apparent.⁷ The $^{16}\text{O}(\text{p},\alpha)^{13}\text{N}$ reaction is also less prolific than the others shown in Fig. 1, a disadvantage which is compounded if one wishes to use a solid target such as Li_2CO_3 where the other atoms provide additional stopping power but no ^{13}N yield. In

addition, it is found¹⁵ that a substantial fraction of the activity is in the form of oxides of nitrogen, placing rigorous demands on the following purification process if one wishes to have pure labeled N_2 gas.

The $^{14}\text{N}(\text{p},\text{pn})^{13}\text{N}$ reaction, while capable of producing amounts of activity¹² comparable to the other reactions at energies of 20 MeV and above, has two flaws. Firstly, solid targets which contain substantial amounts of nitrogen and also tolerate large beams are not available.⁴ The use of gaseous $^{14}\text{N}_2$ targets yields a gas with a relatively low ratio of $^{13}\text{N}^{14}\text{N}$ to $^{14}\text{N}_2$ which tends to reduce the amount of fixed $^{13}\text{N}^{14}\text{N}$. Secondly, the threshold energy for the $^{14}\text{N}(\text{p},\alpha)^{11}\text{C}$ reaction is only 3.13 MeV so that substantial amounts of radioactive $^{11}\text{CO}_2$ and ^{11}CO contaminants are produced.¹

Both the $^{13}\text{C}(\text{p},\text{n})^{13}\text{N}$ and $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ reactions are essentially free of the disadvantages discussed above. The yields are relatively large and convenient solid targets of nearly pure ^{12}C and ^{13}C are available. Table I shows the thresholds for the reactions $^{13}\text{C}+\text{p}$ and $^{12}\text{C}+\text{d}$ leading to all other radioactive products with $\tau_{1/2} > 1$ sec. The most important of these is ^{11}C since its half-life is similar to that of ^{13}N ; other products either decay so rapidly (e.g., ^{11}Be , ^{10}C) that they disappear during the processing time, or so slowly (e.g., ^7Be , ^{10}Be) that they contribute negligible activities. At bombarding energies less than 16.36 MeV for $^{13}\text{C}+\text{p}$ and 14.56 MeV for $^{12}\text{C}+\text{d}$, one is below threshold for producing any $\tau_{1/2} > 1$ sec. radioactivity other than ^{13}N .

* The percentage reduction depends on whether or not the total nitrogen partial pressure saturates the fixation ability of the organism involved (see ref. 8).

the irradiated ^{13}C for further processing: one simply punches through the front Al foil, the target material, and the back Al foil, forcing all material directly into a combustion tube. The gas-tight seal serves to confine in the target cell any gaseous radioactive products which might be formed during the bombardment process, either from ^{13}C or from N_2 absorbed on the target material.

Those parts of the target cell which were struck by the beam were made of pure aluminum to minimize long lived radioactivities from the target cell. The target material was amorphous carbon, usually enriched to 90 or 97 atom % in ^{13}C ,* which had an unpacked density of about 0.3 g/cm.³ Some of the early data was taken with 60% ^{13}C targets.

4. PROCESSING OF IRRADIATED MATERIAL

Following irradiation and ejection into the combustion tube, the target material is mixed with 0.6 ml of fine CuO powder and 0.18 mg of KNO_3 (the latter to provide carrier nitrogen) and is subjected to an automated Dumas combustion procedure using a slightly modified Coleman Model 29 Nitrogen Analyzer.† Similar procedures have been described previously^{10,11} and we give only a brief description here. The process consists of combustion

* The 90% enriched material was obtained from Prochem, 8 Pinebrook Road, P.O. Box 555, Lincoln Park, N.J. 07035 and the 97% from Monsanto Research Corporation, Mound Laboratories, Miamisburg, Ohio.

† Purchased from Coleman Instruments Corp., Maywood, Ill., U.S.A.

Our choice of the $^{13}\text{C}(p,n)^{13}\text{N}$ reaction was based on the larger yield and on the fact that proton beams from our accelerator are available a larger fraction of the time than deuteron beams. The cost of the target is surprisingly low, about \$3.30 for a target enriched to 97 atom-% in ^{13}C .

3. IRRADIATION PROCEDURES

The irradiations were carried out with 1 to 4 μA beams of protons from the Michigan State University sector-focussed cyclotron. Protons entered the target cell after passing through a 28 μm thick Havar window at the end of the beam pipe and 2.5 cm of air. The target cell was transported to and from the beam vault in about 5 sec using a pneumatic shuttle¹⁶ system which also provided cooling air for the target during irradiation. Irradiations usually lasted 20 to 30 min, i.e. two to three half-lives of ^{13}N .

The target cell is shown in Fig. 2. The ^{13}C target is a powder contained in a cylindrical hole with a diameter of about 4.1 mm, as small as is consistent with beam size and multiple scattering in energy degraders and in the ^{13}C . The length of the cell, 4.4 mm, and the total amount of target material, 17 mg, were fixed by the density of the target material and by the requirement that the beam leave the target with an energy of about 4 MeV to permit simple current integration. The target material is confined by 50 μm thick aluminum foils, pressed against silicon-rubber O-rings by the compression-nut, washer configuration shown in the figure. This arrangement permits rapid removal of

at 800°C, of the sample packed with cupric oxide in a quartz tube. Following purging of the system with CO₂ and combustion, CO₂ is used to sweep the combustion products through postheater tubes filled with cupric oxide and copper. In this process oxides are first formed and then reduced, and N₂ is formed. The resultant products then pass through a liquid nitrogen trap which, in addition to freezing out the CO₂, reduces further the concentration of any oxides of nitrogen. Finally the gases enter a previously evacuated Toeppler pump. The gaseous nitrogen (0.02 ml STP) is then compressed into a 1 ml serum vial and the vial is filled to slightly above atmospheric pressure with an inert gas (typically 1% CO₂, 99% Ar) to prevent leakage of atmospheric nitrogen into the vial. Much of this process is carried out automatically by the Coleman Analyzer. The combustion tubes are repacked with fresh CuO following each combustion. The postheater tubes are normally repacked when oxidized or when contaminated with air after several weeks of inactivity. See Table II for further details.

Our requirements were maximum purity of the product gas and freedom from oxides containing radioactive nitrogen. The compound ¹³N₂ is of particular concern because it has a relatively low freezing point and might not be efficiently removed by the liquid nitrogen trap. In order to investigate the sensitivity of the oxide transmission to the parameters of the combustion system, 0.73 mg samples of K¹⁵NO₃ (enriched to 99 atom % in ¹⁵N) were combusted and the gaseous products of the combustion procedure were analyzed using a mass spectrometer. About 0.015% of the total gas produced (predominantly ¹⁵N₂) was in a mass 31 peak.

This peak corresponded to ¹⁵NO as was demonstrated by its absence when K¹⁴NO₃ was processed in an identical fashion. The amount of ¹⁵NO present was found to be essentially independent of oven temperature, so midrange settings were chosen for all ovens.

Because of the nature of the process, some exposure to radiation was inevitable. With reasonable care, whole-body exposures could be kept to about 2 mrem/batch during preparation of the sample for combustion and 4 mrem/batch for the remainder of the process. Substantial reductions in these exposures could be achieved if warranted by the frequency of experiments.

5. RESULTS

It was found that approximately the expected amount of ¹³N activity was present in the irradiated sample and that, corrected for the time consumed in the process (typically 12-15 min from the end of bombardment), a large fraction (4/5) of this activity could be converted to ¹³N¹⁴N gas. The efficiency of the procedure appeared to be crucially dependent on thorough mixing of the ¹³C with the CuO powder prior to combustion. The results of a typical run with a current of 1 μA are summarized in Table III.

In order to determine the isotopic purity of the irradiation products, the disintegration rate was measured as a function of time, with the results shown in Figs. 4-6. The curves in Figs. 4 and 5 are for the unprocessed target material from irradiation with approximately 1 μA beam, of targets enriched to 90 atom% in ¹³C. The decay curve immediately following bombardment was

fraction of possible long-lived activities were obtained from a least-squares fit which allowed two activities, one with a half life of 9.96 min (^{13}N) and another with a half life of either 20.4 min (^{11}C) or 60 min. It was found that at the end of the processing period, 13 minutes after completion of the irradiation, the relative decay rates of ^{13}N and the longer-lived activities were $^{13}\text{N}/^{11}\text{C} = (2.5 \pm 1.3) \times 10^5$ and $^{13}\text{N}/X(t_{1/2} = 60 \text{ min}) = (9.4 \pm 4) \times 10^7$.

While the isotopic purity of the radioactivity is therefore more than adequate for most needs, the preferential absorption by some biological systems of compounds other than $^{13}\text{N}^{14}\text{N}$ requires that an estimate of their abundance be made. Based on the results of the mass spectrometer experiments described in Section 4, the concentration of $^{13}\text{N}_2\text{O}$, the compound most likely to survive our purification process, should be about 10^{-4} that of $^{14}\text{N}^{13}\text{N}$, even in the unlikely event that all the ^{13}N were initially present in oxygen-containing compounds. Thus we expect impurities to be unimportant in most cases. We have, however, made a series of tests involving the organism actually used in our experiments and have described them in detail elsewhere.⁸ The procedure was to compress the radioactive gas into two 1 ml vials instead of one, and to add the biological material to both vials. One of the samples was then treated in the usual way. The procedure for the other, however, was altered so as to prevent the assimilation of ^{13}N as $^{13}\text{N}^{14}\text{N}$ or $^{13}\text{N}^{13}\text{N}$ but to permit its assimilation in other forms. It was found that at most 1-3% of the fixed nitrogen was obtained from compounds other than $^{13}\text{N}^{14}\text{N}$.

Finally, it is perhaps worth noting that even if the processed gas initially contains ^{13}N only in the form of nitrogen gas, decay

obtained with an ionization-chamber radiation monitor placed 20 cm from the sample and is shown in Fig. 4. It is clear that even for the shortest times the decay is dominated by the $t_{1/2} = 9.96$ min activity of ^{13}N . While there is some indication of longer lived activities for $t > 5t_{1/2}$, deviations from the $t_{1/2} = 9.96$ min curve are not larger than uncertainties in the ion-chamber reading for such low intensities. However, at much later times long-lived activities definitely do appear in the unprocessed sample as shown in Fig. 5. These activities do not seem to be dominated by a single half-life. Presumably they arise from reactions on impurities in the C target and in the small pieces of Al window which are punched out with the carbon sample.

The important point to note, however, is that these activities do not survive the processing described in section 4. Figure 6 shows the decay curve of the final gaseous sample. After processing, the 1 ml serum vial was filled to 1 atm with a scintillating liquid, placed in scintillation vials filled with the same liquid and then counted in a Beckman CPM-100 liquid scintillation counter* set to count ^{32}P . The same instrument was used to obtain the data in Fig. 5.

No appreciable long lived activities are visible by eye at least up to 30 half lives of ^{13}N and all of the data are very well fitted by assuming a single activity with the ^{13}N half-life of 9.96 min ($\chi^2 = 13.9$ for 13 degrees of freedom). Estimates of the

* Purchased from Beckman Instruments, Inc., Fullerton, California. The scintillation liquid was Beckman Cocktail D.

of the ^{13}N can lead to the formation of labeled impurities. This occurs because a certain fraction of the gas is in the form $^{13}\text{N}^{13}\text{N}$. When one nucleus of such a molecule decays, the remaining ^{13}N atom might react with the surrounding material, forming labeled compounds (e.g., oxides) which could be assimilated. The importance of this contamination must be considered on an individual basis. Thus, in our experiments, we were particularly concerned about labeled oxides such as ^{13}NO , so that care was taken to eliminate O_2 from the reaction vials. One can estimate a lower limit for the effect as follows. Assuming random pairing, the ratio of $^{13}\text{N}^{13}\text{N}$ to $^{13}\text{N}^{14}\text{N}$ is $1/2(n_{13}/n_{14})$ where n_{13} and n_{14} are the numbers of atoms of ^{13}N and ^{14}N simultaneously present in the postheaters of the Coleman Analyzer. At 20 mCi/ml, 0.02 atm N_2 in the vial, these numbers are approximately $n_{13}=6.4 \times 10^{11}$ and $n_{14}=1.1 \times 10^{18}$ so that the ratio is at least 2.9×10^{-7} . Although this estimate may be lower than the actual value because random pairing is not always obtained in the Coleman Analyzer, it is small enough so that the problem will not often be serious.

6. SUMMARY

A technique has been developed for the production and partially automated purification of $^{13}\text{N}^{14}\text{N}$ using the $^{13}\text{C}(\text{p},\text{n})^{13}\text{N}$ reaction followed by a Dumas combustion incorporating a Coleman Nitrogen Analyzer. Specific activities of 20 mCi/ml are obtained for a 1 μA , 11 MeV proton beam and the gas produced is essentially free both

of radioactivities other than ^{13}N and of ^{13}N -labeled compounds other than $^{13}\text{N}^{14}\text{N}$. The process can be used anywhere that there is an accelerator capable of producing a reasonably intense ($\sim 1 \mu\text{A}$) proton beam with energy $\gtrsim 7$ Mev. If only lower energy beams are available, the $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ reaction coupled with the purification procedure described in this paper should also yield high quality labeled nitrogen gas, but higher beam intensity will be required to produce comparable ^{13}N activity. Application of the techniques described here with higher intensity beams available elsewhere should yield labeled nitrogen with specific activities greater by an order of magnitude than those presently available.

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Table I. Threshold energies for reactions on ^{12}C , ^{13}C , ^{14}N and ^{16}O

Product (half-life) ^a	Threshold Energies (MeV) ^{b,c}		
	$^{12}\text{C}+\text{d}$	$^{13}\text{C}+\text{p}$	$^{14}\text{N}+\text{p}$ $^{16}\text{O}+\text{p}$
^3H (12.3 y)	14.56	16.36	
^7Be (53.4 day)	20.49	21.83	
^{10}C (19.4 sec)	29.88	30.50	
^{10}Be (1.6×10^6 y)	25.33	26.31	
^{11}C (20.4 min)	14.56	16.36	3.13
^{11}Be (13.7 sec)	33.76	34.08	
^{13}N (9.96 min)	0.328	3.24	8.93
			5.55

a. From ref. 17.

b. Calculated from the mass excesses given in ref. 18.

c. The threshold energy given is that for the least endoergic reaction leading to each nuclide.

Table II.--Processing procedure

Stage of Process	Time	Comments
Purge 1 [*]	30 sec.	CO_2 flow rate = 300 ml-STP/min
Purge 2 [*]	~60 sec.	Total flow of 30 ml-STP in three 10 ml batches.
Preheat and Combustion [*]	350 sec.	Combustion tube isolated from remainder of system.
Sweep [*]	100 sec.	CO_2 flow rate = 30 ml-STP/min. Nitrogen gas expands into 200 ml volume Toepller pump.
Compression		Nitrogen gas compressed into 1 ml vial by forcing mercury into 200 ml bulb of Toepller pump.
Fill		Vial is filled to atmospheric pressure with 1% CO_2 -99% Ar.

* These steps are carried out automatically by the Coleman Analyzer.

Table III.--Results of a typical run with 1 μ A beam

Location	Radioactivity (mCi)
Unprocessed target	95
Combustion tube at beginning of combustion	75
Combustion tube after combustion	5 ^{*a}
Liquid N ₂ trap	10 [#]
Dead volume ^b	20 [*]
1 ml vial	40 [*]
1 ml vial (at end of processing)	16 ^c

- a) The numbers marked with an asterisk have been corrected for decay from the beginning of combustion. Thus rows 3-6 should sum to row 2. The numbers are not the result of an individual run, but a summary of typical experience.
- b) Dead volume is the volume connected to 1 ml vial at the time of compression.
- c) Calculated for a typical processing time of 13 min.

FIGURE CAPTIONS

Fig. 1-- Production of ^{13}N by several nuclear reactions. The thick sample is presumed to be bombarded for a time long compared to the ^{13}N half-life, so the saturation activity is attained; the activity is then obtained from the production rate by division by 3.7×10^7 disintegrations $\text{sec}^{-1} \text{mCi}^{-1}$. The vertical bars on the curves mark the threshold energies for the production of ^{11}C .

Fig. 2-- Schematic diagram of the target cell. Parts of the cell which could be struck by the beam were constructed of pure aluminum to minimize the production of long-lived activities.

Fig. 3-- Schematic diagram of the sample processing apparatus. The portion enclosed in the dashed line is the Coleman Nitrogen Analyzer. Combustion and postheater tubes are 10 mm o.d., 8 mm i.d. quartz tubes. The combustion tube contains the sample packed in 30 cm column of CuO (Coleman "Cuprox"). The first (second) postheater tube contains 8 cm (13 cm) of Cuprox and 9.5 cm (20 cm) of Cu (Coleman "Cuprin"). These ovens are at the temperatures shown on the figure. The plumbing is mainly glass and some of the valves shown are actually stopcocks. Regions shown as were constructed to obtain a minimum total volume. The valve marked with

an * was opened only after the region had been pressurized to above atmospheric pressure. For further details of the purification process see the text and Table II.

Fig. 4-- Decay curve for the unprocessed target material at short times. Time is measured from the end of the irradiation. The curve corresponds to a halflife of 9.96 min. The uncertainty in the ionization gauge reading is about 10% over most of the range, increasing to about 20% near the end of the range.

Fig. 5-- Decay curve for the unprocessed target material at long times. Time is measured from the end of the irradiation. The dashed curve is drawn to guide the eye. The variation with time expected for half lives of 9.96 and 20.4 min is also shown.

Fig. 6-- Decay curve for the processed ^{13}N -labeled nitrogen gas contained in a 1 ml vial. Time is measured from the end of the irradiation. Uncertainties are shown where they are larger than the plotted points. The background was measured during the interval 363 to 685 min and was constant within statistics during that time. The curves are, in order of the legend, for the half life of ^{13}N , 9.96 min; for the best fit assuming only ^{13}N and ^{11}C ($\tau_{1/2}=20.4$ min) contribute; and for the best fit assuming only ^{13}N and a contaminant with a half life of 60.0 min.

