^{232}Th(d,xn)^{230,232,233}Pa Cross Section Measurements

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Cross sections for the (d,n), (d,2n) and (d,4n) reactions on ^{232}Th were measured using the stacked-foil technique with beams provided by the ARRONAX cyclotron. These data are of relevance for the production of radionuclides. The measured cross sections were compared with previous measurements as well as with theoretical calculations using the code TALYS.

I. INTRODUCTION

The ARRONAX cyclotron [1], is a new facility installed in Nantes, France. A dedicated program has been launched on production of innovative radionuclides for PET imaging and for β^- and α targeted radiotherapy using proton or α particles. In this study, we have focused on cross section measurements using the stacked-foil technique [2] of one isotope dedicated to α radio-immunotherapy (α RIT) [3].

The α RIT [3] consists in binding an α emitter to an antibody in order to target and then to destroy tumor cells. Their high linear energy transfer give to α particles an important cytotoxic effect especially on small clusters of tumor cells ($\leq$ 100 μm) or isolated cells, minimising damage of surrounding healthy cells. ^{226}Th ($T_{1/2}$ = 31 min) is a novel therapeutic nuclide of interest since it has been found a more potent α emitter for leukemia therapies than ^{213}Bi [4]. Indeed, the ^{226}Th decay produces a cascade of four α particles with a cumulative energy of 27.7 MeV. An additional interest is the possibility to use a radionuclide generator system ^{230}U/^{226}Th. ^{230}U ($T_{1/2}$ = 21 days) could be produced directly via ^{231}Pa(p,2n)^{230}U, and indirectly via ^{230}Pa ($T_{1/2}$ = 17.4 days) using proton or deuteron beams through ^{232}Th(p,3n)^{230}Pa$\rightarrow^{230}$U, ^{232}Th(d,4n)^{230}Pa$\rightarrow^{230}$U. Twelve data sets are published concerning the ^{230}Pa cross section induced by protons [5] and only one by deuterons. As sometimes deuteron induced reactions give higher cross section values, it seems interesting to focus our study on their use as projectile on ^{232}Th target to produce ^{230}Pa. ^{230}Pa cross sections are measured, as well as contaminants created during irradiation. Our new sets are compared with the only existing ones [6] and with the TALYS code calculations [7].

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II. SET-UP AND DATA MEASUREMENT

Our experiment took place at the ARRONAX cyclotron, in the AX hall devoted to experiments in physics, radiolysis and radiobiology. The stacks were placed in air, on an irradiation station called Nice-III. The beam line is closed using a kapton foil (75 μm) and the stack was located about 7 cm away. Since the ^{232}Th(d,4n)^{230}Pa reaction has a threshold of 16 MeV and the deuteron energy available at ARRONAX does not exceed 35 MeV, two stacks were irradiated with, respectively, 22 and 30 MeV deuteron beam in order to cover the energy range of interest. All foils were purchased from Goodfellow® with high purity (> 99%). Each thin foil has been weighed before irradiation using a precise scale ($\pm$ 10^{-5}g) and scanned, to determine the thickness ($\approx$ 45 μm for ^{232}Th). We used a titanium monitor foil to record the particle flux through the stack with the ^{nat}Ti(d,x)^{48}V reaction, as suggested by IAEA [8]. In each foil, the ^{48}V activity value has been determined after the complete decay of ^{48}Sc ($T_{1/2}$ = 43.67 h). The half-life of ^{48}V is 15.9735(25) days [5]. Its three main γ lines [5] at energy of E_γ = 944.104 keV (I_γ = 7.870(7)%), E_γ = 983.517 keV (I_γ = 99.98(4)%) and E_γ = 1312.096 keV (I_γ = 98.2(3)%) were used.

In addition to monitor foils, a Faraday cup was placed after the stack to collect charges and control the intensity during the irradiation. The cross section values obtained from this intensity are in good agreement with the one extracted from the monitor (within 4.5%). The incident beam energy was fixed by the setting parameters of the cyclotron. The energy through each thin foil was determined in the middle of the foil using the SRIM software [9]. Energy losses in the kapton foil and air were taken into account. Our stacks were irradiated during 30 mn. After some cooling time, measurements were done using a high purity germanium detector with low-background lead shield from Canberra®. Gamma spectra were recorded using the LVis software from Ortec® in a suitable geometry previously calibrated with standard

γ sources (^{57,60}Co and ^{152}Eu) from Lea Cerca (France). The full widths at half maximum were 1.04 keV at 122 keV (^{57}Co γ ray) and 1.97 keV at 1332 keV (^{60}Co). The activity values of the produced radionuclides were derived from the spectra and the nuclear decay data given in table I, using the Fitzpeak spectroscopy software package [10]. The dead time during the counting was always lower than 10% in order to reduce the effect of sum peaks.

TABLE I. Produced radioisotope parameters [5][11]

Radioisotope	$T_{1/2}$(days)	E_γ(keV)	I_γ(%)	Reaction
^{230}Pa	17.4(5)	951.95	29.1(14)	^{232}Th(d,4n)
^{232}Pa	1.31(2)	969.315	42.3(6)	^{232}Th(d,2n)
^{233}Pa	26.967(2)	312.17	38.6(4)	^{232}Th(d,n)

Production cross section values can be determined from the activation formula (1) with the appropriate projectile flux:

$$\sigma = \frac{Act.A}{\Phi.N_a.\rho.e_f(1-e^{-\lambda.t})} \quad (1)$$

In this equation, the production cross section σ of a radioisotope depends on its measured activity (Act), its decay constant (λ), the target thickness (e_f), its atomic number (A), its density (ρ), the irradiation duration (t) and the projectile flux (Φ).

In our experiment, each target foil received the same projectile flux as the monitor foil that follows. It is then easier to use the relative equation (2) in which the knowledge of the projectile flux is no longer necessary. In this equation, the prime parameters are associated to ^{48}V monitor while the others relate to Pa isotopes.

$$\sigma = \sigma'.\frac{Act.A.\rho'.e'_f.(1-e^{-\lambda'.t})}{Act'.A'.\rho.e_f.(1-e^{-\lambda.t})} \quad (2)$$

To determine the activity associated to each radionuclide of interest, all the target and monitor foils were counted twice with an interval of 2 weeks and during more than 24 hours. The cross section uncertainty is estimated with a propagation error calculation. Since all the parameters of equation (2) are independent, the total error is expressed as a quadratic form. The main error sources come from the recommended cross section (around 12%), ^{230,232,233}Pa activity (up to 12%) , ^{48}V activity (less than 2%) and thickness uncertainty (around 1%). The contribution of the irradiation time uncertainty is not significant and has been neglected.

III. RESULTS

In order to be able to get data from all the Pa isotopes, the first ^{232}Th foil spectrum were acquired 2 days after the end of irradiation. All the nuclear parameters used for Pa isotopes are summarized in table I.

A. Production of ^{230}Pa

In the spectrum, we used the 951 keV γ line to determine the ^{230}Pa activity value. Several other ^{230}Pa γ lines have been also identified between 397 and 1027 keV with a branching ratio higher than 1%. Results from these γ lines were consistent, giving us confidence in our results. Using the second counting, we have verified that the γ line used was not fed by the decay of an other isotope and that the measured activity was consistent with the first one. The ^{230}Pa production cross section as a function of the deuteron energy is plotted in Fig. 1. Our data points are presented as full circles whereas data from Rama Rao *et al.* [6] are plotted as empty triangles. TALYS 1.4 [7] calculation values, performed using default parameters, are plotted as a solid line.

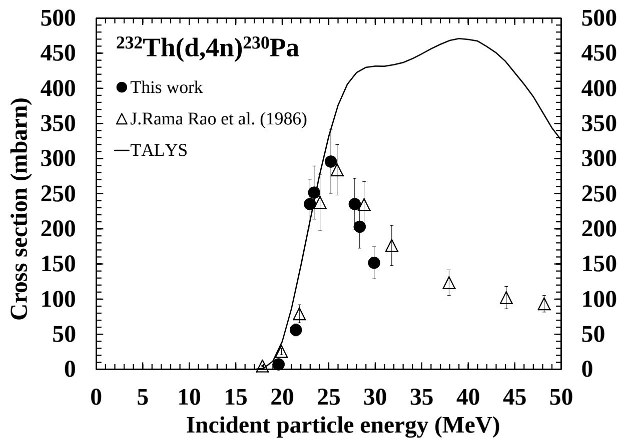

FIG. 1. Experimental cross section of ^{232}Th(d,4n)^{230}Pa

Our new data set is consistent with the energy threshold associated to ^{232}Th(d,4n)^{230}Pa and show a maximum of 320 mb at 24.35 MeV. Compared to the existing data of ref. [6], the shape of our data set as well as the maximum value of the cross section are in good agreement. However, the position of the maximum is slightly shifted. The TALYS calculation shows that neither the shape nor the maximum value of the cross section are reproduced.

B. Production of ^{232}Pa

^{232}Pa ($T_{1/2}$ = 1.31 d) emits many detectable γ lines. In their work, Rama Rao *et al.* choose to use the 894 keV line (I_γ = 22%). In our case, looking at the second counting when ^{232}Pa has totally disappeared, a peak at this energy is still present, which is coming from a sum peak between Pb X-ray (75 keV) from our shielding and ^{136}Cs γ line (819 keV) produced during the irradiation. Even if this contribution in the 894 keV γ line is small, we have preferred to use the 969 keV γ line with a higher branching ratio, 42.3% (table I). In this latter case, the contribution of ^{228}Ac (E_γ = 969 keV, I_γ = 15.8%) has been subtracted using its other γ lines. The ^{232}Pa cross section is represented in Fig. 2 with the existing data and TALYS calculation values.

Due to the ^{230}Pa energy range of interest, we only have data points in the tail of the cross section curve. The trend is consistent with the existing data set and our val-

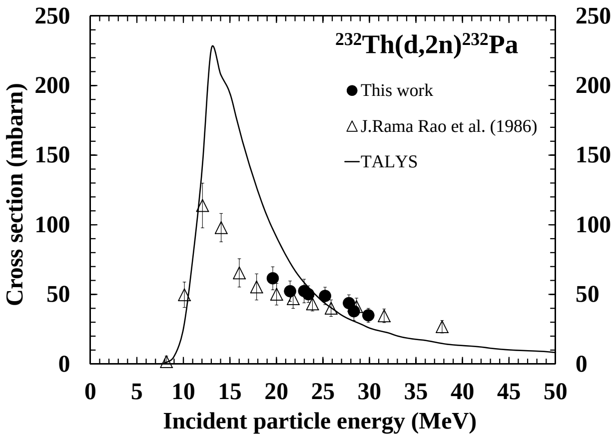

FIG. 2. Experimental cross section of ^{232}Th(d,2n)^{232}Pa

ues are slightly higher, mainly due to the different γ line used. To make a better comparison with ref.[6], if the cross section had been calculated, as in their work, using a branching ratio value $I_\gamma = 22\%$ for the 894 keV γ line and omitting the peak sum effect, the two data sets would be in total agreement. Using the actual branching ratio value, 19.8% as in [11], for this same γ line, the cross section values become 20% higher. It demonstrates the sum peak effect influence and the importance of the nuclear decay data updating. TALYS results using default parameters are not in agreement with data even if the shape agreement can be considered as not too bad.

C. Cumulative production of ^{233}Pa

^{233}Pa is produced directly through ^{232}Th(d,n) but also indirectly by the decay of ^{233}Th which is obtained via ^{232}Th(d,p). Since ^{233}Th has a short half life ($T_{1/2}$ = 21 min), we were only able to measure the ^{233}Pa cumulative cross section. These values compared to Rama Rao *et al.* and TALYS are presented in Fig. 3. To follow the decay of ^{233}Pa ($T_{1/2}$ = 26.967 d), we used the 312 keV γ line. Its high branching ratio leads to a small uncertainty associated to the activity value (around 2.7%). Our data are very similar to those of ref.[6].

The small difference can be accounted by the branching ratio they used($I_\gamma = 37\%$), which is lower than the actual value, 38.6% [5],[11].

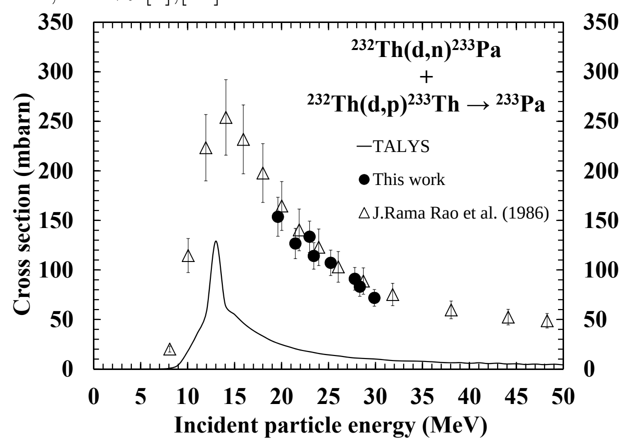

FIG. 3. Cumulative production cross section of ^{233}Pa

Repeating the calculation with their branching ratio value, our data increase by 4%, coming closer to the existing data. TALYS results underestimate the cross section and the peak width is poorly reproduced.

IV. CONCLUSION

In this work, new data sets concerning the ^{230,232,233}Pa production cross sections induced by deuterons have been obtained in order to compare with the values published by Rama Rao *et al.* in 1986. Despite same trends, some disagreement in the ^{230}Pa cross section magnitude are underlined. The ^{232,233}Pa trends show clearly that a difference between the branching ratio values can affect in a significant way the data. The TALYS 1.4 code is not yet capable of making good predictions for deuteron induced Pa isotopes on ^{232}Th and further developments, especially for break-up reactions, are necessary.

V. ACKNOWLEDGMENTS

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