Production of ^{13}N radioactive nuclei from ^{13}C(p,n) or ^{16}O(p,α) reactions

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Total cross sections for the $p+{}^{13}$C and $p+{}^{16}$O reactions were measured by the activation method up to 30 MeV. Yields of ^{13}N and of other produced nuclei (^{11}C,^{15}O) were obtained.

I. INTRODUCTION

A project aiming at the production and acceleration of intense radioactive ion beams has been recently started at Louvain-la-Neuve.[1] Among the light radioactive ions, ^{13}N is of particular interest for the astrophysicists: the ^{13}N(p,γ)^{14}O reaction is indeed the first reaction involving radioactive species in the hot CNO cycle.[2] In the present work the total cross section for ^{13}N production was measured using the activation method, by two different ways, i.e., the ^{13}C(p,n) and the ^{16}O(p,α) reactions, up to a proton energy of 30 MeV in the laboratory system. The goal of this work was not to scan the excitation function in very small energy steps as should have been done by people interested in the level structure of the compound nucleus; we wanted rather to measure only the shape of the excitation functions in order to calculate the ^{13}N yields. Cross sections and yields of other radioactive nuclei (^{11}C, ^{15}O) produced in the $p+{}^{13}$C and $p+{}^{16}$O reactions were also obtained.

The ^{13}C(p,n)^{13}N reaction had been previously studied from threshold (3.2 MeV) to 13 MeV (Refs. 3–5) and at 16.3 and 22.8 MeV;[6] the last work was the only one using the activation method. The ^{16}O(p,α)^{13}N reaction had been measured from 6 to 16 MeV (Refs. 7, 8) and from 12 to 18 MeV.[9]

II. EXPERIMENTAL METHOD

A. Beam and targets

The Louvain-la-Neuve isochronous cyclotron (CYCLONE) delivered a 100-nA proton beam which was focused on a solid (^{13}C) or gas (^{16}O) target. The solid target consisted of a precisely weighed amount of ^{13}C powder (99.9%) uniformly pressed between two 3-cm diameter and 0.5-mm-thick aluminum windows; the typical ^{13}C areal density was 20 mg/cm^2. The gas target was a 2-mm-thick stainless-steel cylinder of 4-cm diameter and 2-cm length closed by two 1-mm-thick aluminum windows of purity larger than 99%; the windows were maintained towards the cell body by stainless-steel rings via 1-mm-diameter indium wires; the rings were bolted together. The typical gas pressure was 10 bar. Aluminum degraders could be placed immediately in front of the solid or the gas target to easily change the proton beam energy. Behind the target, an aluminum beam stopper collected the protons to monitor the beam intensity. A guard-ring (-2.8 kV) repelled the secondary electrons towards the beam stopper.

B. Principle of the measurement and procedure

^{13}N is a positron-emitter nucleus with a half-life of 9.96 min and a maximum positron energy of 1.2 MeV. The measurement consisted in detecting off-line the two 511-keV γ rays emitted at opposite angles after positron annihilation. This method was chosen to extract the source signal from the hot radioactive environment. The γ-ray detectors were two 6.3-cm-diameter and 10-cm-thick NE102 plastic scintillators coupled to fast XP2020 photomultipliers and placed at equal distance from the source. Coincident pulses were required and counted in a multiscaling mode. The lifetimes measurement was used to select the radioactive products. At each energy, the irradiation time was 10 min. After the irradiation, two different procedures were used: (1) The ^{13}C powder was taken out of the aluminum container and was put in a glass vessel which was then placed between the two detectors outside the beam area, and (2) the gas target was counted in the beam line from a few minutes after the end of irradiation, in order to allow for the decrease of the aluminum beam-stopper activity. Aluminum was indeed chosen for the short-lived isotopes which were produced.

The counting time was longer than 1 h at each energy. The multichannel spectrum was fitted with a sum of exponentials, using the MINUIT code.[10] The chi square was always within the calculated expectation. For the $p+{}^{13}$C reaction, at most two radioactive nuclei were seen, i.e., ^{13}N and ^{11}C (20 min); for the $p+{}^{16}$O reaction, 3 lifetimes had to be introduced above 18 MeV, i.e., ^{15}O (2 min), ^{13}N, and ^{11}C.

C. Check of a possible ^{13}N escape

During the transfer of the irradiated ^{13}C powder (procedure 1, mentioned previously), some ^{13}N loss could possibly occur. At two energies (11.5 and 18.5 MeV), the irradiated ^{13}C powder was left in its frame and counted in the beam line; the powder was then contained between two 4-cm-diameter and 1-mm-thick beryllium windows. The beam stopper was made also of beryllium. Before the counting started, a 1-mm-thick copper plate was moved down on each side of the target to keep the positron annihilation very close to the target volume.

D. Data

The fit provided the amplitude of the exponentials at the beginning of the counting time and an extrapolation permitted us to find the saturation activity. Thereafter, knowing the target thickness and the integrated charge, one could calculate the cross section, providing the detection efficiency was determined. This last factor was measured as follows. (a) For the solid target, the glass bottle containing the ^{13}C powder during the counting time was thick enough to stop ^{13}N and ^{11}C positrons (1.2 and 1.0 MeV maximal energy, respectively). The efficiency was measured by putting in a similar bottle a small amount (about 100 μCi) of ^{18}F in a solution representing approximately the same volume as the ^{13}C powder, and by counting the ^{18}F activity successively with a Ge-Li detector of known efficiency and with the two plastic detectors. (b) For the gas target, the situation was slightly more complicated because of its large volume. It was assumed that owing to the low areal density of the gas, all the positrons annihilated on the cell surface, mostly in the thick stainless steel part. The entrance and exit aluminum windows were indeed not very effective because γ rays emitted from the windows have to cross a much larger amount of material in order to reach the detectors, i.e., at least part of the surface of the window plus the 1-cm-thick stainless steel rings. A calibrated ^{22}Na source was thus successively put all around the cell surface, and a mean efficiency was measured. This mean value coincided within 5% of a Monte Carlo calculated efficiency in which positrons emitted isotropically from the gas volume annihilated within the stainless steel surface or within the aluminum windows, or crossed the windows and were lost; annihilation γ rays were subsequently tracked down to the detectors. (c) For the solid target used in Sec. II C, the efficiency was measured by counting a calibrated ^{22}Na source located at the beam-spot place between the Be and Cu windows.

The cross sections for the production of ^{13}N are presented in Figs. 1 and 2. Statistical error bars (typically less than 1%) are always contained in the size of the data points. An additional 10% systematic error was added in quadrature to take into account the uncertainty in the target thickness (areal weight for the ^{13}C powder, or gas pressure and cell thickness for the ^{16}O target), the beam integration and the efficiency determination. Table I quotes all the cross sections measured in this work. The proton energy was calculated from range-energy tables;[11] the uncertainties here represent the target thickness.

As an independent check of our data, a measurement of the $^{13}C(p,n)^{13}N$ thick target yield up to 13 MeV was done using a hyperpure Ge detector of which the absolute efficiency was known. The agreement between both methods was very good.

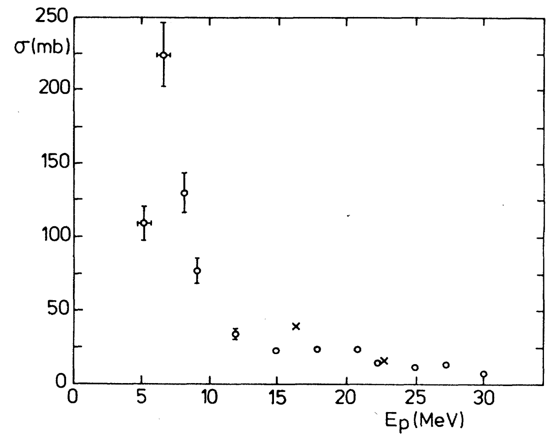

FIG. 1. Cross section for the $^{13}C(p,n)$ ^{13}N reaction measured in this work (open circles) and in Ref. 6 (crosses). Error bars are calculated as the quadratic sum of the statistical and the systematic errors (see text). Data from Ref. 6 are affected by a 20% normalization uncertainty.

III. DISCUSSION

A. The $p+{}^{13}C$ reaction

The cross section data for the ^{13}N production was presented in Fig. 1. Up to 13 MeV, taking into account the poor energy resolution of our measurement, our data

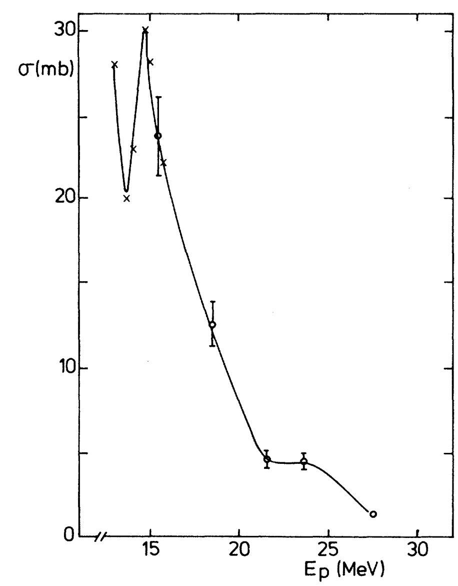

FIG. 2. Cross section for the $^{16}O(p,\alpha)$ ^{13}N reaction from this work (open circles) and from Ref. 8 (crosses). The solid line is an eye guide. Error bars on our data points have the same meaning as in Fig. 1. Data from Ref. 8 are affected by a 7% uncertainty.

TABLE I. Cross sections (in mb) measured in this work. Statistical errors are always less than 1%. An additional systematic error of 10% affects the data. The uncertainty on the proton energy reflects the target thickness and was calculated from the range-energy tables (Ref. 11).

	Cross section (mb)				
	$p+^{13}$C		$p+^{16}$O		
Ep (MeV)	^{11}C	^{13}N	^{11}C	^{13}N	^{15}O
5.2±0.5		109			
6.6±0.4		224			
8.2±0.4		130			
9.2±0.3		77			
12.1±0.3		34			
15.1±0.2		23			
15.6±0.2				24	
18.2±0.2	2.3	24			
18.6±0.2			0.76	14	11
21.2±0.2	6.9	24	0.69	4.8	19
22.7±0.2	7.1	15			
23.5±0.2			1.1	4.8	47
25.5±0.2	8.3	12			
27.8±0.1	12	14	1.2	1.6	61
30.6±0.1	10	8.2			

are in good agreement with Refs. 3 and 4. The agreement with the 22.8-MeV data point of Ref. 6 is also excellent; only the 16.3-MeV data from Ref. 6 is out of line with the trend of our measurements. A possible loss of ^{13}N activity during the powder transfer was excluded by the good agreement between our data at 11.5 and 18.5 MeV, and the rest of our measurements. Only one other radioactive element, i.e., ^{11}C, was detected, above 18-MeV proton energy.

Figure 3 shows the integrated yields of ^{13}N and ^{11}C up to 30 MeV. Below 13 MeV, the data of Ref. 3 were used to calculate the ^{13}N yield. The expected 500-μA proton intensity foreseen for the production of radioactive atoms[1] should thus provide us with an ^{13}N intensity of about 4.5×10^{12} sec^{-1}.

B. The $p+^{16}$O reaction

The cross section for ^{13}N production (Fig. 2) is quickly decreasing from 15 to 28 MeV; our data point at 15.5 MeV agrees within 5% with the Sajjad *et al.* measurement[8] which in general is also in line with the Whitehead and Forster[7] data from threshold up to 13 MeV. Thus, above 13 MeV, where the measurements of Refs. 7 and 8 differ by about 20%, our data strongly favor the results of Ref. 8.

Two contaminants, i.e., ^{15}O and ^{11}C, were measured by us and also by Sajjad *et al.*:[12] for the ^{15}O yield, though the trend of both sets of data are similar, our cross sections are about 20% lower at higher energy; however, the ^{15}O integrated yield from 25 to 26.5 MeV obtained by Beaver[13] agrees very well with the same quantity calculated from our cross section data. The cross section for the production of ^{11}C is very small (≤ 1 mb). The integrated yields for ^{13}N, ^{15}O, and ^{11}C are plotted in Fig. 4. Below 15 MeV, the yield of ^{13}N was taken from Ref. 8.

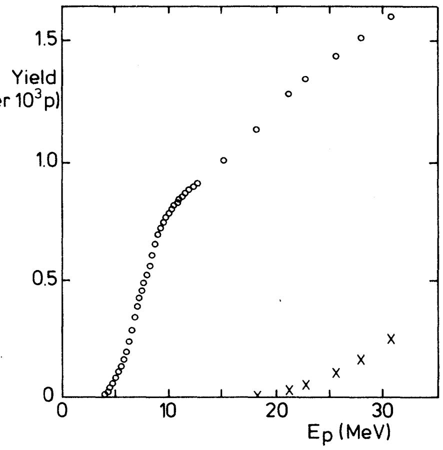

FIG. 3. Thick target yield (in outgoing nuclei per 10^3 incident protons) of ^{13}N (open circles) and of ^{11}C (crosses) produced in the $p+^{13}$C reaction, versus the proton energy. Below 13 MeV, the ^{13}N yield was calculated from the data of Ref. 3. The yield can be expressed also in mCi/μA (1 mCi/μA represents 5.9 outgoing nuclei per 10^6 incident protons); the ^{13}N thick target yield at 30 MeV corresponds roughly to 250 mCi/μA.

IV. CONCLUSION

The yield of ^{13}N from the $p+^{13}$C reaction at 30 MeV was found to be three times higher than from the $p+^{16}$O reaction, and "contaminants," i.e., other β^+ emitter isotopes, were much less abundant in the former case. Moreover, in the future setup for radioactive ions production, an oxygen gas target could hardly sustain a 500 μA proton beam, and thus solid oxides should be used,

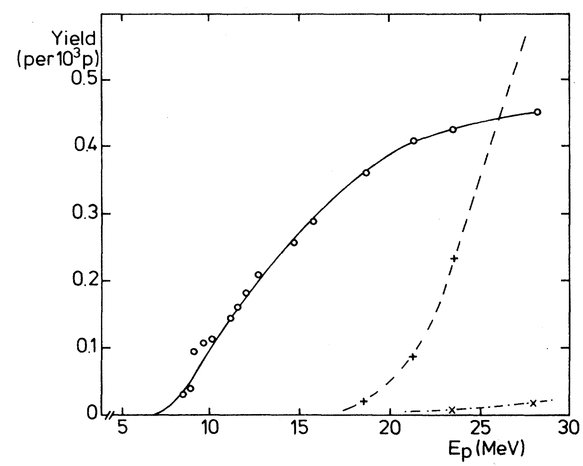

FIG. 4. Thick target yield (in outgoing nuclei per 10^3 incident protons) of ^{13}N (open circles), of ^{13}C (crosses), and of ^{15}O (plus sign) produced in the $p+^{16}$O reaction, versus the proton energy. The ^{13}N yield below 15 MeV was calculated from the data of Ref. 8. Curves (solid for ^{13}N, dashed for ^{15}O, dot-dashed for ^{13}C) were drawn to guide the eye.

again decreasing the ^{13}N production in a compound target. Two factors however are favoring the $^{16}O(p,\alpha)$ reaction: (i) the neutron production is small (the Q value for the $^{16}O(p,n)$ reaction is -16.2 MeV), and (ii) the price of the target is a few orders of magnitude smaller. Taking these factors together, it was decided to build a ^{13}C target for the ^{13}N production.

From an experimental point of view, we have developed a fast detection system which is able to sustain very high counting rates (individual counting of each detector was a few millions per sec during the irradiation time) while keeping its performance (no dead-time effects were present during the counting time, as the expected lifetimes were never distorted). This system will be used to measure the production of short-lived isotopes, down to lifetimes of a few seconds.

We are aware that uncertainties in the proton stopping powers can affect the reaction yields. Different tables can differ by at most 15% in our energy range. However, the conclusions of the present work with respect to the two ^{13}N production reactions remain valid. Moreover, in order to provide a comparison with other data, the tables that we have used[11] are the most commonly used by people measuring the yields of β^+ emitter isotopes of medical interest in the 10–20 u range.

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