話題・解説(II)

Status of (n,xn) cross section measurements at Bruyères le Châtel: a quite special nuclear data paper

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Last year was issued a quite special nuclear data paper of a great value for everyone interested in nuclear reactions induced by fast neutrons. The paper I am talking about is the "CEA-Report" by Joël Fréhaut entitled "Status of (n,xn) cross section measurements at Bruyères le Châtel" [1].

Although this report appears more than 35 years after the experiments were completed, it is still signed by the original investigator, Joël Fréhaut, a retiree for about 15 years from the nuclear physics division of the Bruyères le Châtel research center (today often called CEA DAM DIF). It provides a revision of the (n,2n) and (n,3n) cross sections measured by Fréhaut between 1973 and 1982 at Bruyères le Châtel. In total, about 400 data points are given for a wide variety of targets, separated isotopes or natural elements, namely: ^{2}H, ^{13}C, ^{63}Cu, ^{65}Cu, ^{76}Se, ^{78}Se, ^{80}Se, ^{82}Se, ^{103}Rh, ^{142}Nd, ^{144}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd, ^{148}Sm, ^{150}Sm, ^{152}Sm, ^{154}Sm, ^{151}Eu, ^{93}Nb, ^{151}Eu, ^{155}Gd, ^{156}Gd, ^{157}Gd, ^{158}Gd, ^{160}Gd, ^{169}Tm, ^{182}W, ^{183}W, ^{184}W, ^{186}W, ^{203}Tl, ^{205}Tl, ^{206}Pb, ^{207}Pb, ^{208}Pb, ^{235}U, ^{238}U, natural Ti, V, Cr, Fe, Cu, Ga, Zr, Mo, W, Pt, Pb, and Ce and a range of mono-energetic incident neutron energies.

These measurements constitute still today the most extensive set of experimental (n,2n) energy-differential cross sections and for some nuclei they even are the unique source of experimental data. Part of these measurements had been already reported in the 1980's [2] but since the (n,xn) cross sections were measured relative to the neutron-induced fission cross section of ^{238}U they had to be updated by taking into account the current standard for this cross section which significantly differs from that of the 1970's used originally. Also, the statistics of the ^{238}U(n,2n) and ^{238}U(n,3n) updated cross sections have been significantly increased and the data for ^{13}C and Cu isotopes are reported for the first time. Moreover, this new report provides detailed information on the analysis method and on corrections that were applied to raw data in order to take properly into account the detection efficiency.

Before going deeper into the substance of those measurements, one precision, this CEA-R report is written in Molière's and Victor Hugo's language, that is to say French, as it is Fréhaut's mother tongue. However, nowadays, language is not a big problem anymore for the worldwide readers since fair quality online translation tools are freely available. An English translation of the introduction in Ref. [1] is given in Appendix.

The experimental method used to perform those (n,xn) measurements was the detection of the reaction neutrons thanks to a high volume gadolinium-loaded liquid scintillator-detector. It was a spherical tank of 76 cm diameter, the efficiency of which was quite high, about 75% in average for fission neutrons emitted in the spontaneous fission of ^{252}Cf. The 7 MV Tandem Van de Graaff in Bruyères le Châtel was used to accelerate in pulsed mode deuterons onto a gaseous deuteron target in order to induce the ^{2}H(d,n)^{3}He reaction which is well known for producing quasi mono-energetic neutrons in a given angle with respect to the incident deuteron beam. Downstream from the deuteron target, a collimation system was used to create a quasi mono-energetic neutron beam used to irradiate the target with the isotope to be studied, located in the middle of the large scintillator-detector. The latter was able to detect with a high efficiency the neutrons produced by the induced nuclear reactions. As usual when dealing with a detector, the detection efficiency had to be taken into account. This efficiency depends on the energy of the detected neutrons. Detection efficiency as a function of neutron energy was determined through calculations and also measured for ^{252}Cf fission neutrons using a ^{252}Cf source. Experimental checks with this source were performed all along the experiments. In order to apply the necessary correction for the (n,xn) neutrons, theoretical assumptions on the (n,xn) neutron energy spectra based on the statistical model and even taking into account a preequilibrium component were used.

The reader could ask: "Why on earth is this report written now, 35 years after the last measurements were performed?" Well, there was a need within the nuclear data community for a correct update of these data and a complete, detailed publication. (n,xn) cross sections are indeed important observables for the applications of fast neutron physics. Moreover, neutron data evaluators are eager to get precise constraints in every reaction channel. It is true that some of those data were already available from the EXFOR database, extracted from the original publications when they existed or sometimes directly sent by Joël decades ago. But people did not always know how to correct properly for the normalization to the ^{238}U(n,f) cross section. Also, some neutron data evaluators wanted more information on the physical hypotheses on the (n,xn) neutron energy that were followed to apply the detection efficiency correction.

After several invitations to perform the job from the nuclear physics division in Bruyères le Châtel, Fréhaut surrendered and took the road again to the lab like in the old days. To be honest the road was not too long. Indeed, Joël lives really nearby, within the administrative limits of the town of Bruyères le Châtel. He even endorsed, some years ago, the role of mayor of this town for a few years.

Let's speak a little bit of Joël Fréhaut as a nuclear physicist. Joël joined the Bruyères nuclear physics division in the 70's. That was André Michaudon at that time division head, who proposed him to move from Saclay (another CEA center mostly dedicated to nuclear power and basic research) to "B-III". B-III was the code name of the Bruyères le Châtel Center at its creation in the 50's and people were used to use this nickname for a long time. Michaudon was the instigator of the opening of the nuclear physics division, making eventually its premises open to visitors unlike the rest of BIII well protected by high electric fences. At the beginning Fréhaut worked with Soleilhac who was developing a gadolinium loaded detector dedicated to prompt fission neutron measurements, in particular the mean neutron multiplicity also known as Nu-bar. That was the beginning of a big story. Soon, after Soleilhac retired, Fréhaut became the European pope of such measurements. Even today his Nu-bar measurements for the major actinides for incident neutrons from 1 MeV up to 28 MeV are the most extensive and precise.

But let's go back to Fréhaut's 2016 come back to B-III which ended up in the report we are talking about. There, with the technical help of his old colleague and friend Gérard Haouat, another brilliant retired experimental nuclear physicist and of younger colleagues, Fréhaut could update his good old data and write this excellent and very complete report. Now the (n,xn) results are gathered in this report including the results never published before. They have been updated according to the state of the art knowledge of the ^{238}U(n,f) cross section. Detailed information is provided on the analysis and the theoretical assumptions made. These are very reasonable but thanks to the information provided, everyone can use their own theoretical model and update the results with an efficiency correction modified accordingly even though the effect is expected to be small.

Well, this is certainly an excellent thing that good old data can now be exploited at their best by nuclear data evaluators and others. Good old data freshly refurbished!

Another excellent news is that Bruyères le Châtel's Tandem Van de Graaff accelerator which had been shut down for about 10 years has been recently refurbished and improved and is now being used again to perform reaction cross section measurements. Let's hope new exciting data will go out soon!

References

[1] Joël Fréhaut, Le point sur les sections efficaces de réaction (n,xn) mesurées à Bruyères-le-Châtel, CEA-R-6447 Report, november 2016, CEA.

[2] J. Fréhaut, A. Bertin, R. Bois, J. Jary, Status of (n,2n) cross section measurements at Bruyères-le-Châtel, Symposium on neutron cross sections from 10 – 15 MeV, Upton, L. I. (USA), May 1980, BNL-NCS-51245, Ed. M. R. Bhat and S. Pearlstein, Vol. I, P. 399.

Appendix English translation of the introduction of Joël Fréhaut's report [1]

INTRODUCTION

(n,xn) cross-section measurements were performed over 10 years (1973 to 1982). The experimental method, based on detection of the reaction neutron, was published in 1976 [A1].
That 1976 publication also presents the various corrections applied to the measurements to obtain the cross sections. Among them, the correction for the neutron detection efficiency, which takes into account the difference between the energy spectra of the (n,xn) reaction neutrons and that of the neutrons from the spontaneous fission of the ^{252}Cf source used to calibrate the detection efficiency, was evoked only qualitatively.

This correction, which can exceed 10%, is based on the use of the nuclear evaporation model for the calculation of the energy spectra of the (n,xn) neutrons, which made it possible to implement relatively simply the calculation of the corrective factors for the about 400 experimental data points, especially with regard to the rather limited means of calculation at that time. The correction method is presented below in detail in order to make it possible to change this correction if required.

We were then aware that the evaporation spectra were perhaps not sufficient, and that pre-equilibrium had also to be taken into account though we did not have a suitable local code for its calculation. However, the participation of Mike Cates of LANL in our measurements on thallium (Tl) in 1977 allowed us to obtain some calculations of neutron spectra with the GNASH code [A2]. If the energy spectra are different with and without the pre-equilibrium process, it can be seen that the detection efficiency correction was about the same, because of a compensation effect between the first neutron and the second neutron.

Most of our results appear in the proceedings of the "Symposium on neutron cross sections from 10 - 15 MeV" held in Upton in 1980 [A3]. These are the same data that make the EXFOR entry 20416.

These published data are normalized to the ^{238}U fission cross-section resulting from the now outdated evaluation done by Sowerby [A4], and different users did not always take care to perform the renormalization on a more recent evaluation. This renormalization will be mentioned in the last part of the present report and updated cross sections will be given for all the measured nuclei.

References

[A1] J. FRÉHAUT, “Use of the large gadolinium-loaded liquid scintillator technique for (n,2n) and (n,3n) cross section measurements”, Nucl. Inst. Meth. 135 (1976) 511.

[A2] P. G. YOUNG and E. D. ARTHUR, “GNASH: A Preequilibrium, Statistical Nuclear Model Code for Calculation of Cross Sections and Emission Spectra,” Los Alamos National Laboratory report LA-6947 (1977).

[A3] J. FREHAUT, A. BERTIN, R. BOIS, J. JARY, “Status of (n2n) cross section measurements at Bruyères-le Châtel”, Symposium on neutron cross sections from 10 – 15 MeV, Upton, L. I. (USA), May 1980, BNL-NCS-51245, Ed. M. R. Bhat and S. Pearlstein, Vol. I, P. 399.

[A4] M. G. SOWERBY, B. H. PATRICK, D. S. MATHER, « A detailed report on the simultaneous evaluation of the fission cross sections of ^{235}U, ^{239}Pu and ^{238}U and the ^{238}U capture cross section in the energy range 100 eV to 20 MeV »,Rapport AERE-R-72/73, U.K. Atomic Energy Authority, Harwell,(1973).