Advances in the measurement of the $^{13}\text{C}(d,p)^{14}\text{C}$ cross section using AMS

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Abstract. An experimental protocol to study the total cross section for the $^{13}\text{C}(d,p)^{14}\text{C}$ nuclear reaction via AMS is being currently developed for deuteron energies between 1 and 4 MeV. We started a series of experiments in which two aluminium cathodes filled with natural-graphite (98.9% $^{12}\text{C}$, 1.1% $^{13}\text{C}$) samples were irradiated with a deuterium beam of 4 MeV in a 150° RBS configuration at Instituto Nacional de Investigaciones Nucleares 6.0 Tandem Van de Graaff Accelerator (ININ, Mexico). The relative concentrations of $^{14}\text{C}/^{12}\text{C}$ were subsequently analyzed using AMS at Laboratorio Nacional de Espectrometría de Masas con Aceleradores (LEMA, UNAM).

1. Introduction
The use of the Accelerator Mass Spectrometry (AMS) facilities to measure nuclear reaction cross sections is still pretty new, it was first implemented in 1980 by M. Paul to study $^{26}\text{Mg}(p,n)^{26}\text{Al}$ at energies between 5.2 and 6.9 MeV at ANL [1]. This was also the first work that used this technique for laboratory experiments in nuclear astrophysics.

In the present work, an experimental methodology to measure the cross section of $^{13}\text{C}(d,p)^{14}\text{C}$ by obtaining the deconvolution of the thick target reaction yield at 4 MeV lab energy using AMS is proposed. Moreover, this is one of the first works using this technique in both nuclear physics and nuclear astrophysics [2] in the only AMS facility available in Mexico, the Accelerator Mass Spectrometry National Laboratory (LEMA) [3].

The aim of this experiment is to be able to compare with existing experimental data [4] [5], establishing a methodology to perform this kind of studies. Such methodology could be applied in the future to other systems were the cross section is not well known. On the other hand, the $^{13}\text{C}(d,p)^{14}\text{C}$ reaction is of interest to study compound nucleus effects [4] and also as a new possible channel for studying the CNO synthesis in the Big Bang Nucleosynthesis (BBN) [6]. Experimental data on this reaction are scarce, so confirming previous result is also of scientific interest.
2. Experiment

Originally, we had planned to irradiate graphite samples for deuterium beam energies between 1 and 3 MeV at the 5.5 MV Van de Graaff accelerator at IFUNAM, as stated in our previous paper [7], however, this was not possible due to a malfunctioning of the accelerator’s ion source. We had to redraw the experiment for the working conditions of the ININ 6.0 MeV Tandem Van de Graaff Accelerator, where stable beam currents could only be achieved at higher energies. Thus, we irradiated two samples at $E_d = 4$ MeV for 3 hours each. One solid-state detector was placed at 150° with respect to the beam direction in order to detect the backscattered deuterons. The relative concentrations had to be sufficient to obtain minimum measurable $^{14}\text{C}/^{12}\text{C}$ suitable for the LEMA AMS-facility sensitivity-range conditions ($10^{-16} - 10^{-11}$), and consequently to produce optimum quantities of $^{14}\text{C}$. The irradiation time was previously estimated by computing the reaction rates for the relative concentrations of $^{14}\text{C}/^{12}\text{C}$ (hereinafter $N_{14}/N_{12}$) for energies between 1 and 3 MeV, where concentrations of the order of $10^{-12}$ were achieved for all these energies after 3 hours.

The targets consisted of two cylindrical aluminium cathodes, each containing 4.6 g of graphite in its natural concentration (98.9% $^{12}\text{C}$, 1.1% $^{13}\text{C}$). The surface area of the graphite was of 1.3 mm and the thickness was of 1600 μm. The geometry of the cathodes is shown in Figure 2.

The experimental setup we used at ININ bears a close resemblance to the one at IFUNAM. The difference lies mainly on the reaction chamber at ININ, which has a mechanical system to align the targets from outside the chamber in order to irradiate several samples one after another without breaking vacuum. Hence we used the sample holder shown in Figure 1 (right) made out of nylon.

After the irradiation, the samples as well as another two non-irradiated ones were analyzed by AMS at the LEMA facility.

3. Results and discussion

The number of deuterons impinging on each sample was determined from the backscattered particle spectra using SIMNRA [8] (Table 1) to simulate each spectrum using experimental data of the cross sections of the reaction channels available at IBANDL [9] at the same conditions, namely data that were available at 4 MeV at the and at 150° as well as the elemental concentration of the composition of the target, which showed that the carbon concentrations were of 31% and 32%, for samples C2 and C3 respectively. The rest corresponds to the aluminium from the cathodes, and also, oxygen and nitrogen as contaminants.

Figure 2 shows an RBS spectrum obtained and set in logarithmic scale, where the reaction and elastic channels identified in the target can be seen as well as some elastic channels from other nuclei that were also present. The reaction channels for $^{12}\text{C}+d$ and $^{13}\text{C}+d$ were clearly
located. For instance, $^{12}\text{C}(d,p)^{13}\text{C}$ proved to be an important reference since its cross section has been widely measured, hence we were able to recognize its strong contribution in our experiment thanks to the measurements made by Kashy et al. at 165° in the incident energy range of $E_d=\{802-1956\}$ keV [10], Jarjis et al. at 135° [11] within $E_d=\{2700-2960\}$ keV and at $E_d=\{3211-4333\}$ keV at 150° measured by Bonner et al. [12]. Regarding the exit channels for $^{13}\text{C}+d$, only $^{13}\text{C}(d,\alpha)^{11}\text{B}$ [13] and $^{13}\text{C}(d,p)^{14}\text{C}$ measured by Marion and Weber were available and thereby located, however, the cross section of the latter was measured at 135° [4] and therefore it was not possible to use it to fit our data. It is worth mentioning that the sum of the individual contributions should reproduce our experimental data, however, since some of the data available for the simulation are from experiments at angles different from 150° (this experiment), the strength is not reproduced. Therefore, those reactions that were present at backscattered angles different from 150° were used for identification only.

Table 1. Number of particles by unit solid angle (particles*sr) obtained by SIMNRA and the total number of particles calculated by the solid angle

| Sample | Particles*sr | No. particles |
|--------|--------------|---------------|
| C2     | 9.00E+13     | 1.70E+17      |
| C3     | 7.23E+13     | 1.27E+17      |

Figure 2. Backscattering protons spectrum obtained (shown is dark red points connected by a red line to guide the eye) set in logarithmic scale, compared with the simulated contributions from individual reactions performed with SIMNRA (shown in different colors).

The relative concentrations of $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ measured in the AMS system at the LEMA facilities are summarized in Table 2, where a clear difference of one order of magnitude can be seen between the irradiated samples and the non-irradiated ones, which demonstrates the production of $^{14}\text{C}$. Also there is an important difference in the relative concentrations for C2 and C3. This can be attributed in part to the different incident deuteron flux received by the samples (Table 1). Other effects like the ratio of the irradiated area in the graphite surface ($1.33 \text{ mm}^2$) and the area of the deuteron beam ($\sim 2.69 \text{ mm}^2$) might also influence the measured concentration and further studies are required for this.
the following equation:

\[ \sigma \] produced nuclei in the number, \( f \) the isotopic fraction of the target nuclei of interest, \( M_B \) the atomic weight of the target, \( dE/d(\rho x) \) the stopping power of beam in the target per unit of mass thickness and \( \sigma_i \) the cross section for the production for \( i \). 

For the nuclear reactions of interest, namely \( ^{12}\text{C}(d,p)^{13}\text{C} \) and \( ^{13}\text{C}(d,p)^{14}\text{C} \) we have correspondingly:

\[ y_{13}(E_0) = \frac{N_A f_{13}}{M_B} \int_{E_0}^{0} \sigma_{13}(E) \left[ \frac{dE}{d(\rho x)} \right]^{-1} dE \]  

and

\[ y_{14}(E_0) = \frac{N_A f_{14}}{M_B} \int_{E_0}^{0} \sigma_{14}(E) \left[ \frac{dE}{d(\rho x)} \right]^{-1} dE \]  

where \( f_{12}=0.989 \) and \( f_{13}=0.01 \) are the isotopic fractions for \( ^{12}\text{C} \) and \( ^{13}\text{C} \) respectively, \( \sigma_{13} \) and \( \sigma_{14} \) the cross sections for the production of \( ^{13}\text{C} \) and \( ^{14}\text{C} \).

Similarly, considering the reaction channels available at \( E \) for \( ^{12}\text{C}+d \) and \( ^{13}\text{C}+d \) we will have the following equation:

\[ Y_{12,13}(E_0) = \frac{N_A f_{12,13}}{M_B} \int_{E_0}^{0} \Sigma_{12,13}(E) \left[ \frac{dE}{d(\rho x)} \right]^{-1} dE \]  

where \( \Sigma_{12} \) and \( \Sigma_{13} \) are the sums of the reaction-channel cross sections of \( ^{12}\text{C}+d \) and \( ^{13}\text{C}+d \) respectively, except for \((d,p)\) at \( E \).

Likewise, we consider the yield for \( ^{13}\text{C}(d,p)^{14}\text{C} \) as \( Y_{14} = N_{\text{net}14}/N_d \), where \( N_{\text{net}14} \) is the difference between the \( ^{14}\text{C} \) present nuclei in the irradiated and non-irradiated samples; \( N_d \), the number of incident deuterons. Thus, if there are \( N_d \) incident projectiles, the total number of produced nuclei in the \( i-th \) process will be \( N_i = N_d Y_i \).

On the other hand, since several reactions take place simultaneously, the total number of particles of \( ^{12}\text{C}, \ ^{13}\text{C} \) and \( ^{14}\text{C} \) will be correspondingly:

\[ N_{12} = N_{12}^0 - N_d y_{13} - N_d \frac{N_A f_{12}}{M_B} \int_{E_0}^{0} \Sigma_{12}'(E) \left[ \frac{dE}{d(\rho x)} \right]^{-1} dE \]

Table 2. Relative concentrations of \( ^{14}\text{C}/^{12}\text{C} \) and \( ^{13}\text{C}/^{12}\text{C} \)

| Sample   | \( ^{14}\text{C}/^{12}\text{C} \)     | \( ^{13}\text{C}/^{12}\text{C} \)     |
|----------|--------------------------------------|--------------------------------------|
| C2       | 7.76E-12 ± 9.27E-19                  | 1.43E-02 ± 6.59E-25                  |
| C3       | 1.53E-12 ± 3.10E-19                  | 1.34E-02 ± 2.03E-25                  |
| C4       | 1.46E-13 ± 3.35E-19                  | 1.31E-02 ± 7.31E-26                  |
| C6       | 1.72E-13 ± 3.73E-19                  | 1.35E-02 ± 9.27E-26                  |

Since the thickness of the targets were of 1.6 mm, an order of magnitude larger than the projected range of the deuterons going through graphite (165 \( \mu \text{m} \) at \( E_{lab}=4 \text{ MeV} \)), the targets were treated as thick and therefore it was possible to compute the yields as follows:

\[ y_i(E_0) = \frac{N_A f_i}{M_B} \int_{E_0}^{0} \sigma_i(E) \left[ \frac{dE}{d(\rho x)} \right]^{-1} dE \]  

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where \( y_i \) corresponds to the yield of the reaction channel of interest, \( N_A \) is the Avogadro’s number, \( f_i \) the isotopic fraction of the target nuclei of interest, \( M_B \) the atomic weight of the target, \( dE/d(\rho x) \) the stopping power of beam in the target per unit of mass thickness and \( \sigma_i \) the cross section for the production for \( i \).

For the nuclear reactions of interest, namely \( ^{12}\text{C}(d,p)^{13}\text{C} \) and \( ^{13}\text{C}(d,p)^{14}\text{C} \) we have correspondingly:

\[ y_{13}(E_0) = \frac{N_A f_{13}}{M_B} \int_{E_0}^{0} \sigma_{13}(E) \left[ \frac{dE}{d(\rho x)} \right]^{-1} dE \]  

and

\[ y_{14}(E_0) = \frac{N_A f_{14}}{M_B} \int_{E_0}^{0} \sigma_{14}(E) \left[ \frac{dE}{d(\rho x)} \right]^{-1} dE \]  

where \( f_{12}=0.989 \) and \( f_{13}=0.01 \) are the isotopic fractions for \( ^{12}\text{C} \) and \( ^{13}\text{C} \) respectively, \( \sigma_{13} \) and \( \sigma_{14} \) the cross sections for the production of \( ^{13}\text{C} \) and \( ^{14}\text{C} \).

Similarly, considering the reaction channels available at \( E \) for \( ^{12}\text{C}+d \) and \( ^{13}\text{C}+d \) we will have the following equation:

\[ Y_{12,13}(E_0) = \frac{N_A f_{12,13}}{M_B} \int_{E_0}^{0} \Sigma_{12,13}(E) \left[ \frac{dE}{d(\rho x)} \right]^{-1} dE \]  

where \( \Sigma_{12} \) and \( \Sigma_{13} \) are the sums of the reaction-channel cross sections of \( ^{12}\text{C}+d \) and \( ^{13}\text{C}+d \) respectively, except for \((d,p)\) at \( E \).

Likewise, we consider the yield for \( ^{13}\text{C}(d,p)^{14}\text{C} \) as \( Y_{14} = N_{\text{net}14}/N_d \), where \( N_{\text{net}14} \) is the difference between the \( ^{14}\text{C} \) present nuclei in the irradiated and non-irradiated samples; \( N_d \), the number of incident deuterons. Thus, if there are \( N_d \) incident projectiles, the total number of produced nuclei in the \( i-th \) process will be \( N_i = N_d Y_i \).

On the other hand, since several reactions take place simultaneously, the total number of particles of \( ^{12}\text{C}, ^{13}\text{C} \) and \( ^{14}\text{C} \) will be correspondingly:

\[ N_{12} = N_{12}^0 - N_d y_{13} - N_d \frac{N_A f_{12}}{M_B} \int_{E_0}^{0} \Sigma_{12}'(E) \left[ \frac{dE}{d(\rho x)} \right]^{-1} dE \]
From equations 5, 6 and 7 it is possible to obtain the expressions for \( y_{13} \) and \( y_{14} \) in terms of irradiated and non-irradiated relative concentrations \( (N_{14}/N_{12}, N_{13}/N_{12} \) for the former, and \( N_{14}^0/N_{12}^0 \) and \( N_{13}^0/N_{12}^0 \) for the latter), the number of incident deuterons \( (N_d) \) and the yields for reaction channels other than \( (d,p) \). These expressions provide a way to extract \( y_{13} \) and \( y_{14} \) from the experiment, with the exception of \( Y_{12} \) and \( Y_{13} \), which can be calculated from equation 4 using experimental cross sections reported in the literature [15]. Moreover the experimental results for \( y_{13} \) and \( y_{14} \) can be compared to those calculated using equations 2 and 3 and cross sections from the literature. Table 4 shows the experimental and calculated values of the yields.

\[
y_{13} = \left[ 1 + \frac{N_{13}^0}{N_{12}^0} \right]^{-1} \cdot \left[ \frac{N_{12}^0}{N_d} \left( \frac{N_{13}}{N_{12}} - \frac{N_{13}^0}{N_{12}^0} + \frac{N_{14}}{N_{12}} - \frac{N_{14}^0}{N_{12}^0} \right) + Y_{13} - \frac{N_{13}^0}{N_{12}^0} Y_{12} - \frac{N_{14}^0}{N_{12}^0} Y_{12} \right] \\
y_{14} = \frac{N_{12}^0}{N_d} \left( \frac{N_{14}}{N_{12}} - \frac{N_{14}^0}{N_{12}^0} \right) - \frac{N_{14}^0}{N_{12}^0} \left[ \left[ 1 + \frac{N_{13}}{N_{12}} + \frac{N_{14}}{N_{12}} \right]^{-1} \cdot \left[ \frac{N_{12}}{N_d} \left( \frac{N_{13}}{N_{12}} + \frac{N_{14}}{N_{12}} \right) + Y_{13} - \frac{N_{13}^0}{N_{12}^0} Y_{12} - \frac{N_{14}^0}{N_{12}^0} Y_{12} \right] + Y_{12} \right]
\]

Table 3. Yield calculation by two different ways. In the first column shows the yields derived using Tendl-2014 library data are enlisted. In the last column the values of the yields obtained using experimental data obtained in the present work

|        | Yields using library data | Yields experimental data (this work) |
|--------|--------------------------|-------------------------------------|
|        |                          | C2                          | C3                          |
| \( Y_{12} \) | 2.05E-04                | -                           | -                           |
| \( Y_{13} \) | 3.64E-05                | -                           | -                           |
| \( y_{13} \) | 4.61E-04                | 4.67E-03                    | 4.23E-03                    |
| \( y_{14} \) | 1.26E-02                | 3.57E-11                    | 8.33E-12                    |

Table 3 describes the yield values calculated using theoretical data (TENDL 2014) [14] retrieved from ENDF data base [15] and the values of the yields obtained using experimental data obtained in the present work, such as those enlisted in Table 2 and the number of incident deuterons in carbon, \( N_d \). The significant disagreement between these values can be be better explained observing Figure 3, which shows the comparison of \( \Delta E \cdot E \) energy spectra for the concentration of \( ^{14} \text{C} \) between a non-irradiated sample (left) and an irradiated one (right). The dark-shaded area shown in both spectra is the region of interest (ROI), where the \( ^{14} \text{C} \) produced was expected to appear. However, as seen in the spectrum for the irradiated sample, a considerable amount of contamination can be clearly observed; on one hand, a significant
Figure 3. (left) $\Delta E \cdot E$ energy spectrum for the concentration of $^{14}$C of a non-irradiated sample (C4); (right) $\Delta E \cdot E$ energy spectrum for the concentration of $^{14}$C of an irradiated sample (C2).

disagreement between the TENDL data and the experimental data [4] [5] is consistent with this result (see Figure 4); on the other hand, a possible contamination with the ($^{12}$C + $^2$H)$^{++}$ molecular state (and maybe other contaminants) in our AMS measurements cannot be ruled out.

Figure 4. The red line shows the experimental data for the cross section of $^{13}$C($d$, $p$)$^{14}$C measured by Marion and Weber [4]. The blue line shows the cross section data of $^{13}$C($d$, $p$)$^{14}$C retrieved from TENDL 2014 library [14].

4. Conclusions
The production of $^{14}$C was successfully achieved by performing the $^{13}$C($d$, $p$)$^{14}$C nuclear reaction using natural graphite as a target and 4 Mev deuterium beam. The AMS analysis performed at LEMA of the relative concentration of $^{14}$C/$^{12}$C shows a significant amount of $^{14}$C in the irradiated samples compared to those that did not undergo irradiation. The sample geometry used for the deuterium irradiation was that required at LEMA to enable direct AMS analysis after irradiation, however, this complicated the process because the graphite diameter was only 1.3 mm. On the other hand, the sample preparation implies working with thick targets, which also imposes difficulties in the extraction of cross sections of interest. A deconvolution of the yield is possible in order to extract the cross section as a function of energy. However, a series of measurements of the yield at different energies is needed. This work has helped to set the basis for future experiments using the AMS technique in the study of cross sections induced by
charged particles. Additional experiments and improvements, mainly in target preparation and beam focusing, are in order to complete a protocol to perform this type of measurements.

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