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Theoretical calculations for the reaction $^{241}Am(n,2n)$ in the framework of the Hauser-Feshbach model.

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Abstract

The cross section of the reaction $^{241}Am(n,2n)$, has been measured by the activation method in the range from 9.6 to 11.4 MeV, at the Tandem accelerator facility of NCSR Demokritos. Statistical model calculations in the framework of the Hauser-Feshbach theory have been performed, and the first results are presented. Experimental data on the neutron induced fission cross section of ^{241}Am, have been used as a constraint for the calculations. The results of the investigation are presented in comparison with experimental data and previous theoretical evaluations.

1 Introduction

The world energy consumption growth, is one of the main social problems that will have to be addressed successfully in the years to come. The amount of energy produced from fossil fuel and alternative energy sources—e.g. solar wind and hydroelectric energy—will be unable to meet the consumption needs, in some years time. Hence, nuclear energy is arising as an appealing solution, provided the drawbacks related to the environmental impact of its use, will be addressed succesfully.

Many solutions have been proposed for these problems, the most promising of which, are related to the so called Accelerator Driven Systems (ADS) [1]. The ADS, is a hybrid subcritical system, consisting of the combination of a high energy accelerator with a reactor. Neutrons produced at an accelerator through a spallation reaction, will be used for energy production in the core of the reactor, and for transmutation or/and incineration of radiotoxic nuclear waste, through neutron induced fission. One key element for the success of

such projects, is whether a fast or a thermal neutron spectrum is more adequate for the transmutation of actinides, which are contained in large amounts in spent nuclear fuel. According to recent calculations [2], in a fast neutron spectrum, all the actinide nuclei, could be transmutated effectively, producing at the same time, energy through fission. This is not the case, however, for a thermal spectrum, were some of the actinide nuclei would not fission at all. The characteristics of the neutron spectrum produced by the accelerator, depend mainly on the construction material of the spallation target. A typical upper threshold for the energy of neutrons produced in an ADS system using a Lead target, is of the order of 1 GeV. In such a neutron spectrum, the neutron balance in the core of the reactor would be affected by reactions of the (n,xn) and (n,xnf) type, with the actinide nuclei. In order to determine the contribution of these reactions to the neutron flux of the core, the corresponding cross sections have to be determined, for nuclei with $90 \leq Z \leq 96$, up to energies of the order of 1 GeV. These measurements are in general, quite difficult. This explains in a way, the lack of experimental data on the n,xn and n,xnf reactions of actinides. Due to this lack of data, the design of ADS systems has to rely to a large extent, on theoretical calculations. However, the consistency of theoretical calculations has to be verified, experimentally. In the context of this idea, in the present work, the cross section for the reaction $^{241}Am(n,2n)^{240}Am$ has been investigated experimentally and theoretically.

2   Summary of Experimental data

For ^{241}Am, one of the most abundant nuclei in nuclear waste, and one of the most radiotoxical ones, the existing data are summarized in table 1. For the $^{241}Am(n,2n)$ reaction, the existing experimental data, are limited to a narrow energy region from 13.4 up to 15MeV. In the present investigation,

Reaction	Energy Range	Comments
$^{241}Am(n,\gamma)$	up to 500 keV	important for low energies
$^{241}Am(n,2n)$	13.4 to 15.1 MeV	limited energy range
$^{241}Am(n,total)$	up to 18 MeV	
$^{241}Am(n,f)$	up to 20 MeV	individual channels admixture above 6 MeV

Table 1
Summary of existing measurements concerning ^{241}Am neutron induced reactions

the cross section of the reaction $^{241}Am(n,2n)$, has been measured by the activation method at the Tandem accelerator of NCSR Demokritos, in the energy range from 9.7 to 17MeV. A 37GBq Americium source encapsulated in a lead shielding, served as the activation target for the 5-day long irradiation of each run.

3   Theoretical Description

The nucleus ^{241}Am, absorbs a neutron to form the compound nucleus ^{242}Am. The compound system deexcites, mainly by sequential emission of neutrons to ^{240}Am, and through fission of all the compound nuclei involved. In the framework of the statistical model of Hauser and Feshbach, the cross section for each exit channel of the compound system, is given in the most general case, by equation 1.

$$\sigma_b(E, J, \pi) = \sigma_a(E, J, \pi) \frac{\Gamma_b(E, J, \pi)}{\sum_c \Gamma_c(E, J, \pi)} \qquad (1)$$

Thus,it depends on the cross section for the formation of the compound nucleus, σ_α, and also on the widths (probability) for fission or for the evaporation of a particle or a gamma ray (corresponding to Γ_b in equation 1). The widths corresponding to the main exit channels in our case (that is, for fission and neutron emission), are given by the following equations respectively:

$$\Gamma_n(E, J, \pi) = \frac{1}{2\pi\rho_{CN}(E, J, \pi)} \cdot$$
$$\cdot \sum_{J'=0}^{\infty} \sum_{\pi'} \sum_{j=J'-J}^{J+J'} \int_0^{E-B_n} \rho_n(E', J', \pi') \cdot T_n^{ij}(E - B_n - E^J) dE' \quad (2)$$
$$\Gamma_f(E, J, \pi) = \frac{1}{2\pi\rho_{CN}(E, J, \pi)} \cdot$$
$$\cdot \int_0^{E-E_{sad}(J)} \rho_f(\epsilon, J, \pi) T_f(E - E_{sad}(J) - \epsilon) d\epsilon \qquad (3)$$

The widths depend primarily on the corresponding transmission coefficients and level densities, which are the most critical quantities entering the theoretical description of any reaction.Since the fission process is the dominant mechanism of deexcitation in this energy and mass region, in order to describe theoretically the $(n, 2n)$ reaction, it is absolutely necessary, to consistently describe the fission process as well. The fission process for an actinide nucleus, is described in the calculation, as a sequential crossing of the two humps of a double humped fission barrier, according to figure 1. The humps, correspond to transition states related to the local maxima of the shell correction for the deformation energy of the nucleus. A nucleus that will get so deformed that will cross the two humps and reach the scission point, will fission immediatelly. The probability for fission to happen, is given by the transmission coefficient for penetrating this barrier. The transmission coefficient for each one of the humps is related to the density ρ of the transition states at the corresponding
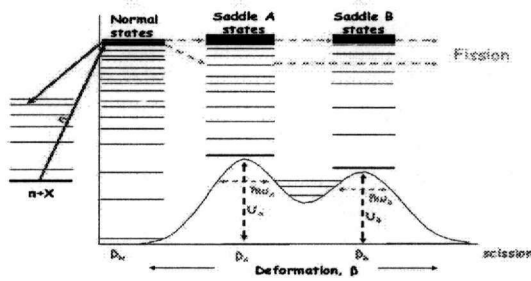
Fig. 1. *Outline of the fission process in the context of the double humped fission barrier model*

deformation, the height of the barrier U, and its curvature $\hbar\omega$, according to equation 4.

$$T_{A,B}^{J,\pi}(U') = \int_0^U [1 + \exp(\frac{2\pi}{\hbar\omega_{A,B}}(U_{A,B} - U' + \epsilon)]^{-1} \cdot \rho_{A,B}(\epsilon, J, \pi) \cdot d\epsilon \qquad (4)$$

For the theoretical calculations of the reaction $^{241}Am(n, 2n)$, the transmission coefficients for the input channel, were deduced using a coupled channels optical potential originally developed for ^{238}U that presents a weak isotopic dependence [3], and has been previously proven succesful in describing the fission cross sections of Americium and other isotopes of the actinide family [?]. The height of the barriers used in the calculation, were adopted from [6]. The nuclear level densities at saddle and at normal deformation, are determined in the framework of the Generalized Superfluid Model of the Nucleus [4], which is a phenomenological model, that takes into account, shell, collective and superfluid effects. The implementation of such effects in the level density formulation, is of utmost importance for the consistent description of reactions with heavy deformed nuclei like the actinides, that undergo fission. Collective effects, are included in the calculation, in the form of the multiplying factors K_{rot} and K_{vib} in the following formulas used for the level density:

$$\rho(U', J) \propto (2J + 1) \cdot \rho(U') \cdot K_{vibr}(U')K_{rot}(U', J) \cdot \exp(-\frac{J(J+1)}{2\sigma^2}) \qquad (5)$$

$$\rho(U') \propto \frac{\exp(2\sqrt{\alpha U})}{\sqrt{2\pi\sigma^2}} \qquad (6)$$

$$\alpha = \tilde{\alpha}[1 + \delta w \cdot f(U - E_{cond})] \qquad (7)$$

The parametre K_{rot} is related to the shape of the nucleus according to the adiabatic aproximation [5], while the parametre K_{vib} is based on a liquid drop estimate of the density of multipole oscillations of the nuclear surface [4].

From the parametres used for the calculation of the nuclear level densities, the asymptotic parametre $\tilde{\alpha}$, for nuclei with A=241 and A=242, were based on fitting of resonances of the reaction $^{241}Am(n,\gamma)$ at the resolved resonance region, while for the other nuclei, were selected in a way that the $n, 2n$ and n, f_{obs} reactions could be fitted reasonably.

4   Results

Using the set of parametres described, a satisfactory fitting of the reactions $^{241}Am(n, 2n)$ and $^{241}Am(n, f_{obs})$ could be achieved. The comparison between calculation and experiment is illustrated on figures 2,3. The calculated values for the fission cross section are in fair agreement with the experimental data at the whole energy region. At the same time, the same calculation, reproduces in a reasonable fashion, the data for the $(n, 2n)$ reaction. The agreement in
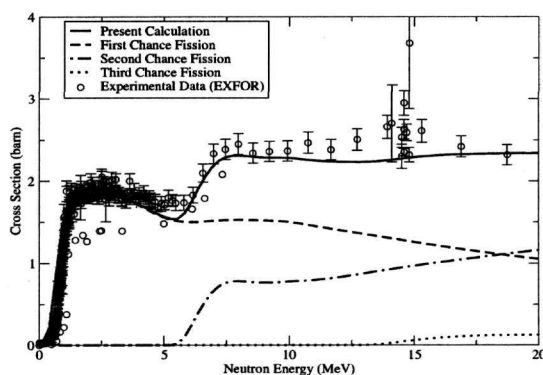

Fig. 2. Comparison of Experimental and Calculated Data for the reaction $^{241}Am(n, f)$

the case of the $(n, 2n)$ reaction, is better in the higher energy region, above 10MeV. At lower energies, further experimental and theoretical investigation is needed. The apparent bump of the experimental data for the (n, f) reaction at the energy region above 12MeV, can not be reproduced by this calculation. The possible cause for this is the adopted optical model potential. Further investigation of the parametres related to the optical potential, could improve the agreement. The behavior of the reaction $^{241}Am(n, 2n)$, near the threshold, could also provide additional information connected to the description of the level density at lower excitations, near the threshold of the (n, nf) reaction channel.
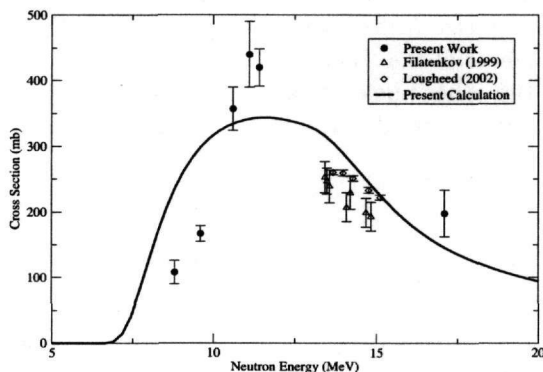
Fig. 3. Comparison of Experimental and Calculated Data for the reaction $^{241}Am(n,2n)$

5   Conclusion

The cross section of the reaction $^{241}Am(n,2n)^{240}Am$, has been investigated for the first time experimentally and theoretically, in a wide energy range, from 9.7 up to 17MeV. The preliminary results of the theoretical calculation are within reasonable agreement with the experimental data presented in this work, as well as with previous measurements. The agreement is better at higher energies than at energies closer to the threshold of the reaction. For the future, it is planned to continue the investigation concerning the $^{241}Am(n,2n)$ reaction near the threshold, as well as, at higher energies, to obtain a consistent description, at the whole energy range from threshold to 20MeV.

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