Investigating $^{13}C+^{12}C$ Reaction by the Activation Method. Sensitivity Tests

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Abstract. We have performed experiments to check the limits of sensitivity of the activation method using the new 3 MV Tandetron accelerator and the low and ultra-low background laboratories of the "Horia Hulubei" National Institute of Physics and Nuclear Engineering (IFIN-HH). We have used the $^{12}C+^{13}C$ reaction at beam energies E_{lab}= 6, 7 and 8 MeV. The knowledge of this fusion cross section at deep sub-barrier energies is of interest for astrophysical applications, as it provides an upper limit for the fusion cross section of $^{12}C+^{12}C$ over a wide energy range. A ^{13}C beam with intensities 0.5-2 particleμA was provided by the accelerator and used to bombard graphite targets, resulting in activation with ^{24}Na from the $^{12}C(^{13}C,p)$ reaction. The 1369 and 2754 keV gamma-rays from ^{24}Na de-activation were clearly observed in the spectra obtained in two different laboratories used for measurements at low and ultralow background: one at the surface and one located underground in the Unirea salt mine from Slanic Prahova, Romania. In the underground laboratory, for E_{lab} = 6 MeV we have measured an activity of 0.085 ± 0.011 Bq, corresponding to cross sections of 1-3 nb. This demonstrates that it is possible to measure ^{12}C targets irradiated at lower energies for at least 10 times lower cross sections than before. $\beta-\gamma$ coincidences will lead us another factor of 10 lower, proving that this installations can be successfully used for nuclear astrophysics measurements.

INTRODUCTION

With the final goal of establishing a solid line of research in nuclear astrophysics (NA) at the Bucharest accelerators and laboratories of IFIN-HH, we have performed experiments to check the limits of one method that seems appropriate and for which the institute has or could acquire installations: the activation method. We used for irradiation one of the new tandem accelerators which can provide good intensities for light ions and the low and ultralow background laboratories, situated above ground and underground, respectively, for activation measurements. We have chosen the $^{13}C+^{12}C$ reaction, which leads to an activation appropriate for our tests: ^{24}Na, with a half-life of 15.0 hours, formed by one proton evaporation.

Nuclear astrophysics, or more precisely nuclear physics for astrophysics, is becoming more and more an explicit motivation for nuclear physics research, for European laboratories programs, in the USA, Japan and China, but also for the ones from Romania: through direct measurements (at low energies as in stars) or indirect methods (at the most common energies in nuclear physics laboratories). Direct measurements are very difficult because of the low cross sections involved and require dedicated facilities: proton or alpha particle accelerators of very high intensities at low energies and, if possible, low background and special detection systems. Such a facility did not exist in Romania and therefore, direct measurements were not made in Romania. The use of indirect methods involve typically radioactive beams, which were also not available locally. We wanted to prove that we can do direct measurements now, using newly available installations [1,2].

The reaction $^{12}C + ^{12}C$ in the low energy region is of great interest in astrophysics (see eg [3].) because of its essential role in studying a wide range of burning scenarios in carbon-rich stellar environments. It is important for understanding carbon burning nucleosynthesis that occurs in stars with more than 10 solar masses during late evolutionary periods [4], in intermediate mass stars (8-10 solar masses), which can lead a detonation wave and a supernova explosion [5], in binary systems, where a massive carbon-oxygen white dwarf exceeds the Chandrasekhar mass limit accumulating material from its partner star. The temperatures at which the carbon burnout occurs are found in the range of 0.5-1.2 GK corresponding to the center-of-mass energy range of 1 to 3 MeV. To verify all these scenarios and put constraints on models requires a detailed knowledge of the carbon fusion processes at these energies. Considerable efforts have been made to measure the cross section of $^{12}C + ^{12}C$ reaction at astrophysical energies, involving both the detection of charged particles and gamma-ray spectroscopy. However, previous measurements were made for $E_{c.m} \geq 2.1$ MeV, the upper region of astrophysical interest. Also, as $E_{c.m} = 3.0$ MeV cross sections reported are not consistent and are quite uncertain [6-8]. Moreover, the extrapolation procedure in the case of $^{12}C + ^{12}C$ from current experimental data at ultra-low energies is complicated by the presence of possible resonant structures even in the low energy excitation function. Measurements that could extend to below $E_{c.m} = 2.1$ MeV would be extremely important. It was found, however, that the $^{13}C + ^{12}C$ and $^{13}C + ^{13}C$ reactions do not have such resonances and provide material for understanding fusion at low energies, and ways to determine the maximum cross section for the reaction $^{12}C + ^{12}C$.

A University of Notre Dame group [9] has proposed a $^{13}C + ^{12}C$ experiment in collaboration with us and a group of Lanzhou, China at 3 MV Tandem from IFIN-HH. It is the motivation for our choice of measurements here: irradiations with a ^{13}C beam followed by measurement of activities at both surface and underground laboratory characterized by an ultra-low background radiation.

EXPERIMENTAL METHODS FOR INVESTIGATION OF THE $^{12}C + ^{13}C$ REACTION BY THE ACTIVATION METHOD

The *HVEE Tandetron 3 MV electrostatic accelerator* - recently installed at IFIN-HH is dedicated to:

1) Ion Beam Analysis (IBA) - analytical techniques that use accelerated ion beams: Rutherford backscattering spectrometry (RBS), X-ray emission induced by charged particles (PIXE), nuclear reaction analysis (NRA), etc.

2) Testing the radiation resistance of the materials or implants.

3) Nuclear astrophysics.

For nuclear astrophysics we assess that this facility is suitable for direct measurements of cross sections induced by α particles (He-burning) and light ions (^{6}Li, ^{12}C, ^{13}C, ^{16}O ...), due to relatively low energies and high intensities and its stable functioning, as tested by us last year.

The *GammaSpec laboratory* is an above ground installation in IFIN-HH main campus, in the same location as the tandem accelerators, consisting of a HpGe detector very well shielded, and carefully calibrated with sources and international inter-laboratory comparisons [10, 11].

The *Underground Laboratory in the Unirea salt mine, Slanic Prahova (MicroBequerel or "μBq")*, is located in a salt mine, about 2 hours drive North of Bucharest. Environmental conditions in the salt mine are very stable year round: temperature between 12 and 13^{o} C, humidity 67-70% approximately, area of ~70,000 m^2, height between 52 and 57 m, depth is 208 m below ground (approximately 600 m.w.e), the distance between the walls is between 32 and 36 m, volume is 2.9×10^6 m^3 [12]. In this mine a laboratory was built to perform measurements using gamma-ray spectrometry in ultralow radiation background. The average dose underground was found 1.29 ± 0.30 nSv/h, approximately 70-80 times lower than the dose at the surface. As ambient background radiation comes from: i) natural radioactivity (especially from the decay of ^{238}U, ^{232}Th and ^{222}Rn present in the atmosphere and ^{40}K); ii) cosmic rays (μ, ^{1}H, ^{3}H; ^{7}Be, ^{14}C ...); and iii) neutrons from (α, n) reactions and fission, the i) and iii) sources are particularly low in this mine due to its thick and compact salt walls. Figure 1 compares γ-ray spectra measured above ground and underground. The top spectrum shows that the strongest component of the γ rays spectrum at $E\gamma$<3MeV is associated with the natural environment radioactivity and exhibits intense characteristic lines. At higher energies, the background radiation originates mostly from cosmic rays. The natural radioactivity is significantly reduced for measurements in the underground laboratory (bottom spectrum). From Fig. 1 it can be seen that the measured background radiation (using a protection shield consisting of 15 cm Pb and 5 cm Cu produced by Canberra Ind.) is about 4000 times smaller compared to the background spectrum measured at the surface. This is the major advantage we want to test and use in the current measurements [13, 14].

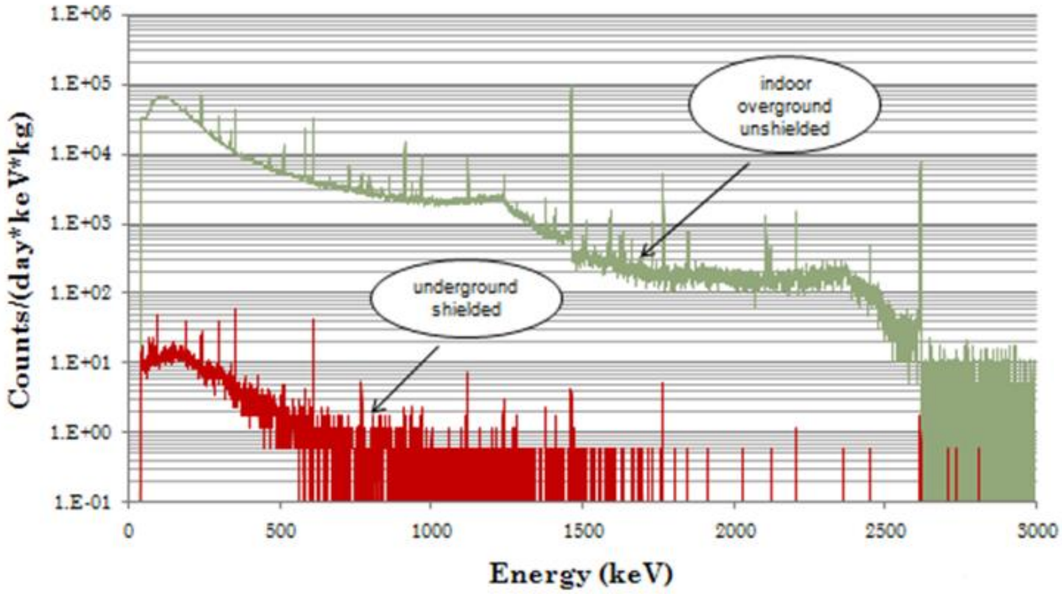

FIGURE 1. Typical spectrum of γ rays measured at the Earth's surface and underground

EXPERIMENTAL RESULTS

In this experimental phase we studied ^{12}C + ^{13}C fusion reaction in the laboratory energy range of 6 to 8 MeV. A $^{13}C^{+3}$ beam with intensity 0.5 μA, at the first irradiation (E_{lab}=8 MeV), and 1.9 μA, for the irradiations at energies E_{lab}= 6 and 7 MeV, provided by the 3 MV Tandetron accelerator, impinged on a 1 mm thick natural carbon (graphite) target. A gas stripper system was used to increase the intensity of the $^{13}C^{+3}$ charge state.

Cross section of the $^{12}C(^{13}C, p)\ ^{24}Na$ reaction can be determined by measuring the γ radiation corresponding to nucleus ^{24}Na ($T_{1/2}$ = 15.00 h), using the activation method. The irradiated carbon targets were measured in the GammaSpec laboratory and in the underground laboratory. The cascading γ rays (1369 and 2754 keV) were detected with germanium detectors. The detection systems have been protected with lead castles to reduce ambient background radiation. The first case studied was a C target irradiated for 15 hours with an 8 MeV beam. γ rays were measured in the underground laboratory 4 times successively, 82.000 s each measurement (comparable to $T_{1/2}$ of ^{24}Na) using a germanium detector with 120% relative efficiency, in a protective castle as described before. We found an activity of 4.44 ± 0.19 Bq and evaluated the minimal detectable activity at 0.048 Bq. In the four the γ-ray spectra we could observe the decreasing activity of the irradiated target and the gradual relative increase of the background radiation.

The following two steps consisted of the activation of C targets at two different beam energies, 6 and 7 MeV, and from measuring them both in the underground laboratory and in the GammaSpec laboratory located at the surface. In this latter laboratory, the spectrometric system is based on an Ortec HPGe detector 30185 GEM, resolution 2.1 keV at 1332 keV of ^{60}Co, and relative efficiency 30% (compared to 3 "x 3" NaI (Tl) standard). This spectrometric system is protected by a lead cylindrical shield (10 cm thick), covered on the inside with tin (1 mm thick) and copper (1.5 mm thick) foils. Thus for γ rays of energies between 20 and 2750 keV in a 24 hours measurement one obtains a count rate of 1.2-1.8 events/sec (depends mainly on the concentration of ^{222}Rn in natural background).

For the target irradiated (23 hours) at E_{lab} = 7 MeV, and measured in the GammaSpec laboratory, the beam intensity was 1.87 μA, yielding an activity at the end of irradiation equal with 5.20 ± 0.40 Bq. This activity was calculated after corrections were made for the efficiency and the time needed to transport the target from the reaction chamber to the GammaSpec laboratory. For measurements made in the underground laboratory another C target was irradiated using the same parameters, but for a longer irradiation time of about 25 hours.

Activity values measured in the two laboratories are shown in Tables 1 and 2; the two sets of measurements gave comparable results, within the evaluated uncertainties. The incident ^{13}C beam energy (E_{lab}) in MeV, beam current (I) in μA, and counting time of the irradiated targets (t_c) in seconds are also given in these tables. Knowing the activated targets activity at the measurement moment and the background rate of accumulation we determined the limit of detection for the evaluation of the ^{12}C + ^{13}C fusion reaction cross sections. The minimum measurable cross section results to be about 3 nb using beam intensity around 0.6 pμA (particleμA, $^{13}C^{+3}$ charge state) , as in these cases. That is an order of magnitude below the lowest value measured until now in other laboratories. Increasing the beam intensity to approximately 6-10 pμA, it is possible to decrease the limit of detection of 10 more times, so we can measure at the energies lower than those now existing in the literature.

Tests conducted at the lowest $E_{lab}(^{13}C)$ = 6 MeV have revealed low activities of the activated targets, but to which the experimental setups are still sensitive. Barely in the surface lab, but clearly in the underground one (see Fig. 2) [15]. Reducing the limit by an order of magnitude is still possible by increasing the beam intensity. There will be, however, limitations on the extent to which the current intensity can be increased without damaging the targets. A high current beam raises problems with sputtering effect (some produced ^{24}Na's are sputtered away from the target surface during irradiation) and with heating effects. In a test at 10 μA we had visible signs of carbon sputtering from the target. For future measurements it will be necessary to construct a target cooling system. But again there is a limitation on how heat can be dissipated in the target.

Another way to improve the signal-to-noise ratio in de-activation measurements is using the β−γ coincidence method. This method allows to suppress the ambient background γ rays from natural radioactive isotopes such as ^{40}K and ^{208}Tl. In the Notre Dame experiment the peaks at 1369 keV and 2754 keV of ^{24}Na could be observed only in the β gated γ-ray spectra. It is obvious that this experimental setup made now at IFIN-HH, will allow decreasing the total fusion cross section from this measurement with another order of magnitude.

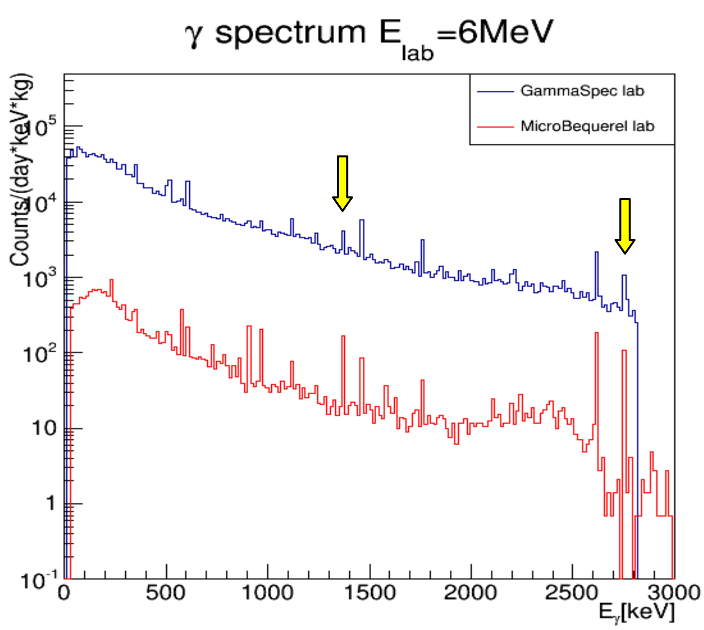

FIGURE 2. Comparison between γ spectra (E_{lab} = 6 MeV) measured in underground laboratory and GammaSpec laboratory (arrows-the cascading γ rays 1369 and 2754 keV)

TABLE 1. Experimental results obtained in GammaSpec laboratory

$E_{lab}(^{13}C)$ (MeV)	I(μA)	t_c(s)	^{24}Na (Bq)
7.0	**1.87**	**81000**	**5.20 ± 0.40**
6.0	**1.90**	**86400**	**0.115 ± 0.018**

TABLE 2. Experimental results obtained in the underground laboratory

$E_{lab}(^{13}C)$ (MeV)	I(μA)	t_c(s)	^{24}Na (Bq)
7.0	**1.87**	**86400**	**5.23 ± 0.043**
6.0	**1.90**	**84480**	**0.085 ± 0.011**

CONCLUSIONS

Study of carbon burning is an open question in nuclear astrophysics. This process represents the third stage of stellar evolution of massive stars with mass greater than 8 stellar masses that continue mainly through ^{12}C + ^{12}C fusion processes and to a lesser extent by ^{12}C + ^{16}O. Direct measurement at the Gamow window energies are therefore essential, but are difficult to carry due to the background from the cosmic rays, terrestrial environment and/or accelerator beams. Major improvements can be achieved by using high intensity accelerators, advanced detection techniques and/or underground measurements. ^{12}C + ^{13}C fusion process gives information about the fusion mechanism at low energies and can be studied both in-beam γ spectroscopy and activation method using experimental setups that consists of an accelerator and detectors for γ spectroscopy.

To determine the optimum parameters of this experiment, stability and resolution tests of ^{12}C beam obtained at the 3 MV accelerator of IFIN-HH were conducted last year. Following these tests, it turns out that the accelerator has the characteristics required for nuclear astrophysics measurements, namely: allow the terminal voltage between 0.1-3.2 MV, stable while providing stability of incident beam energy used, the currents are stable over time, allowing precise measurements. In particular, the intensities of the order of 10 pμA obtained for ^{12}C, an order of magnitude higher than those obtained from the University of Notre Dame FN tandem, make possible to carry the proposed experiments in collaboration with the group from there.

We studied the ^{12}C+^{13}C fusion reaction in the energy range $E_{c.m}$ = 2.9 - 3.8 MeV using the activation method and gamma-ray spectroscopy. Activities of irradiated targets measured both in the underground and surface laboratories allowed to determine the limit of detection of cross sections of the order of 1-3 nb. By increasing the intensity it is possible to gain a factor of 10 in sensitivity and by using β–γ coincidences, another factor of 10. However, this will imply a good cooling of the graphite targets. We emphasize that the minimum value of the measurable cross sections in general, is dependent on the specific characteristics of the produced isotope and of the γ transition(s) used, but the order of magnitude set here (nanobarns) remains valid, as remains the possibility to reduce it by increasing the intensity and using β–γ coincidences. Calibrations and measurements performed in identical or similar conditions will also allow us to reduce the uncertainties associated with the experimental data corresponding with range $E_{c.m}$ =

2.6-5.0 MeV below 20%, and to determine the cross section for the ^{12}C + ^{13}C process at an energy lower than $E_{c.m}$ = 2.6 MeV.

In conclusion, the 3 MV accelerator is suitable for nuclear astrophysics measurements due to energies and intensities provided and stability in operation. Low (DFN) and ultralow ("μBq" Slanic) background laboratories of the institute can be successfully used for measurements by activation with lifetime greater than ten minutes and several hours, respectively, necessary to transport the probes. These facilities have been included recently in a European project proposal Horizon 2020 program, called the European Laboratory Astrophysics Network (ELAN) as TA (Transnational Access facility), in a select group of seven multi-disciplinary laboratories of atomic and molecular spectroscopy or radiation installations and of only two other nuclear astrophysics labs.

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