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Neutron-induced cross sections of actinides via the surrogate-reaction method

Q. Ducasse[1,2,a], B. Jurado[1,b], M. Aiche[1], T. Tornyi[3], A. Görgen[3], J.N. Wilson[4], G. Barreau[1], I. Companis[1], S. Czajkowski[1], F. Giacoppo[3], F. Gunsing[5], M. Guttormsen[3], A.C. Larsen[3], M. Lebois[4], J. Matarranz[1], T. Renstrøm[3], S. Rose[3], S. Siem[3], I. Tsekhanovich[1], G.M. Tveten[3], T.W. Hagen[3], M. Wiedeking[6], O. Serot[2], G. Boutoux[7], P. Chau Huu-Tai[7], V. Méot[7], and O. Roig[7]

[1] CENBG, Chemin du Solarium, BP. 120, 33175 Gradignan, France
[2] CEA-Cadarache, DEN/DER/SPRC/LEPh, 13108 Saint Paul lez Durance, France
[3] University of Oslo, Department of Physics, PO Box 1048, Blindem 0316 Oslo, Norway
[4] IPN Orsay, 15 rue G. Clémenceau, 91191 Gif-sur-Yvette Cedex, France
[5] CEA Saclay, DSM/Irfu, 91191 Gif-sur-Yvette Cedex, France
[6] iThemba LABS, PO Box 722, 7129 Somerset West, South Africa
[7] CEA, DAM, DIF, 91297 Arpajon, France

Abstract. The surrogate-reaction method is an indirect way of determining cross sections for reactions that proceed through a compound nucleus. This technique may enable neutron-induced cross sections to be extracted for short-lived nuclei that otherwise cannot be measured. However, the validity of the surrogate method has to be investigated. In particular, the absence of a compound nucleus formation and the J^{π} dependence of the decay probabilities may question the method. In this work we study the reactions ^{238}U(d,p)^{239}U, ^{238}U(^{3}He,t)^{238}Np, ^{238}U(^{3}He,^{4}He)^{237}U as surrogates for neutron-induced reactions on ^{238}U, ^{237}Np and ^{236}U, respectively, for which good quality data exist. The experimental set-up enabled the measurement of fission and gamma-decay probabilities. The first results are hereby presented.

1. Introduction

Neutron-induced cross sections of short-lived nuclei are crucial for fundamental nuclear physics, astrophysics and also for applications such as reactor physics. In particular, (n,γ) cross sections on minor actinides are one of the largest sources of uncertainty in modelling new reactors for nuclear waste transmutation using fast neutrons. However, very often the high radioactivity of the actinide samples makes the direct measurement of these cross sections extremely difficult. The surrogate-reaction method is an indirect way of determining cross sections for compound-nuclear reactions. This method was first

[a] e-mail: ducasse@cenbg.in2p3.fr
[b] e-mail: jurado@cenbg.in2p3.fr

proposed by J. D. Cramer and H. C. Britt [1] in the seventies. It consists of using a transfer reaction to produce the same decaying nucleus as the one formed in the desired neutron-induced reaction. The transfer reaction leads to the heavy recoil nucleus of interest and to an ejectile. The identification of the ejectile permits to determine the mass A and charge Z of the decaying nucleus. In addition, one can deduce the excitation energy E^* of the heavy nucleus by measuring the kinetic energy and the emission angle of the ejectile. The measurement of the number of coincidences between the ejectiles and the decay products normalized to the total number of detected ejectiles allows one to extract the decay probability for the corresponding decay channel $P^{A,exp}_{decay}(E^*)$. According to the surrogate-reaction method, the neutron-induced cross section for the nucleus $A-1$ is then given by the equation:

$$\sigma^{A-1}_{decay}(E_n) \cong \sigma^{A}_{CN}(E_n) \cdot P^{A,exp}_{decay}(E^*), \tag{1}$$

where $\sigma^{A}_{CN}(E_n)$ is the calculated cross section for the formation of the compound nucleus A after absorption of a neutron with energy E_n; this quantity is usually obtained using optical model. In our case, we use the phenomenological optical model from TALYS for the ^{238}U + n reaction and the ECIS code for the ^{237}Np + n and ^{236}U + n reactions. The incident neutron energy E_n and the excitation energy E^* of the compound nucleus A are related by the expression $E^* = S_n + E_n \cdot (A-1)/A$, where S_n is the one-neutron separation energy in the nucleus A. The benefit of the surrogate method is that in some cases the target needed is stable or less radioactive than the target of the corresponding neutron-induced reaction. For the surrogate method to work, the decaying nucleus has to be a compound nucleus. In addition, one has to consider the spin-parity differences between the neutron-induced and the surrogate reactions.

Indeed, at low excitation energies the decay probability strongly depends on J^π. Therefore, important deviations between the neutron-induced results and the ones obtained with the surrogate method may exist if the populated spin distribution in the neutron-induced and surrogate experiments are different. While it is rather well established that the surrogate method works well for fission at sufficiently high E^* (see e.g. [2]), several recent experiments have shown that gamma decay is very sensitive to the differences in the populated J^π distributions ([3–5]), which leads to important discrepancies between the surrogate results and the neutron-induced data at low excitation energies. This is probably due to the spin-parity selectivity of neutron emission [5]. This selectivity decreases strongly as the level density of the residual nucleus after neutron emission increases. Therefore, the discrepancies between the surrogate results and the neutron-induced data are expected to decrease with increasing mass of the decaying nucleus and with increasing excitation energy. In this work we study the validity of the surrogate method in the actinide region using an improved experimental set-up that enables the measurement of fission and gamma-decay probabilities. Thanks to this we can investigate the two main issues that determine the validity of the surrogate method: the "compound" character of the decaying nucleus and the J^π dependence of the decay probabilities.

2. Experiment

The experiment was performed in June 2012 at the Oslo cyclotron. We used a ^{238}U target with 99.7% isotopic purity produced at GSI on a ^{12}C backing. Two different beams were used, a deuteron beam of 15 MeV and a ^{3}He beam of 24 MeV. The experimental set-up is represented in Figure 1. The ejectiles were detected at backward angles with the SiRi multi-strip silicon ΔE/E detector [6]. SiRi provided the identification of the ejectiles, as well as their kinetic energy and angle. Fission fragments were detected in coincidence with the ejectiles. The fission detector was located at forward angles and consisted of 4 PPACs covering a solid angle of 41.1% out of 2π. The reaction chamber housing SiRi, the PPACs and the ^{238}U target were surrounded by the CACTUS array made of 27 high-efficiency NaI detectors. CACTUS was used to detect gamma rays with energies ranging from several hundreds keV to about 10 MeV in coincidence with the ejectiles. The aim of the experiment was to study the transfer reactions

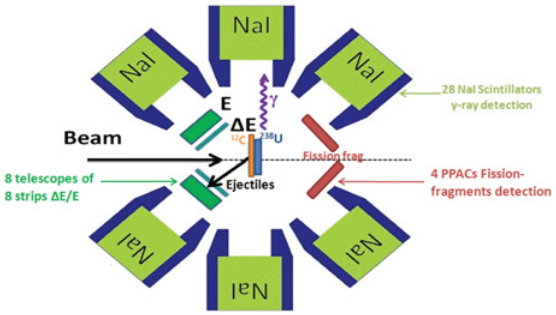

Figure 1. Schematic view of the set-up for decay-probability measurements with the SiRi telescope and the fission detector surrounded by the CACTUS NaI array.

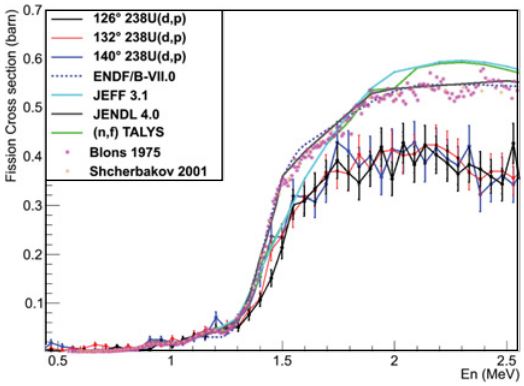

Figure 2. Preliminary fission cross section for three angles obtained with the $^{238}U(d,\,p)$ reaction compared with experimental neutron-induced data and several evaluations.

^{238}U(d,p), ^{238}U(^{3}He, t) and ^{238}U(^{3}He,^{4} He) serving as surrogates for the neutron-induced reactions ^{238}U + n, ^{237}Np + n and ^{236}U + n, respectively.

Fission or gamma-decay probabilities are obtained following the expression:

$$P^{A,exp}_{decay}(E^*) = \frac{N_{coinc}(E^*)}{N_{singles}(E^*) \cdot \epsilon_{decay}(E^*)} \tag{2}$$

where N_{coinc} is the number of coincidences between the ejectiles and the fission or the γ-ray detector, $N_{singles}$ is the total number of ejectiles and ϵ_{decay} is the efficiency of the fission or the γ-ray detector.

3. Preliminary results

3.1 The ^{238}U(d,p)^{239}U reaction

The results presented in this subsection follow from the analysis of data for three telescope strips out of 64. The selected strips were located at 126, 132 and 140 degrees relative to the beam direction. Because of the relatively low statistics, these results should be considered as preliminary.

Figure 2 shows fission cross section of $^{239}U^*$ as a function of neutron equivalent energy obtained from the ^{238}U(d,p) reaction. Our data are compared with different evaluations and experimental neutron-induced data. The geometrical fission efficiency has been measured by using a ^{252}Cf source of known activity. The effective fission efficiency has to include the effects due to the angular anisotropy of the

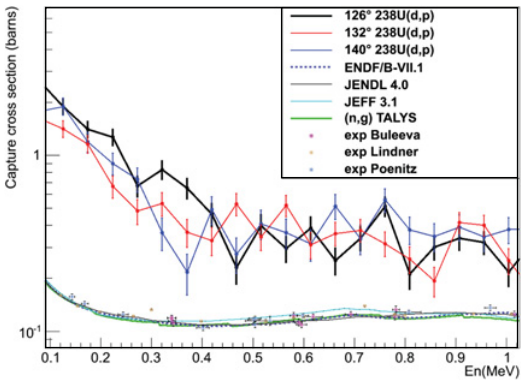

Figure 3. Preliminary capture cross section for three angles obtained with the $^{238}U(d, p)$ reaction compared with experimental neutron-induced data, TALYS(n,gamma) calculation and several evaluations.

fission fragments. The angular anisotropy has been determined as a function of E^* for the ^{238}U(d,p) reaction by Britt and Cramer [1]. We have included these results in a Monte-Carlo simulation that reproduces the experimental solid angle of our fission detector. Our simulation leads to a final fission efficiency of 45.5% (±3.7%) for the range of equivalent neutron energy considered in this work. Figure 2 shows that the fission cross section does not depend on the detection angle of the proton, although there are some differences between the two curves in the 1.3–1.5 MeV energy range that may be the consequence of an angular-dependent spin distribution. However, the statistics of the data is still too low to confirm this. Up to 1.7 MeV, the surrogate results are in good agreement with the neutron data. However, above 1.7 MeV the results from the surrogate reaction are clearly below the neutron-induced results. The difference amounts to 35% at most. The reason for this discrepancy could be that the neutron that is transfered in the stripping (d,p) reaction to the ^{238}U target nucleus escapes into the breakup phase space before the nucleus $^{239}U^*$ becomes a compound nucleus. This leads to a background of "sterile" protons that contaminates the singles proton spectrum. These protons are not correlated with the compound-nucleus formation decreasing the fission probability as shown by Eq. (2). This phenomenon was already observed by Britt and Cramer [7] but only now it starts to attract theoretical efforts [8]. Another process that has to be taken into consideration is the fusion of the deuteron beam with oxygen and the subsequent evaporation of protons. Again, this leads to the production of sterile protons decreasing the fission probability. The oxidation of the target can not be complety avoided, this is why this process could explain in a minor way the encountered differences. To address this issue fusion-evaporation calculations will shortly be performed with the PACE4 code.

Figure 3 represents the result for the capture cross section of ^{238}U as a function of equivalent neutron energy obtained from the ^{238}U(d,p) reaction for three angles in comparison with neutron-induced data and several evaluations. As can be seen from Figure 2, fission sets in at equivalent neutron energy of about 0.9 MeV. Therefore, up to this energy, the capture cross section of ^{238}U is not affected by the gamma emission of the fission fragments. The gamma efficiency of the experimental set-up has been determined by using the EXEM method [9]. After neutron emission of $^{239}U^*$, the residual nucleus $^{238}U^*$ may still emit gamma rays. We have removed these gammas by applying a threshold to the NaI energy spectra as discussed in [5].

The capture cross section obtained with the surrogate method is several times higher than the neutron-induced one, in the whole excitation-energy range. The agreement is better at higher excitation energy (a minimum factor of about 2.5 is reached at 1 MeV). The reason for these discrepancies is probably that the angular momentum induced by the (d,p) reaction is larger than the one populated in the neutron-induced reaction. Under these conditions neutron emission becomes very improbable

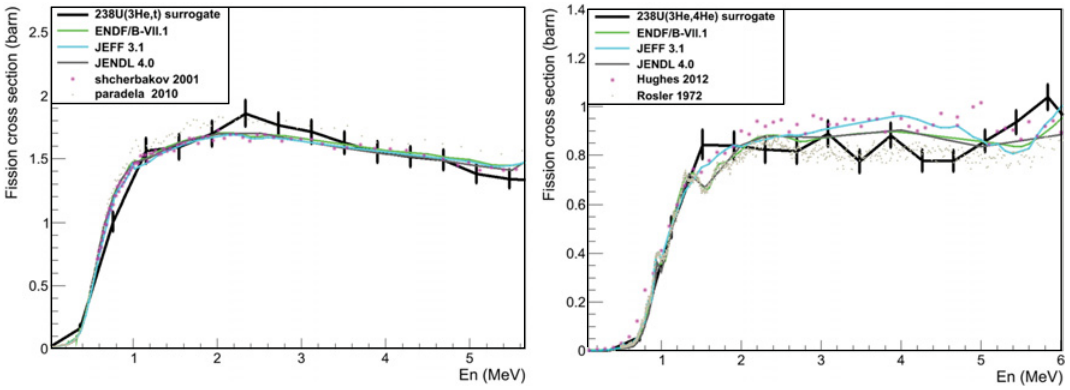

Figure 4. Preliminary neutron-induced fission cross section of ^{237}Np obtained with the $^{238}U(^{3}He,t)$ reaction (left) and of ^{236}U obtained with the $^{238}U(^{3}He,^{4}He)$ reaction (right) compared with neutron-induced data and evaluations.

leading to a strong increase of gamma emission which is here the only way of decay. Note that we have to perform some corrections at the data that can modify the cross sections at the highest excitation energies.

3.2 The ^{238}U(^{3}He,t)^{238}Np and ^{238}U(^{3}He,^{4}He)^{237}U reactions

The results presented in this subsection follow from the analysis of three groups of 8 strips. Each group of strips was located at a particular angle, namely 128, 132 and 140 degrees relative to the beam direction. As for the first subsection, because of the relatively low statistics, these results should be considered as preliminary. The left part of Figure 4 shows the fission cross section of ^{237}Np as a function of equivalent neutron energy obtained from the ^{238}U(^{3}He, t) reaction and the right part shows the fission cross section of ^{236}U obtained from the ^{238}U(^{3}He,^{4} He) reaction. The effective fission efficiency for these two reactions has been taken the same as for the (d,p) reaction.

Figure 4 shows an overall good agreement with the neutron data up to about 6 MeV for both reactions meaning there is no particle escape for the (^{3}He,^{4} He) and (^{3}He,t) reactions contrary to the (d,p) reaction. This result can be easily understood for the (^{3}He,^{4} He) reaction where a neutron is removed from the target. Moreover the fusion of the ^{3}He beam with oxygen and the subsequent evaporation of alphas occurs, but the energies of the latter (obtained by PACE4 code calculations) are not high enough to traverse the first part of the telescopes. Concerning the (^{3}He,t) reaction, the fusion of the ^{3}He beam with oxygen and the subsequent evaporation of tritons is very improbable. For these reasons, the fission cross sections for these two reactions are not underestimated as it is for the (d,p) reaction.

4. Conclusions and perspectives

We performed an experiment at the Oslo Cyclotron to investigate the validity of the surrogate method in the actinide region. The first results for the neutron-induced fission cross section of ^{238}U with the ^{238}U(d,pf) reaction show a systematic lowering relative to the neutron-induced data that could be due to the escape of the transfered neutron and/or to deuteron fusion with oxygen followed by proton evaporation. Model calculations will be performed to understand this issue. The capture cross section measured is about several times higher than in the neutron-induced reaction, although the disagrement decreases with increasing excitation energy. These discrepancies are probably due to differences between the spin/parity distributions populated in the surrogate and the neutron-induced

reactions. Note that the (d,p) reaction is particularly interesting to simulate neutron-induced reactions in inverse kinematics using radioactive beams. The first results for the fission cross sections of ^{237}Np and ^{236}U induced in the ^{238}U(^{3}He, tf) and ^{238}U(^{3}He,^{4}Hef) reactions, respectively, show an overall good agreement with neutron-induced data. A complete analysis of the results requires the use of all the 64 telescope strips. Moreover, we will also determine the capture cross section associated to the reactions ^{238}U(^{3}He,^{4} He) and ^{238}U(^{3}He, t). The ensemble of results expected from this work will provide an important step forward for understanding to which extent the surrogate method can be applied to extract neutron-induced cross sections in the actinide region.

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