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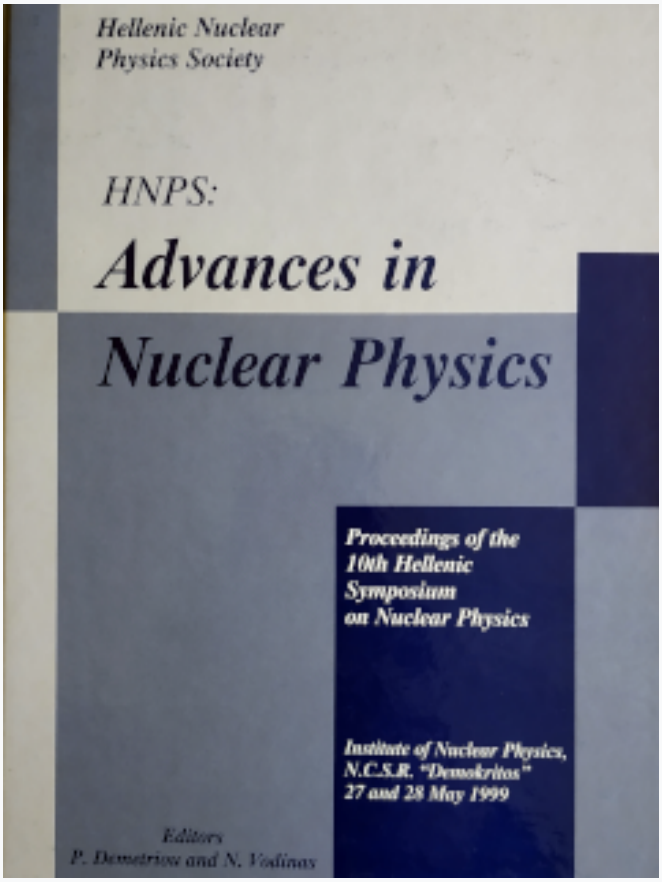

Neutron capture cross section measurements of 232Th in the 30-600 keV energy range

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Abstract

Neutron capture cross sections of ^{232}Th have been measured relative to ^{197}Au in the energy region from 30 keV to 600 keV. The neutron source was the reaction $^{7}Li(p,n)$ produced on the 4 MV Van de Graaff Accelerator of the Centre d' Etudes Nucleaires de Bordeaux (CENBG). Preliminary analysis of the measurements indicates that the cross sections are closer to the JENDL database values.

1 Introduction

One of the most relevant problems with nuclear power is the production of large amounts of highly radioactive nuclear wastes, which represent a major concern due to the huge lifetime of some of the elements. Incineration of transuranic elements and long-lived fission fragments is presently under consideration as a promising option to handle the radioactive waste problem and alternative to the deep underground geological storage with its uncertain long term consequences.

There are two incineration methods under investigation in international studies, actinide burner reactors and accelerator-based transmutation systems. Furthermore, accelerator driven systems (ADS) based on the ^{232}Th-^{233}U cycle, have been proposed as an alternative way for energy production [1,2,3]. In these systems, an intense proton beam bombarding a spallation target of a heavy material, is coupled to a sub- critical reactor core. High fluxes of neutrons are produced and are used to drive the sub-critical blanket where energy is produced and long lived nuclear waste can be burned.

A successful application of the ^{232}Th-^{233}U mixture as combustible requires that the quality of the nuclear data is of the same standard as that of the U-Pu Cycle.

However, the existing data of the nuclear databases are insufficient to enable a reliable Th cycle analysis. Among the most crucial reaction channel is the neutron capture of ^{232}Th which leads to ^{233}Pa and ^{233}U. In fast reactors, the above reaction is required with an accuracy of 2%. The situation for ADS is similar. A 10% change in the ^{232}Th capture cross section gives rise to a 30% change in the needed proton current of the accelerator if the system has to be operated at a subcritical level of $K_{eff} \simeq 0.97$ [4]. However, for this reaction, the evaluated data in the energy range 0.02-1.5 MeV differ by $10 - 30\%$ [5].

Hence, there is a great need for new measurements and evaluations. In this context, a program at the 4 MV Van de Graaff has started lately, in order to measure the neutron capture cross section of ^{232}Th in the energy interval from 30 keV to 3 MeV. In this report, preliminary results in the energy range from 30 keV to 600 keV will be presented.

2 Experimental

The neutron capture cross section of ^{232}Th was measured by the activation method relative to the standard neutron capture cross section of ^{197}Au. This method was chosen due to its simplicity and selectivity for a given nuclide and its sensitivity when combined with low background gamma ray spectroscopy. Mono-energetic neutrons were produced by bombarding water cooled LiF targets with a proton current of $15\mu A$ from the 4 MEV Van-de-Graaff (VDG) accelerator of the Centre d'Etudes Nucleaires de Bordeaux Gradignan(CENBG). The dedicated neutron line of CENBG with a "neutron hole" of 1.7 meters deep, was used. The threshold of the reaction $^{7}Li(p,n)^{7}Be$ was used for the energy calibration of the VDG accelerator. Furthermore, the factor relating the analyzer's magneting field to the proton energy, was determined by varying the magneting field and measuring the produced neutrons with a He^3 detector. Several thickness of LiF target were used and the relation $B(gauss) = 2238.89\sqrt{E_p(MeV)}$ was determined.

Thorium metal targets were of high purity (99.5%), thickness of 1 mm and surface of 1 cm^2. They were packed together with two Au foils on each side (same surface and 0.5 mm thickness) and enclosed in an Cd box of 1 mm thickness. Cadmium was used to eliminate the contribution from thermal neutrons that were produced in the experimental hall. The irradiation was performed in the 0° with respect to the proton beam with typical bombarding time of the order of 20 hours.

Prior to the irradiation, the neutron thermal background from scattering in the experimental hall, was measured using the coincidence technique of the alpha and triton produced in the $^{6}Li(n,t)a$. It was found to be negligible when the cadmium shielding was applied. During the irradiation, the neutron flux was monitored with a He^3 detector that was far away from the neutron source. In this way, irradiations were performed for neutron energies between 30 to 600 keV.

At the end of irradiation, the intensity of the gamma ray lines emitted from the de-excitation of the produced nucleus ^{198}Au (412 keV) and ^{233}Pa (312 keV), was measured separately with gamma spectroscopy. Two High Purity Germanium Detectors (HPGe) (40% and 80%) and an X-ray detector were calibrated and tested with standard electronics for various source to detector distances. The optimal conditions were achieved with the 40% HPGe which was subsequently used for all the gamma activity measurements of the gold and thorium foils. The acquisition time varied between 1-2 hours for Au and 1-2 days for Th. The pulser method was used for dead time correction. The photopeak areas were determined with the program PAW while two more fitting programs were used for comparison.

3 Analysis

For relative measurements with the Au standard in the same conditions, the ^{232}Th(n,γ) capture cross section was calculated using the formula:

$$\frac{^{Th}\sigma(n,\gamma)}{^{Au}\sigma(n,\gamma)} = \frac{\epsilon(412)}{\epsilon(312)} \cdot \frac{^{Th}I(312)}{^{Au}I(412)} \cdot \frac{^{Th}N}{^{Au}N} \cdot \frac{f(T^{Th},t)}{f(T^{Au},t)} \quad (1)$$

where I and ϵ are the photopeak intensity and efficiency for the respective γ lines, N the number of atoms and f the time factor relating the measured activities to the end of irradiation. The capture cross sections of ^{197}Au were taken from the ENDF/B-VI library.

It is evident from Eq. (1) that the knowledge of the efficiencies of the two gamma ray lines is of primary importance in the correct determination of the cross section. Therefore, the self absorption factor that influence the efficiencies, was further investigated. In a first approximation, the analytical expressions

$$F_{self-abs} = 1/(1 - a_s) \quad (2)$$

and

$$a_s = 1 - (1 - exp(-\mu \cdot x))/\mu \cdot x \quad (3)$$

were used. In the above formulas μ is the attenuation coefficient for the lines of Au or Th and x the thickness of the sample. The values of 9% and 21% were determined for Au and Th respectively. Furthermore, the Haase method [6] with total efficiency determination and normalization with a calibrated punctual source, was also applied. The efficiency of the 40% HPGe detector was determined with a ^{152}Eu punctual source and several fitting equations. The best results were achieved using the equation [7]:

$$\epsilon(E) = A \cdot ln(E) + B \cdot \frac{ln(E)}{E} + C \cdot \frac{ln(E)^2}{E} + D \cdot \frac{ln(E)^4}{E} + F \cdot \frac{ln(E)^5}{E} \quad (4)$$

The self-absorption values determined with the Haase method were $6.4 \pm 0.4\%$ for Au and $19.9 \pm 0.5\%$ for Th.

Since there was a discrepancy between the determined values, the two well known MCNP4B and GEANT 3.21 Monte Carlo programs were further used. In this way, a very detailed geometrical detector description was produced by comparison with the experimental efficiency. The agreement achieved was better than 95%. Therefore, the self absorption was determined as $21.0 \pm 0.4\%$ for the 312 keV Th line and $6.1 \pm 0.1\%$ for the 412 keV Au line.

In the values of the neutron radiative capture cross section of ^{232}Th calculated with Eq. (1), different corrections had to be applied due to:

- Neutron energy spread because of proton energy degradation in the source
- Effects of finite dimensions of neutron source and targets on neutron energy spread
- Inelastic and elastic neutron scattering within the intermediate experimental environment
- Multiple elastic and inelastic scattering in the target foils

Therefore, a Monte Carlo code considering all the above effects, is currently under development for an IBM AIX 4.3.2 operating system. The only assumption of the code is that the angular distribution of the produced neutrons is constant in their energy spread interval. This constrain is imposed because of the lack of experimental data for several neutron energies. The architecture of the code is schematically depicted in Diagram 1.

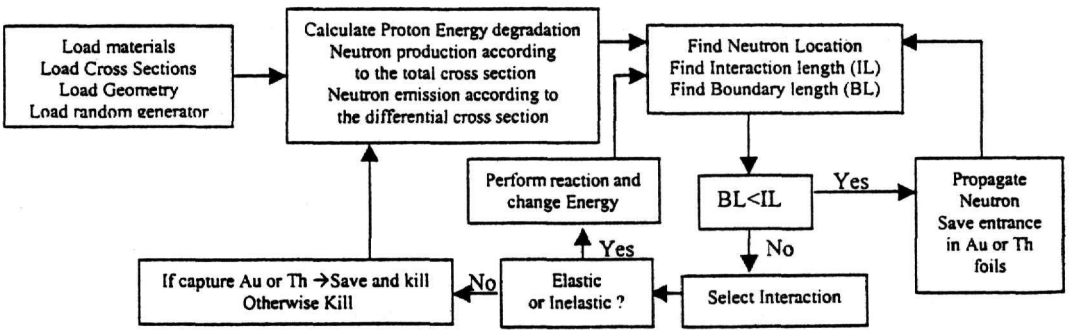

Diagram 1. Flow diagram of the Monte Carlo code for the determination of the correction factors in the neutron captyre cross section of ^{232}Th.

A first version of the code was applied in all the experimental points and the correction factors of the capture cross section of ^{232}Th were determined. With the implication of the above corrections, the values of the neutron radiative cross section of ^{232}Th are given in Fig. 1 with the associated sources of errors. In the same figure the evaluations, of the different databases are also included.

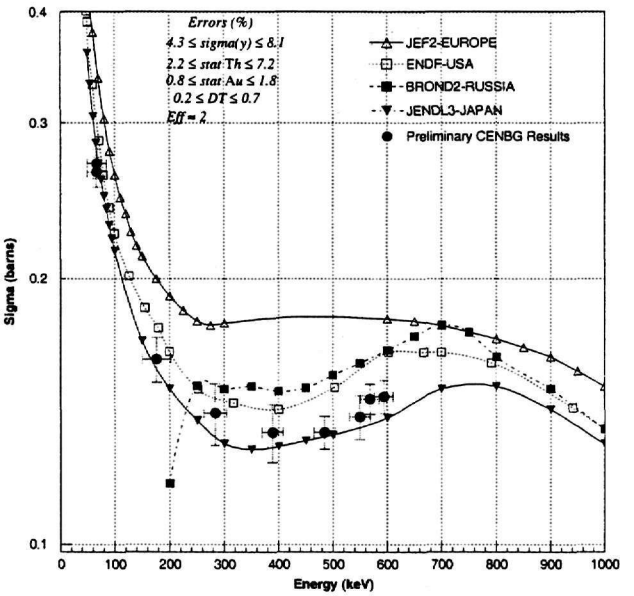

Fig. 1. Neutron radiative capture cross section of ^{232}Th and the existing evaluated data from the four major neutron data reference libraries.

4 Conclusions

It is evident from Fig. 1, that the data are closer to the JENDL evaluation. However, the results presented in this work are preliminary and further analysis is needed. The extension also to higher energies has been started. Complementary to the already used Au standard, the neutron fission of ^{235}U will be used as a second standard. The fission fragments will be detected on line by the use of a fission detector while the off line gamma activity measurements of the Au and Th foil will be performed with the already used methods.

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References

[1] C.D. Bowman et al. NIM A320 (1992) and F. Venneri et al. LA-UR-93-752

[2] T. Takizuka, Proceedings of the Int. Conf. on Accelerator-Driven Transmutation

Technologies and Applications, Las Vegas, NV, USA, August 1994.

[3] C. Rubbia et al. CERN/AT/93-47 (1993) and CERN/AT/95-44 (ET) (1995).

[4] M. Salvatores, Conf. Proceedings Vol. 59, "Nuclear data for Science and Technology", G. Reffo, A. Ventura and C. Grandi (Eds.) Bologna 1997.

[5] D. Karamanis, CENBG/Draft Note/Mars 1999.

[6] G. Haase et al. NIM A 336 (1993) 206.

[7] K. Debertin NIM 226 (1984) 566.