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A study of the tandem-terminal-stripper reaction $^{1}H(^{12}C,\gamma)^{13}N$ with accelerator mass spectrometry

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Abstract

A novel method to determine small cross-sections of nuclear reactions of astrophysical interest was implemented at the Vienna Environmental Research Accelerator laboratory (VERA). This method combines inverse-kinematics reactions taking place inside the stripper of a tandem accelerator with the identification of the recoil products using the accelerator mass spectrometry (AMS) technique. As a first test we studied the $^{1}H(^{12}C,\gamma)^{13}N$ reaction, which is the starting reaction of the stellar CNO cycle of hydrogen burning. The ^{13}N recoil products as well as the interfering isobar ^{13}C were separated from the main ^{12}C beam by common AMS techniques. Measurements with ^{13}N recoil ions of different charge states were performed at center-of-mass energies of 229 and 206 keV, respectively. Notwithstanding the restrictions of VERA, the principle feasibility of the method could be demonstrated.

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1. Introduction

In this work we present a novel method to measure small cross-sections of nuclear reactions at very low energies. In particular we want to investigate the $^{1}H(^{12}C,\gamma)^{13}N$ radiative capture reaction, which is of astrophysical interest since it takes part in the stellar CNO cycle of hydrogen burning [1].

Other groups have already performed measurements of this cross-section from thick-target yields reaching center-of-mass energies as low as about

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70 keV [2–4]. They studied the $^{12}C(p,\gamma)^{13}N$ reaction by either detecting the associated prompt gamma rays or by measuring the positron activity of the product nucleus ^{13}N, respectively. Since at that low energies the capture cross-sections for $^{12}C(p,\gamma)^{13}N$ become very small (about 10^{-10} barn at 100 keV), the background produced by competing reactions poses a severe hindrance for the gamma-ray detection method.

In a pioneering effort, Paul and coworkers proposed that one could use the carbon foil stripper inside the terminal of a tandem accelerator as reaction targets [5]. With the extremely high sensitivity of AMS the separation from the projectiles and background as well as the detection of the reaction product should be possible. In this way, they attempted the measurement of the fusion reaction $^{12}C(^{12}C,\gamma)^{24}Mg$ at a center-of-mass energy of 6.13 MeV. Although they clearly detected ^{24}Mg ions in their final detection system, one could not unambiguously identify them as fusion products from the stripper reaction [5].

The aim of our efforts was to demonstrate that one can use an AMS facility for measuring the $^{1}H(^{12}C,\gamma)^{13}N$ reaction cross-section. At the Vienna Environmental Research Accelerator (VERA) [6] a $^{12}C^{-}$ beam was injected into the tandem accelerator. The stripper canal inside the terminal of the accelerator was filled with hydrogen gas, the produced ^{13}N recoils were further accelerated and then separated from the ^{12}C projectiles and background particles, like ^{13}C, using an analyzing beamline, essentially consisting of a 90° double-focusing magnet (bending radius: 127 cm), a 90° spherical electrostatic separator (bending radius: 200 cm) and an ionization chamber (entrance window: 100 nm thick silicon nitride; gas pressure: 25 mbar isobutane) in combination with a surface barrier detector for ΔE and E_{res} signals, respectively.

2. Experimental considerations

The pelletron accelerator of VERA operates at terminal voltages ranging from 1.0 to 3.5 MV. Therefore, the $^{1}H(^{12}C,\gamma)^{13}N$ reaction can be investigated for center-of-mass-energies ranging from 80 to 270 keV. To achieve even lower energies at the stripper ^{12}C ions can be injected as molecules like $^{12}C_2^{-}$. In any case, the recoil ions have final energies between about 3 and 12 MeV, allowing good particle discrimination.

The cross-section for a reaction can be expressed as $\sigma = N_R/(N_P\delta_{target})$, N_R being the number of the nuclei produced in the reaction, N_P the number of projectiles and δ_{target} the overall thickness of the target. The number of the ^{13}N reaction products, N_R, can be expressed as the ratio between the number of detected ^{13}N particles, N_D and the transmission T from the reaction target to the rare-isotope detector

$$N_R = N_D/T. \tag{1}$$

A calibration of our method implies the measurement of a factor F determined from a known reference cross-section σ'

$$F = T\delta_{target} = N_D/(\sigma' N_P). \tag{2}$$

Measuring F for different energies should give all the information needed to extrapolate to other energies.

The transmission from the gas stripper to the detector depends not only on the charge state distribution of the produced ^{13}N nuclei but also on the emittance properties of the different ionic states and on the energy of the ions.

In this particular case, $^{12}C^{-}$ particles, having after the preacceleration section an energy E_0, are injected into the accelerator, which is operated at terminal voltage $V_{terminal}$. Inside the terminal stripper they can react with ^{1}H at a center-of-mass energy

$$E^{CM} = \frac{M_{^1H}}{M_{^1H} + M_{^{12}C}}(eV_{terminal} + E_0). \tag{3}$$

With deexitation of the compound nucleus by photon emission and neglecting the very small amount of energy transferred to the massive particle, the center-of-mass velocity of the ^{13}N nucleus is calculated to be

$$v^{CM}_{^{13}N} \cong \frac{(E^{CM} + Q)}{M_{^{13}N}c}. \tag{4}$$

Leaving the accelerator, the laboratory energy of the product nucleus is given by

$$E^{\mathrm{lab}}_{^{13}\mathrm{N}} = \frac{M_{^{13}\mathrm{N}}}{2}\left(\left(v^{\mathrm{CM}} + v^{\mathrm{CM}}_{^{13}\mathrm{N}}\cos(\alpha^{\mathrm{CM}})\right)^2 + \left(v^{\mathrm{CM}}_{^{13}\mathrm{N}}\sin(\alpha^{\mathrm{CM}})\right)^2\right) + qeV_{\mathrm{terminal}}. \quad (5)$$

α^{CM} denotes the scattering angle in the center-of-mass system with respect to the incident particle, q the charge state of the reaction product after the stripping process, Q the Q-value of the reaction, equal to $(M_{^{12}\mathrm{C}} + M_{^{1}\mathrm{H}} - M_{^{13}\mathrm{N}})c^2$, e the electron charge, and v^{CM} the velocity of the center-of-mass system.

During the measurement, background signals may arise from ^{13}C ions with the same energy and charge state as the ^{13}N ions. About one percent of $^{13}C^-$ is extracted from the source as $^{13}CH^-$ molecule and pass the electrostatic separator of the injector line. A small amount of these molecules dissociates into $^{1}H^0 + {}^{13}C^-$ before reaching the injection magnet, giving $^{13}C^-$ an energy of $E_0 M_{^{13}\mathrm{C}}/(M_{^{13}\mathrm{C}} + M_{^{1}\mathrm{H}})$, which enables them to pass the injection magnet set for $^{12}C^-$. They are then accelerated together with the ^{12}C projectiles, are stripped and can change their charge again when interacting with residual gas molecules in the high energy-section of the accelerator [7]. A small fraction of them acquires the same magnetic and electric rigidity as the reaction product ^{13}N, which enables them to reach the rare-isotope detector (see Section 4).

The charge state distribution of the produced ^{13}N ions can in principle be derived from measurements with directly injected ^{14}N tuned to the same velocity as ^{13}N.

Based on the Q-value of the ^{1}H(^{12}C,γ)^{13}N reaction, which is 1.943 MeV, and a typical center-of-mass energy of about 230 keV the maximum deflection angle and the corresponding momentum spread could be calculated. Inside the stripper canal the reaction products get a maximum deflection angle of 8.4 mrad. Monte-Carlo simulation calculations of the reaction kinematics showed that about 60% of them leave the stripper canal. At the image slits' position of the 90° analyzing magnet, ions of the same charge state with $\Delta p/p = 1/1250$ are separated 1 mm from each other. This means, due to the reaction kinematics, the $^{13}N^{3+}$-'beam' width at the image slits is more than ±10 mm. This implies additional, not yet in detail quantified losses, along the rest of the analyzing beamline down to the rare-isotope detector.

3. Hydrogen as stripper gas in the terminal

In order to get information on the total stripping efficiency (i.e. sum of all charge states) of our system for H_2, we compared it with the total stripping efficiencies using Ar under identical conditions. ^{13}C accelerated at a terminal voltage of 2.77 MV was used for this purpose. The sum of all charge states was ~90% for Ar and ~70% for H_2 as stripper gas. The gas stripper device inside the terminal of the accelerator is a tube having 780 mm in length and 8 mm in diameter at its entrance and 9 mm in diameter at its exit, respectively. Turbo molecular pumps are placed at both ends, in order to recycle the stripper gas. Though differential pumping avoids excessive gas flow into the accelerator tubes, some gas escapes through the accelerator tubes towards the end of the accelerator. In comparison to heavy stripper gases like O_2 or Ar, H_2 escapes more easily from the stripper device. The possibility for charge exchange processes and scattering after the stripping process is therefore enhanced.

Beyond its purpose as gas stripper, hydrogen also acts as target for the ^{1}H(^{12}C,γ)^{13}N reaction. The determination of the cross-section requires a precise knowledge of the density of hydrogen atoms inside the gas stripper. Methods for the determination of this quantity were developed as a separate project. Two different physical effects are measurable that crucially depend on stripper gas density: charge changing of negative ions to positive ions, and energy loss. Both effects are most easily quantified by using H^- as projectiles since the necessary cross-sections are available with errors of about 20% or better. It has to be mentioned, however, that for a light gas like hydrogen, a quantity not easily measurable has an important influence: the purity of the stripper gas. Recirculating the stripper gas enriches gases heavier than H_2 more effectively. As gases like H_2O, O_2, or Ar have significantly higher energy loss and charge-changing cross-sections, their probable presence in percent quantities limits the

accuracy of the derived H_2 density. Our best estimate for the present experiment is $1 :\rightarrow 10^{17}$ H_2 molecules per cm^2, if the stripper gas is pure H_2, and $0.5 :\rightarrow 10^{17}$ H_2 molecules per cm^2, if the stripper contains about 10% heavier gases.

Independently, the amount of target nuclei was estimated from the measurements of the H_2 gas pressure using a thermocouple gauge (calibrated for air) placed in the middle of the stripper canal. Assuming a heat conductivity of $0.18\,J\,m^{-1}\,s^{-1}\,K^{-1}$ for H_2 and of $0.03\,J\,m^{-1}\,s^{-1}\,K^{-1}$ for ambient air, a reading of 47 μmHg (63 mbar) in H_2 results in a 'true' value of ~10 μbar of H_2 at the center of the stripper canal. Attempts to measure the H_2 target thickness trough energy loss measurements of a ^{197}Au beam turned out to give unreliable results.

4. Measurements

As a viability test for this technique, we investigated the $^1H(^{12}C,\gamma)^{13}N$ radiative capture reaction at energies for which the cross-section is known: i.e. $E^{CM} = 206.8$ keV ($\sigma' = 79$ nb [8]) and $E^{CM} = 229.5$ keV ($\sigma' = 130$ nb [9]). These center-of-mass energies were achieved by accelerating the $^{12}C^-$ projectiles (having an injection energy of 69 keV) with terminal voltages of $V_T = 2619.3$ kV and 2915.3 kV, respectively. The energies of the produced $^{13}N^{3+}$ ions after the second acceleration step were 10 338 keV and 11 499 keV, respectively, which were high enough to identify unambiguously the ^{13}N nuclei and to separate them from the isobar contamination ^{13}C (Fig. 1). The position of the $^{13}N^{3+}$ region in the ΔE–E spectrum was deduced from the positions of $^{13}C^{3+}$ and $^{14}N^{3+}$. These two isotopes, which were separately tuned, were used for the energy calibration of the rare-isotope detector. The number of projectiles was measured every 600 s at the entrance of the accelerator. In order to obtain the dependence of the overall transmission on the tuned charge state, additional measurements at $E^{CM} = 229.5$ keV for $q = 2^+$ and $q = 4^+$ were carried out. In a second measurement, performed about one year after

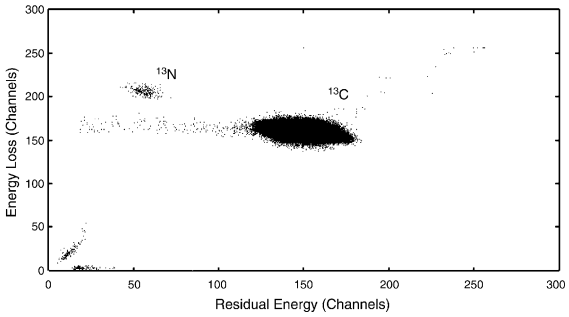

Fig. 1. ΔE–E spectra taken at a center-of-mass energy $E^{CM} = 229.5$ keV. Analyzing beamline tuned for charge state 3+. The products of the $^1H(^{12}C,\gamma)^{13}N$ reaction are clearly separated from the background particles. See last column of Table 1 for more detailed information.

Table 1
Summary of the experimental results used for deducing the transmission T from the production target to the rare-isotope detector (see Section 2 for relevant formulas)

E^{CM} (keV)	Charge state	Accumulation time (s)	Number of ^{13}N detected	Stripper gas pressure[a] (μbar)	Nominal cross-section (nb)	Mean $^{12}C^-$ current (μA)	Deduced transmission[b] (%)
229.5	3+	60 100	123	1	130[c]	10	6
229.5	4+	2700	2	1	130[c]	12	2
229.5	2+	48 300	4	1	130[c]	7	0.3
206.8	3+	61 200	22	1	79[d]	4.5	4
229.5[e]	3+	35 059	243	6	130[c]	8.8	4

[a] The gas pressure is measured in the middle of the gas stripper (assumption for the calculation of T: constant pressure inside the stripper canal). The target thickness (stripper length: 78 cm) for 1 μbar is 4×10^{15} H atoms cm^{-1}.
[b] Due to the rather large uncertainty in the number of target nuclei, no uncertainties are reported.
[c] Rolfs and Azuma [9].
[d] Vogl [8].
[e] The results given in the bottom row were measured in July 2004, whereas the others are results from measurements performed in July 2003.

the first at $E^{CM} = 229.5$ keV we could reproduce the value for the overall transmission of the 3+ charge state (see Table 1).

5. Conclusions

We explored an alternative method for the determination of extremely low astrophysical reaction rates. It is based on the merit of the AMS technique for identifying small amounts of reaction products. Whereas previous measurements [2–4] determined cross-sections from thick-target yields, the present measurement is performed on a thin (hydrogen) target with well-defined reaction energy. However, due to the uncertainties in the determination of the stripper-target thickness, the acceptance of the analyzing beamline of VERA, and the variability of the transmission with energy, a reliable extrapolation of the normalizing factor, F, to much lower energy is currently not feasible. In order to reach energies relevant for stellar reaction rates an improved determination of the above-mentioned quantities must be implemented, and reaction rates over a larger energy range of known cross-sections should be measured.

Nevertheless, we believe that our experiment demonstrates the principle usefulness of combining AMS with an on-line measurement of small reaction cross-sections.

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