Experimental determination of the $^{14}N(n,p)^{14}C$ reaction cross section for thermal neutrons

J. Wagemans
E. C., Joint Research Centre, Institute for Reference Materials and Measurements, Retieseweg, B-2440 Geel, Belgium

C. Wagemans and G. Goeminne
Department of Subatomic and Radiation Physics, RUG, Proeftuinstraat 86, B-9000 Gent, Belgium

P. Geltenbort
Institut Laue-Langevin, Boîte Postale 156, F-38042 Grenoble, France
(Received 6 January 2000; published 28 April 2000)

The $^{14}N(n_{th},p)^{14}C$ reaction cross section was determined at the high flux reactor of the ILL in Grenoble using various polyimide and adenine samples. We obtained a precise value of (1.93 ± 0.05) b for the cross section. A comparison is made with the currently available results in the literature and the astrophysical context is briefly discussed. Also the previously determined $^{17}O(n_{th},\alpha)^{14}C$ cross section is renormalized taking into account this new result.

PACS number(s): 25.40.Fq, 27.20.+n, 97.10.Cv

I. INTRODUCTION

The $^{14}N(n,p)^{14}C$ reaction plays an important role in the *s* process of nucleosynthesis: ^{14}N is very abundant since it is a dominant product of hydrogen-burning in the CNO cycle, the stage prior to the *s* process. So with its relatively high cross section, this reaction can act as a strong neutron poison in the reaction chain to heavier elements. Also, ^{14}N is of crucial importance in the much debated nucleosynthetic origin of fluorine, whose only stable isotope is ^{19}F. The He-burning shell in asymptotic giant branch stars is thought to be the most likely site for the synthesis of fluorine, mainly through the nuclear chain $^{14}N(\alpha,\gamma)^{18}F(\beta+)^{18}O(p,\alpha)^{15}N(\alpha,\gamma)^{19}F$. The protons captured by ^{18}O are produced in the $^{14}N(n,p)^{14}C$ and to a lesser extent in the $^{26}Al(n,p)^{26}Mg$ reaction by neutrons from the $^{13}C(\alpha,n)^{16}O$ neutron source [1].

The first direct measurement of the stellar $^{14}N(n,p)^{14}C$ cross section using neutrons with a quasi-Maxwellian distribution at the astrophysically relevant temperatures ($kT=25$ keV and $kT=52.4$ keV) was done by Brehm *et al.* [2] in 1988. Their result for the reaction rate was about a factor of three smaller than the rate used in most of the previous *s*-process calculations. It was also 2–3 times smaller than rates estimated from the inverse reaction and extrapolations from the thermal cross section, for which an evaluated value of 1.83 b was adopted. Koehler *et al.* [3,4] performed measurements from thermal neutron energy up to 35 keV and found clear evidence for a 1/v behavior of the $^{14}N(n,p)^{14}C$ reaction cross section up to approximately 30 keV. Since they used a thermal value of 1.83 b for the normalization of their data, also their results for the stellar reaction rate at $kT=25$ keV are about a factor of 3 higher than reported by Brehm *et al.* [2]. Measurements with quasimonoenergetic neutrons at 25 keV from Gledenov *et al.* [5] are in fair agreement with the results from Koehler *et al.* [3,4] and with the estimates from the inverse reaction, since again the same thermal value was used for the normalization. Another direct measurement of the $^{14}N(n,p)^{14}C$ stellar cross section at $kT=25$ keV was done by Sanami *et al.* [6]. They found a value approximately a factor of 2 higher than Brehm *et al.* [2] and a rather good agreement with the other results. Last year Kii *et al.* [7] performed measurements at neutron energies of 35.8 and 67.1 keV. Their results support the recently measured values but have rather large (20% and 12%, respectively) uncertainties.

It is clear that the value for the $^{14}N(n_{th},p)^{14}C$ cross section plays a crucial role in all these comparisons and hence needs to be verified: there are indeed still discrepancies between direct measurements, calculations, and measurements normalized on the thermal cross section value. This doubtful situation is illustrated by the fact that several authors use different reaction rates in their nucleosynthesis network calculations [8–10]. An additional reason for a careful verification of the $^{14}N(n_{th},p)^{14}C$ cross section is its role as a reference cross section for the determination of the $^{17}O(n_{th},\alpha)^{14}C$ cross section [11].

So far, five experimental results are reported for the thermal cross section value varying from 1.72 to 1.93 b, so there is more than 10% difference between the extreme values. The available values for the $^{14}N(n_{th},p)^{14}C$ cross section (renormalized to updated values of the reference cross sections used in the original works) and their references are summarized in Table I. The recommended value in the ENDF-B6 data file is (1.85 ± 0.07) b.

II. EXPERIMENTAL PROCEDURE AND MEASUREMENTS

We installed a dedicated setup at the end of the curved neutron guide H22D of the high flux reactor at the ILL in Grenoble (France). The thermal neutron flux at the sample position reached a value of about 5×10^8 n/cm^2 s with a negligible background of epithermal and fast neutrons and γ rays. This enabled a clean detection of the low-energy protons (0.6 MeV) emitted in the $^{14}N(n_{th},p)^{14}C$ reaction. The energy distribution of the neutrons approached a Maxwellian distribution with $kT\approx18$ meV (corresponding to a temperature $T\approx210$ K).

TABLE I. The renormalized literature values for the thermal $^{14}N(n,p)^{14}C$ cross section.

Cross section (b)	Uncertainty (b)	Year of publication, reference
1.83	0.07	1993, [12]
1.83	0.03	1961, [13]
1.93	0.10	1951, [14]
1.92	0.05	1949, [15]
1.75	0.04	1949, [16]

For the particle detection, we consecutively made use of two fully depleted silicon surface barrier detectors. The first one is 29 μm thick, has a surface of 150 mm^2, and has 40 keV resolution and the second detector is 60 μm thick, has a surface of 150 mm^2, and has 25 keV resolution. The energy calibration was done by means of the $^{10}B(n,\alpha)^7Li$ and $^6Li(n,\alpha)t$ reactions. The sample was mounted in a vacuum chamber at 30° with respect to the neutron beam axis. A surface barrier detector was mounted parallel with the sample out of the beam and slightly collimated in order to avoid detection of particles under small incident angles. A schematic view of the experiment is shown in Fig. 1.

The thermal neutron flux determination was done by means of the $^{235}U(n_{th},f)$ reaction using several U samples with a well-determined number of atoms, strictly maintaining the same detection geometry. The flux was verified to be perfectly constant during the short time intervals of the different measurements. For the thermal fission cross section a value of (584.25 ± 1.10) b was adopted as reported in the ENDF-B6 data file. A typical spectrum is shown in Fig. 2.

A variety of ^{14}N samples was used: adenine ($C_5H_5N_5$) containing 51.6% ^{14}N evaporated on thin Al backings and polyimide ($C_{22}H_{10}O_5N_2$) foils containing 7.30% ^{14}N. In Fig. 3, a typical $^{14}N(n_{th},p)^{14}C$ spectrum obtained with an adenine sample is shown. The homogeneity of the samples was tested by doing measurements with a collimated neutron beam.

Several background corrections had to be done. The

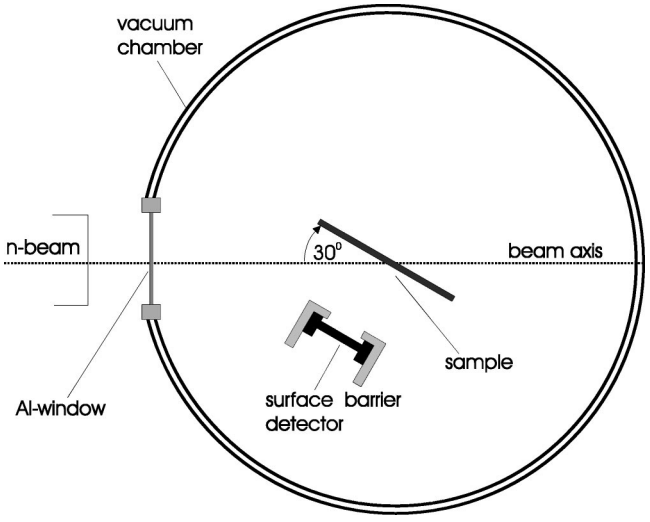

FIG. 1. A schematic view of the experimental setup.

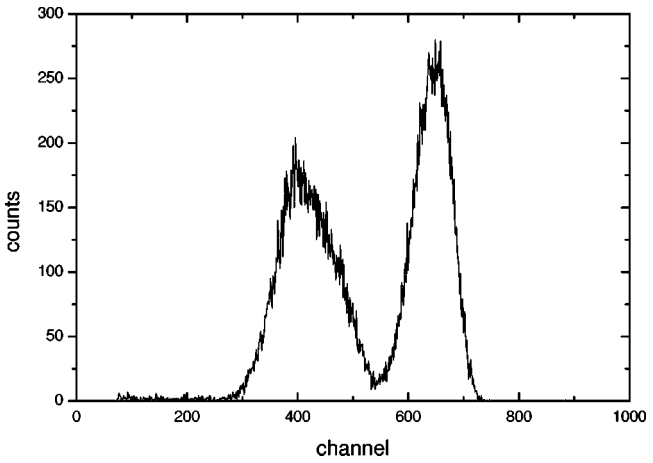

FIG. 2. Fission fragments detected during the $^{235}U(n_{th},f)$ flux calibration.

vacuum during the measurements was about 10^{-3} torr. Since air consists for 77.8% out of ^{14}N, a few protons coming from reactions with the remaining air in the chamber were detected. This contribution can be determined by performing measurements without sample and with a dummy (Al backing) in the neutron beam under the same vacuum conditions as during the actual $^{14}N(n_{th},p)^{14}C$ measurement. Another source of background comes from the interaction of neutrons with boron impurities present in the samples. This is a consequence of the very large cross section (3842 b) of the $^{10}B(n_{th},\alpha_0)^7Li+^{10}B(n_{th},\alpha_1\gamma)^7Li^*$ reactions. Especially the $^7Li^*$ particles are disturbing, because they are detected in the same energy region as the protons. Since the ratio α_0/α_1 (and hence $^7Li^*/^7Li$) for the $^{10}B(n,\alpha)^7Li$ reaction is well known $(6.733\pm0.008\%)$ [17] and the number of detected α particles can easily be integrated ($E_{\alpha_0}=1.8$ MeV, $E_{\alpha_1}=1.5$ MeV), this background contribution can be well estimated.

III. RESULTS AND DISCUSSION

The $^{14}N(n_{th},p)^{14}C$ reaction cross section is determined relative to the $^{235}U(n_{th},f)$ reaction using the following for-

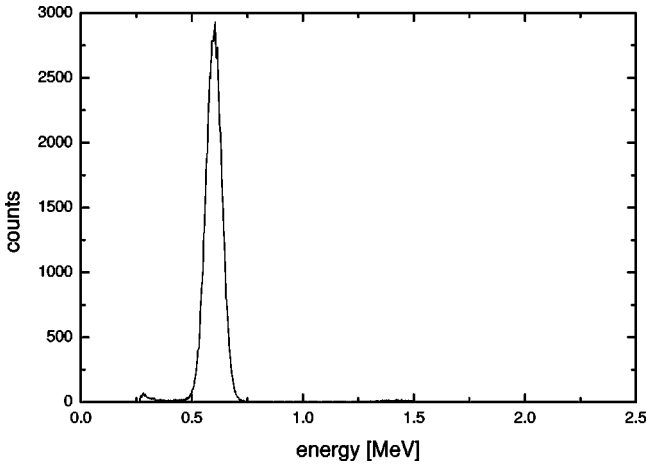

FIG. 3. Energy distribution for the $^{14}N(n_{th},p)^{14}C$ reaction obtained with an adenine sample after 45 min of data taking.

TABLE II. The cross-section values obtained with the different adenine and polyimide samples.

Sample composition	Density (μg cm^{-2})	Cross section (b)
$C_5H_5N_5$	204.0±1.7	1.98±0.04
$C_5H_5N_5$	94.3±1.7	1.88±0.05
$C_5H_5N_5$	70.7±1.7	2.02±0.06
$C_{22}H_{10}O_5N_2$	64.2±1.3	1.97±0.06
$C_{22}H_{10}O_5N_2$	60.5±1.2	1.95±0.05
$C_{22}H_{10}O_5N_2$	55.0±1.7	1.81±0.08
$C_{22}H_{10}O_5N_2$	52.5±1.6	1.87±0.08

mula [18]:

$$\sigma_p = \frac{N(^{235}\mathrm{U})}{N(^{14}\mathrm{N})} \frac{Y_p(^{14}\mathrm{N})}{Y_f(^{235}\mathrm{U})} \frac{g(T)(^{235}\mathrm{U})}{g(T)(^{14}\mathrm{N})} \sigma_f, \tag{1}$$

where $N(^{235}\mathrm{U})$ and $N(^{14}\mathrm{N})$ are the number of atoms/cm^2 of the samples, $Y_f(^{235}\mathrm{U})$ and $Y_p(^{14}\mathrm{N})$ are the counting rates of the $^{235}\mathrm{U}(n_{\mathrm{th}},f)$ and $^{14}\mathrm{N}(n_{\mathrm{th}},p)^{14}\mathrm{C}$ reactions, $g(T)(^{235}\mathrm{U})$ and $g(T)(^{14}\mathrm{N})$ are the corresponding Westcott factors at a neutron temperature T, and σ_f is the $^{235}\mathrm{U}(n_{\mathrm{th}},f)$ reference cross section. Since Koehler *et al.* found that the $^{14}\mathrm{N}(n,p)^{14}\mathrm{C}$ reaction cross section follows a 1/v shape below approximately 30 keV [3,4], $g(T)(^{14}\mathrm{N})=1$ is adopted. Wagemans *et al.* [18] reported that for the $^{235}\mathrm{U}(n_{\mathrm{th}},f)$ reaction $g(T)(^{235}\mathrm{U})=(0.995\pm0.002)$ for the neutron spectrum used.

The cross section values and their respective uncertainties obtained in this way for the seven different ^{14}N samples are shown in Table II. In order to calculate a final $^{14}\mathrm{N}(n_{\mathrm{th}},p)^{14}\mathrm{C}$ cross section value, we applied the following method. The counting rates for the $^{14}\mathrm{N}(n_{\mathrm{th}},p)^{14}\mathrm{C}$, $^{235}\mathrm{U}(n_{\mathrm{th}},f)$ reactions, respectively, can be written as follows:

$$Y_p(^{14}\mathrm{N}) = \sigma_p \times N(^{14}\mathrm{N}) \times \Phi \times S \times g(T)(^{14}\mathrm{N}), \tag{2}$$

$$Y_f(^{235}\mathrm{U}) = \sigma_f \times N(^{235}\mathrm{U}) \times \Phi \times S \times g(T)(^{235}\mathrm{U}), \tag{3}$$

where Φ is the thermal neutron flux and S the sample surface (equal for all the ^{14}N and ^{235}U samples). Transformation of Eqs. (2) and (3), adopting $g(T)(^{14}\mathrm{N})=1$ and $g(T)(^{235}\mathrm{U})=0.995$, gives

$$\frac{Y_p(^{14}\mathrm{N})}{S\times\Phi} = \sigma_p \times N(^{14}\mathrm{N}), \tag{4}$$

where the thermal neutron flux Φ is given by

$$\Phi = \frac{Y_f(^{235}\mathrm{U})}{\sigma_f \times N(^{235}\mathrm{U}) \times 0.995}. \tag{5}$$

This means that if we plot the normalized proton counting rate $Y_p(^{14}\mathrm{N})/S\times\Phi$ versus the number of ^{14}N atoms $N(^{14}\mathrm{N})$ in the appropriate units, we should obtain a straight line with as slope the $^{14}\mathrm{N}(n_{\mathrm{th}},p)^{14}\mathrm{C}$ cross section value σ_p. Since our data points have uncertainties on both the abscissa x and the ordinate y, it is not straightforward to make a weighted linear fit. Therefore, we calculated for each data point a generalized uncertainty on the ordinate following the effective variance method [19,20],

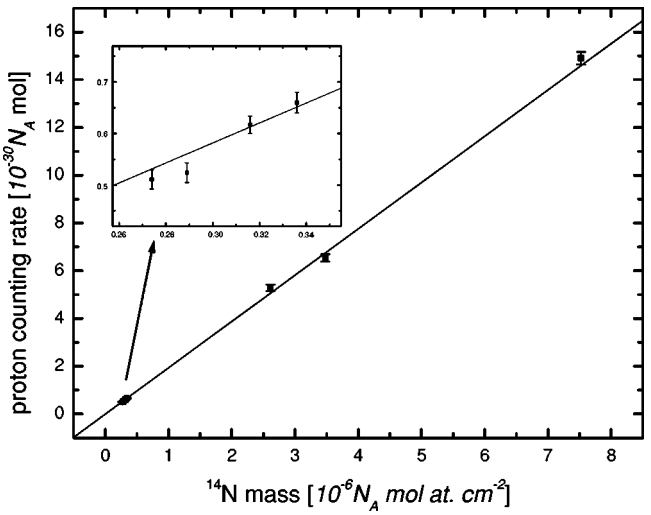

FIG. 4. A weighted linear fit through the origin for the normalized proton counting rates versus the ^{14}N mass. The slope is the $^{14}\mathrm{N}(n_{\mathrm{th}},p)^{14}\mathrm{C}$ reaction cross section σ_p.

$$\delta^2 = \left(\frac{\partial f}{\partial x}\right)^2 (\delta_x)^2 + (\delta_y)^2 \tag{6}$$

within our case $(\partial f/\partial x)=\sigma_p$. Since we plot the net counting rate versus the ^{14}N mass, we can add as additional point (0,0) with zero uncertainty in order to obtain a more precise linear fit. The result of this weighted linear fit through the origin (Fig. 4) is $\sigma_p=(1.93\pm0.03)$ b with regression coefficient $R=0.999\,18$.

Special attention has to be given to the treatment of the uncertainties. It is recommended to add a systematic uncertainty as can be seen from Fig. 5. Here we plotted the same quantities as in Fig. 4 but now for the three adenine (top) and four polyimide (bottom) samples separately. When again a weighted linear fit through the origin is applied we find $\sigma_p=(1.95\pm0.04)$ b for the adenine samples and $\sigma_p=(1.90\pm0.04)$ b for the polyimide samples. This tendency to higher, respectively, lower values might indicate that there is some ''hidden'' systematic uncertainty in the mass determination. Indeed, two different methods were applied: the adenine samples were all calibrated via differential weighing and the masses of the polyimide foils were determined via spectrophotometric transmission and reflection measurements. Moreover, the uncertainty from the linear fit is obtained by supposing that all the values are independent. This gives a slight underestimation when the uncertainties of the different values are partly dependent. We, therefore, prefer a conservative approach by adding 1% to the obtained uncertainty, which results in a final value for the $^{14}\mathrm{N}(n_{\mathrm{th}},p)^{14}\mathrm{C}$ reaction cross section of $\sigma_p=(1.93\pm0.05)$ b.

When comparing our results with those given in Table I, several things can be noticed. As mentioned before, there is more than 10% difference between the extreme values. Nevertheless some authors claim to have a very small uncer-

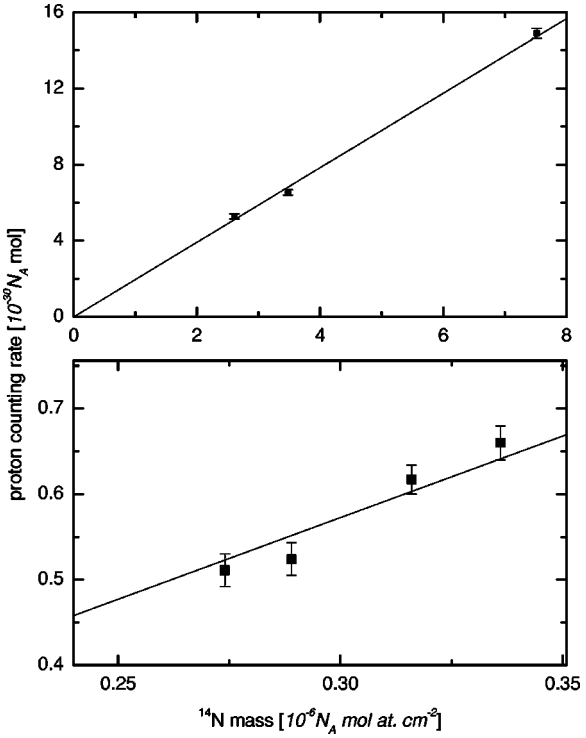

FIG. 5. Same as Fig. 4 but now with the adenine (top) and polyimide (bottom) samples taken separately.

tainty, sometimes even less than 2%. It is clear that when making a weighted average of our result with the five results from Table I, those values with a very small uncertainty will almost completely determine the final result. Indeed, a weighted average of the six values results in $\sigma_p=(1.84\pm0.02)$ b. When having a closer look at the previous experiments we see that in our work a better resolution and better background conditions were obtained since we could take advantage of more suitable detectors and of a cleaner neutron beam. It cannot be excluded that previous experiments were also subject to some hidden uncertainty and that therefore the quoted uncertainties are slightly underestimated. For this reason we prefer not to make a weighted average but instead we will use the value determined in the present work as the normalization factor in the Appendix.

IV. CONCLUSION

In the present work we have determined an accurate value of (1.93 ± 0.05) b for the $^{14}N(n_{th},p)^{14}C$ cross section. When comparing this value with the previously found results we see that it is in good agreement with some of them but differs by 10% with the lower extreme value. We believe that a careful new evaluation is needed in order to come to a new recommended value.

The present result confirms that the $^{14}N(n_{th},p)^{14}C$ reaction is a strong neutron poison in the *s* process of stellar nucleosynthesis. It also supports the idea that it can act as an important proton supplier for the synthesis of ^{19}F.

ACKNOWLEDGMENTS

This Research was sponsored by NATO Collaborative Research Grant No. 960455 and by the Fund for Scientific Research Flanders.

APPENDIX

In 1998 Wagemans *et al.* [11] reported a value of (244 ± 7) mb for the $^{17}O(n_{th},\alpha)^{14}C$ reaction cross section. A series of measurements was performed with five different gas samples with ^{17}O enrichments of 58.2 and 85.5 at. % resulting in eight experimental values for the $^{17}O(n_{th},\alpha)^{14}C$ cross section. The thermal neutron flux was determined via the $^{14}N(n_{th},p)^{14}C$ reaction adopting a cross section value of $\sigma_p=(1.83\pm0.03)$ b as recommended at that time.

As an additional verification that the result is not influenced by the degree of enrichment of the oxygen gas, we performed a new measurement with 72.1 at. % enriched ^{17}O gas using the same experimental technique as described in detail in [11]. This resulted in $\sigma_\alpha=(240\pm8)$ mb and $\sigma_\alpha=(241\pm8)$ mb for the two detectors, confirming the value reported in [11]. A weighted average was calculated for these two results together with the eight previously obtained values using the statistical errors as weight factors. In addition we now use for the normalization value $\sigma_p=(1.93\pm0.05)$ b instead of the previously adopted $\sigma_p=(1.83\pm0.03)$ b. In this way we obtain a new value for the thermal $^{17}O(n_{th},\alpha)^{14}C$ cross section of $\sigma_\alpha=(257\pm10)$ mb. Note that the increase in uncertainty is fully due to the adoption of a very conservative value for the uncertainty of the $^{14}N(n_{th},p)^{14}C$ reaction cross section.

[1] M. Forestini, S. Goriely, A. Jorissen, and M. Arnould, Astron. Astrophys. **261**, 157 (1992).
[2] K. Brehm, H. W. Becker, C. Rolfs, H. P. Trautvetter, F. Kappeler, and W. Ratynski, Z. Phys. A **330**, 167 (1988).
[3] P. E. Koehler and H. A. O'Brien, Phys. Rev. C **39**, 1655 (1989).
[4] P. E. Koehler, Phys. Rev. C **48**, 439 (1993).
[5] Y. M. Gledenov, V. I. Salatski, P. V. Sedyshev, M. V. Sedysheva, V. A. Pshenichnyj, and J. Andrzejewski, Z. Phys. A **348**, 199 (1994).
[6] Sanami *et al.*, Nucl. Instrum. Methods Phys. Res. A **394**, 368 (1997).
[7] T. Kii, T. Shima, H. Sato, T. Baba, and Y. Nagai, Phys. Rev. C **59**, 3397 (1999).
[8] A. Jorissen, V. V. Smith, and D. L. Lambert, Astron. Astrophys. **261**, 164 (1992).
[9] M. Forestini and C. Charbonnel, Astron. Astrophys. **123**, 241 (1997).
[10] N. Mowlavi, A. Jorissen, and M. Arnould, Astron. Astrophys. **334**, 153 (1998).
[11] J. Wagemans, C. Wagemans, R. Bieber, and P. Geltenbort, Phys. Rev. C **58**, 2840 (1998).
[12] Y. M. Gledenov, V. I. Salatski, and P. V. Seyshev, Z. Phys. A **346**, 307 (1993).

[13] G. C. Hanna, D. R. Primeau, and P. R. Tunnicliffe, Can. J. Phys. **39**, 1784 (1961).
[14] P. Cüer, J. P. Longchamp, and S. Gorodetzky, J. Phys. Radium (Suppl.) **12**, 6S (1951).
[15] J. H. Coon and R. A. Nobles, Phys. Rev. **75**, 1358 (1949).
[16] R. Batchelor and B. H. Flowers, A.E.R.E. N/R, 370 (1949).
[17] A. J. Deruytter and P. Pelfer, J. Nucl. Energy **21**, 883 (1967).
[18] C. Wagemans, P. Schillebeeckx, and J. P. Bocquet, Nucl. Sci. Eng. **101**, 293 (1989).
[19] J. Orear, Am. J. Phys. **50**, 912 (1982).
[20] M. Lybanon, Am. J. Phys. **52**, 276 (1984).