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Neutron production by a ^{13}C thick target irradiated by 20–90 MeV protons

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Abstract

Neutron production using an enriched ^{13}C carbon converter has been measured during the design study of the italian RIB facility SPES. Energy and angular distributions of neutrons emitted by bombarding a ^{13}C target of stopping length with protons in the range of 20 to 90 MeV have been measured by time-of-flight and activation and compared with the prediction of a Monte Carlo code developed at Snezhinsk. At the proton energy of 100 MeV, firstly envisaged for SPES, the gain with respect to a natural C target is less than a factor of two, while yields still compare well with those for 40 MeV deuterons on natural carbon adopted by SPIRAL-II. At energies near 30 MeV the ^{13}C thick target is definitely more prolific than the target of natural carbon, but both yields with protons are clearly lower than the one with deuterons. At the energy of 20 MeV envisaged for a first stage of SPES it might be more efficient to irradiate the uranium target with protons rather than using the two-stage method with converter.

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Keywords: Neutron yield; Thick target; ^{13}C; Protons

1. Introduction

The two-step method for production of intense radioactive beams of neutron-rich nuclei is the scheme considered in the early design of the italian facility SPES (Study for Production of Exotic Species) [1], adopted for SPIRAL-II at GANIL [2] and possibly by the future EURISOL facility [3]. A thick target, the so-called converter, stops the charged-particle beam and generates a neutron flux, subsequently used to induce fissions in a target placed in its vicinity. It is well known that the advantage of this scheme is that it allows high intensities by removing the thermal load due to the electronic stopping power from the fissile target. Very thick targets (of about 1 kg and dimensions near 10 cm) should be developed to take advantage of the longer attenuation length of the neutrons. Our studies of release processes with high-density UC targets are the subjects of other contributions [4,5]. Yet, an issue is the conversion of the charged-particle beam into neutrons in terms of absolute numbers but also angular distribution

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and energy spectrum. For reasons of synergies the beam retained for SPES is a proton beam, of 100 MeV for the full facility and of 20 MeV in a first stage. In contrast to the deuterons chosen for SPIRAL-II protons do not provide the extra mechanism of projectile breakup. Nevertheless, it was conjectured that a highly enriched ^{13}C thick target, as the threshold for (p,n) is lowered from 18 MeV for ^{12}C to 3 MeV, could compensate for the absence of projectile breakup. A first neutron production measurement by time-of-flight was carried out with the HENDES detectors at JYFL-Jyväskylä (Finland) with 30 MeV protons [6] and fully supported this assumption. Subsequently, measurements were undertaken at proton energy close to 100 MeV at the AGOR cyclotron of KVI, Groningen, and then at 20, 25 and 40 MeV at the K-130 JYFL cyclotron, Jyväskylä, in order to evaluate the production for the first stage of SPES. We report briefly these measurements here, since they have been published recently [7,8] and compare the merits of diverse schemes for production of neutron-rich nuclei.

2. Experimental method

2.1. Time-of-flight

In all time-of-flight experiments the neutron detectors were liquid scintillators capable of neutron/γ discrimination based on the difference of pulse shapes.

At KVI-Groningen, the EDEN detectors were placed in two rings covering the same angles. In one of the rings the EDEN modules were sheltered by a lead foil to prevent triggering by occasionally scattered protons and to reduce the γ background. Comparison of event parameters (time and amplitude) in shielded and non-shielded detectors helped to identify true events due to neutrons only. The reference time trigger was generated by the detection of a proton in a phoswich detector upstream of the target.

At Jyväskylä, the HENDES detectors were used. A HENDES detector module is a 1 m long bar with its axis placed at right angle to the incoming neutrons in order to cover a wide solid angle. The time signals of photomultipliers placed at both ends of the bar allow for determination of the location of the impact and for its time. The reference time was given by the cyclotron radio-frequency. With the HENDES set-up the recorded angular distributions are continuous. They are subsequently binned for the ease of presentation.

2.2. Activation

Activation was used in all measurements. Foils placed at various angles θ with respect to the beam axis were submitted to the neutron flux. The basic quantity derived from off-line γ-ray counting of the activities generated during irradiation is an integral y_k, where k is an index for the reaction channel, σ_k the corresponding cross-section and ϕ the neutron flux:

$$y_k = \int_{E_{\min}}^{E_{\max}} \sigma_k(E_n) \frac{d^2\phi}{dE_n d\Omega} dE_n$$

The integration limits are the cross-section threshold ($E_{\min}$) and, by definition, the beam energy ($E_{\max}$). At each angle θ the spectrum shape is deduced by iterations of a trial function in order to reproduce the set of experimental y_k values at this angle. Obviously, the spectrum should exhibit a smooth variation with energy so that it can be well described by few parameters (which, however, need to be less than the number of reaction channels). Our trial function is a mere interpolation between $(E_n, d^2\phi/dE_n d\Omega)$ pairs where the double-differential flux values are to be adjusted. The activation method works very well at a proton energy of 90 MeV, mainly owing to the cross-sections for Bi(n,xn) typically separated by 10 MeV available above a neutron energy of 25 MeV. At lower proton energy, the narrower neutron energy range becomes comparable with the width of cross-sections so that the unfolding method becomes less sensitive to the spectrum shape. It seems hardly possible to use it with the set of Al, Co, Ni, In and Bi foils if the neutron range is less than 20 MeV. Yet, by inserting the spectrum shape obtained from TOF it still provides a check of proper scale, i.e. of the total conversion proton-to-neutron factor.

3. Results

Examples of neutron spectra obtained at the proton energies of 25 and 90 MeV are shown in Figs. 1 and 2.

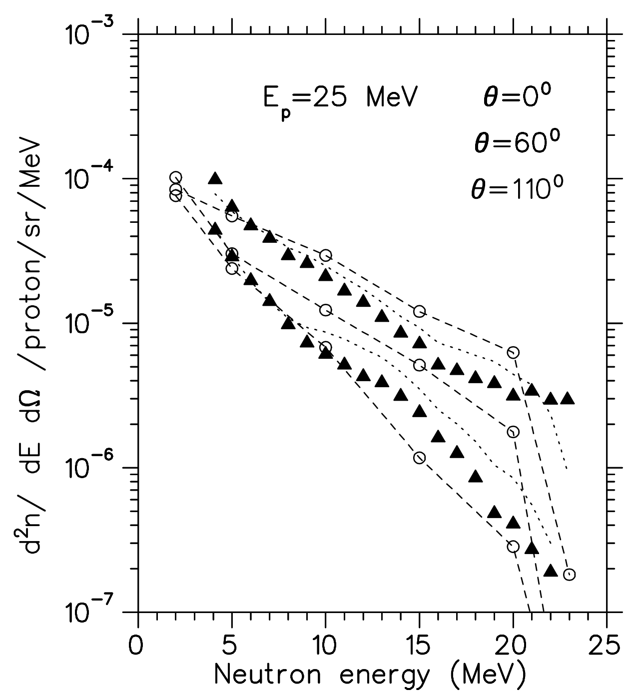

Fig. 1. Some neutron spectra measured at JYFL for 25 MeV $p+^{13}C$ at various angles, showing TOF (triangles) and activation data (circles are the parameter pairs). Dotted lines are the adopted values. There is no TOF spectrum at 110° due to a technical problem.

At low proton energy the agreement of TOF and activation is not very satisfactory. Activation systematically yields a higher production than TOF and the excess increases with increasing angle with respect to the beam. At $E_p = 25$ MeV the deviation may reach near a factor of two at $\theta = 90°$. As we could not find the origin of this discrepancy, we adopted the geometric mean values. These data are thus estimated to have errors of up to a factor 1.4 up or downwards. At the proton energy of 90 MeV the results of both methods are in good agreement.

The data have been compared with the prediction of a Monte Carlo code (PRIZMA) written by the Snezhinsk group. The bench-marking tests with $p+^{12}C$ show that the calculated productions compare well with the experimental values available from 30 to 113 MeV, which actually are references for the widely used MCNPx code. We show a comparison of experimental and calculated neutron spectra for $p+^{13}C$ in Figs. 3 and 4.

The calculations for $p+^{13}C$ were delivered without further tuning before the experiments were completed. The good agreement with the data confirm that the PRIZMA code is very suitable for modelling carbon converters. The most salient discrepancies are an overprediction of the high-energy spectrum near proton energy at the lowest proton energies, see Fig. 3 and another overprediction of the low-energy spectrum at the most forward angles at proton energy of 90 MeV, Fig. 4. It might be accidental, but perhaps worth to note, that the agreement with data from activation alone is better than with the adopted values and in fact very good.

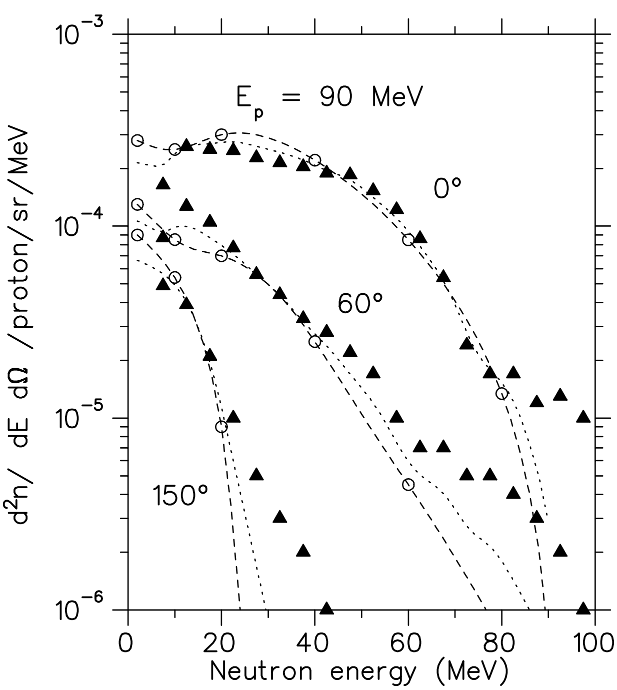

Fig. 2. Some neutron spectra measured at KVI for 90 MeV $p+^{13}C$ at various angles. Symbols are like in Fig. 1.

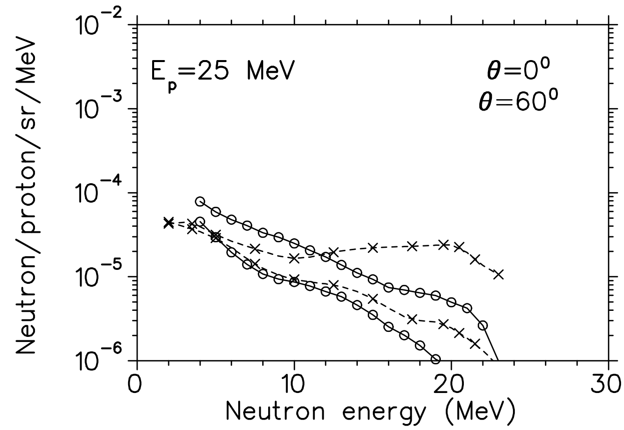

Fig. 3. Comparison of neutron spectra from experiments and simulation at proton energy of 25 MeV. Circles are the adopted values from TOF and activation. The Monte-Carlo calculation with the PRIZMA code is shown by crosses.

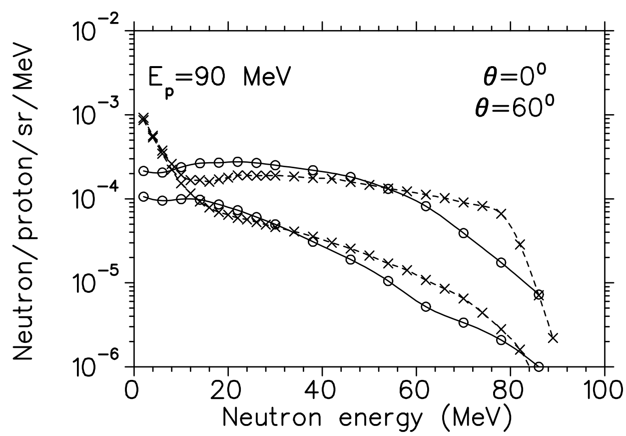

Fig. 4. As Fig. 3 but at 90 MeV proton energy.

4. Discussion

The main outcome of these measurements is twofold. At 90 MeV proton energy the ^{13}C target provides only a small gain, less than a factor of two, with respect to the target of natural carbon. Secondly, the promising measurement at 30 MeV [6] that predicted this reaction to be competitive with deuterons of same energy on natural carbon is not confirmed. Although the shape of neutron energy and angular distributions looks to be correct, the yield has been strongly overestimated. The unexpected evolution of yield from $E_p = 30$ to 90 MeV prompted a paper by the Louvain-La-Neuve group [10], in which they report a much lower value, actually in fair agreement with our new work.

Some alternative methods of neutron production in the intermediate energy range are shown in Fig. 5. The neutron flux has been integrated over all energies above 4 MeV and on a solid angle defined by θ values up to 30°. This corresponds somehow to the flux that shall be experienced by a fissile target in form of a disk placed downstream of the

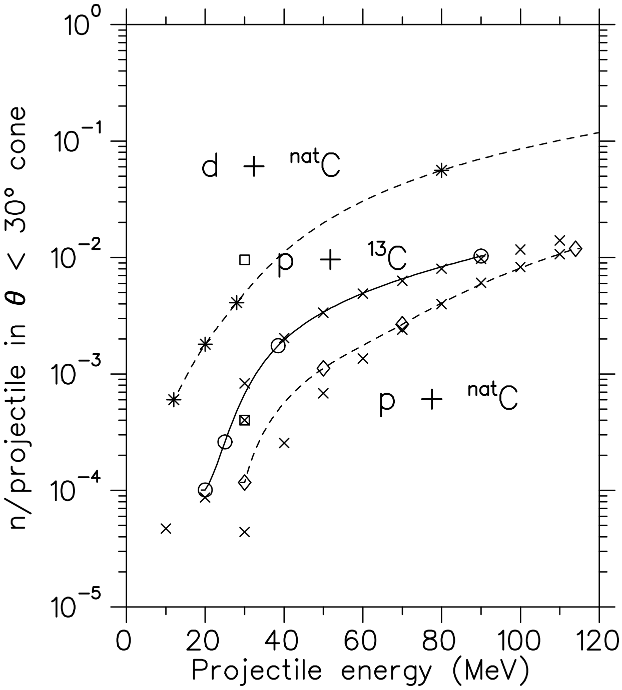

Fig. 5. Comparison of energy and solid angle integrated neutron yield per projectile. Our measurements for protons on ^{13}C are marked by circles connected by the solid line. The squares at 30 MeV are from [6], the highest one, and from the Louvain-La-Neuve group [10], in better agreement with ours. For comparison are shown $p +$ natural carbon used to benchmark MCNPx (diamonds). Calculations with PRIZMA for protons on both targets are marked with crosses. Stars are from works of the GANIL and IPN-Orsay groups for deuterons on natural carbon [11].

converter. The 100 MeV protons aimed at for SPES would be competitive with the 40 MeV deuterons of SPIRAL-II. At low-energy, yields with protons, in spite of the improvement visible for ^{13}C, are too low. Typically a proton-to-neutron conversion factor of one thousand has to be overcome. Possibly, a "direct" solution without converter would be more suitable at low proton energy, in analogy with the already operating Oak-Ridge facility operating with 10 μA of 40 MeV protons. The feasibility of a similar concept but at a beam current of 200 μA is now being investigated at Legnaro [9].

We finally note that regarding the application to nuclide production by fission of natural uranium the isotope distribution via the direct method is centered on less neutron-rich nuclei than via the converter method, the shift being about 2 mass units for a given element [12]. In contrast, and somewhat surprisingly, there is little difference when using intermediate energy protons or deuterons with a converter, in spite of the lower average neutron energy of the neutron spectrum generated with protons on ^{13}C [13].

5. Conclusion

The neutron production by protons on an enriched ^{13}C target has been measured in the range of 20–90 MeV. It remains lower than the one of deuterons on natural carbon at the same beam energy. An increase of the proton energy by about 2–3 times is needed to match the performance of deuterons on natural carbon. The low yields obtained below 30 MeV suggest that the two-stage method with a converter irradiated with protons is not very competitive with respect to the deuteron solution and possibly even with the direct method.

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