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$^{232}\text{Th}(\text{d},\text{xn})^{230,232,233}\text{Pa}$ Cross Section Measurements at ARRONAX facility for the production of ^{230}U

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Abstract

Introduction

The targeted radionuclide therapy is one modality to treat cancer which consists in binding a radioactive isotope to a vector in order to target and then to destroy tumor cells. The choice of the isotope to be used depends on the characteristics of the targeted tumor. α emitters will be well suited for microscopic disease. Many isotopes are considered for this application. Among them, ^{213}Bi ($T_{1/2} = 46$ min) has already shown its toxic effects. It is used in several medical trials conjugated to monoclonal antibodies. However, another α emitter with a half life slightly lower seems more interesting. Indeed, ^{226}Th ($T_{1/2} = 31$ min) took part in an experiment with results published in 2009, showing that it induces cell death and activates apoptosis pathways with higher efficiencies than ^{213}Bi .

Methods

^{226}Th can be obtained via the ^{230}U α decay. The $^{232}\text{Th}(\text{d},4\text{n})^{230}\text{Pa} \rightarrow ^{230}\text{U}$ reaction was used to produce ^{230}U . Experimental cross sections for deuteron

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induced reactions on ^{232}Th were measured using the stacked-foil technique with beams provided by the ARRONAX cyclotron. Indeed, these data are of relevance for the production of radionuclides but only one data set performed 27 years ago is available and need to be change.

Results

Our new cross section values were compared with previous measurements and with results given by the code TALYS. Experimentally, same trends were observed with slight differences in orders of magnitude due to the nuclear data change. The Talys code is not yet able to well reproduce the data for deuterons induced reactions on thorium and improvements are ongoing.

Conclusions

We were able to calculate the thick-target yield in $\text{Bq}/\mu\text{A.h}$ of ^{230}U via the ^{230}Pa cross section measured in this work. This value allows now to a complete comparison between the different production routes.

Keywords: production cross section, stacked-foil technique, alpha targeted therapy, TALYS code, ARRONAX cyclotron, Thick-target yield (TTY)

1. 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. Since the accelerator is also able to deliver deuteron beams up to 35 MeV, we have reconsidered the possibility to use them to produce medical isotopes. ^{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 [2]. 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 $^{230}\text{U}/^{226}\text{Th}$. ^{230}U ($T_{1/2} = 21$ days) could be produced directly via $^{231}\text{Pa}(p,2n)^{230}\text{U}$, and indirectly via ^{230}Pa ($T_{1/2} = 17.4$ days) using proton or deuteron beams through $^{232}\text{Th}(p,3n)^{230}\text{Pa} \rightarrow ^{230}\text{U}$, $^{232}\text{Th}(d,4n)^{230}\text{Pa} \rightarrow ^{230}\text{U}$. Twelve data sets are published concerning the ^{230}Pa cross section induced by protons [3] 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 using the stacked-foil technique [4], as well as contaminants created during irradiation. Our new sets are compared with the only existing ones [5], other production routes [6] and with the TALYS code calculations [7].

2. 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 $^{232}\text{Th}(d,4n)^{230}\text{Pa}$ 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 ($\pm 10^{-5}\text{g}$) and scanned, to determine

the thickness ($\approx 45 \mu\text{m}$ for ^{232}Th). We used a titanium monitor foil to record the particle flux through the stack with the $^{nat}\text{Ti}(\text{d,x})^{48}\text{V}$ reaction, as suggested by IAEA [8]. In each foil, the ^{48}V activity value has been determined after the complete decay of ^{48}Sc ($T_{1/2} = 43.67 \text{ h}$). Nuclear data associated to ^{48}V are summarized in table 1.

Table 1: V-48 half-life and main γ rays [3]

Radioisotope	$T_{1/2}(\text{days})$	$E_\gamma(\text{keV})$	$I_\gamma(\%)$
^{48}V	15.9735 (25)	944.104	7.870 (7)
		983.517	99.98 (4)
		1312.096	98.2 (3)

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 ($^{57,60}\text{Co}$ 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 γ ray). The activity values of the produced radionuclides were derived from the spectra and the nuclear decay data given in table 2,

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 2: Produced radioisotope parameters [3][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)
				²³² Th(d,p) decay

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 ⁴⁸V monitor while the others relate to Pa isotopes.

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

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%), $^{230,232,233}\text{Pa}$ 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.

3. 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 2.

3.1. 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.* [5] are plotted as empty triangles. TALYS 1.4

[7] calculation values, performed using default parameters, are plotted as a solid line.

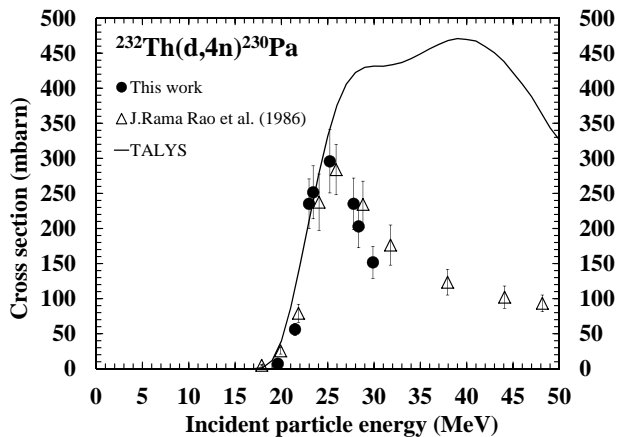


Figure 1: Experimental cross section of $^{232}\text{Th}(d,4n)^{230}\text{Pa}$

Our new data set is consistent with the energy threshold associated to $^{232}\text{Th}(d,4n)^{230}\text{Pa}$ and show a maximum of 320 mb at 24.35 MeV. Compared to the existing data of ref. [5], 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.

3.2. 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 ($I_\gamma = 22\%$). 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 2). In this latter case, the contribution of ^{228}Ac ($E_\gamma = 969$ keV, $I_\gamma = 15.8\%$) 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.

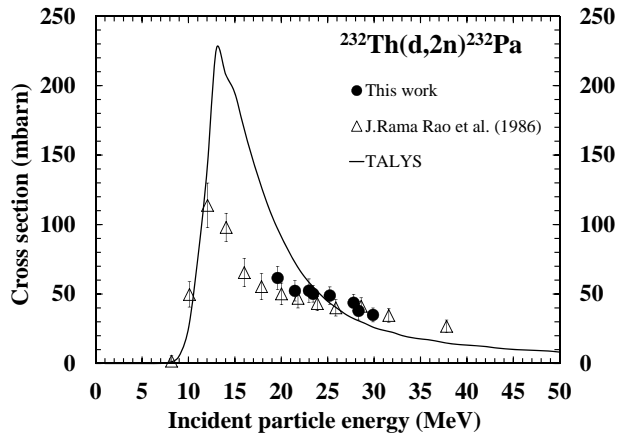


Figure 2: Experimental cross section of $^{232}\text{Th}(d,2n)^{232}\text{Pa}$

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 values are slightly higher, mainly due to the different γ line used. To make a better comparison with ref.[5], 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.

3.3. Cumulative production of ^{233}Pa

^{233}Pa is produced directly through $^{232}\text{Th}(d,n)$ but also indirectly by the decay of ^{233}Th which is obtained via $^{232}\text{Th}(d,p)$. 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 L.M.Slater [12], 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.[5]. The small difference can be accounted by the branching ratio they used ($L_\gamma = 37\%$), which is lower than the actual value, 38.6% [3],[11]. Repeating the calculation with their branching ratio value, our data increase by 4%, coming closer to the existing data.

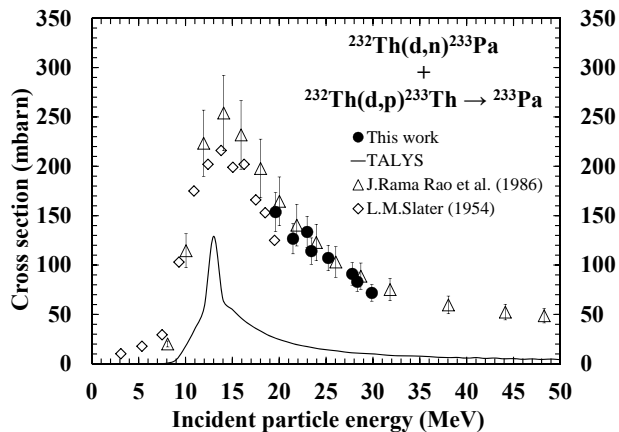


Figure 3: Cumulative production cross section of ^{233}Pa

In 1954, after an experiment looking at the ^{233}Th cross section, the cumulative cross section of ^{233}Pa was deduced [12]. The trend is consistent with

this work and with ref.[5] but also shows a difference of magnitude between the series. The TALYS results underestimate the cross section and the peak width is poorly reproduced.

4. Thick-target yield of ^{230}U

The thick-target yield (TTY) for the production of ^{230}U after the deuteron irradiation of ^{232}Th has been calculated using the ^{230}Pa cross section, previously determined. The results are resumed in table 3 with other production routes and plotted in Fig. 4. The TTY of the indirect production routes corresponds to the maximum activity of ^{230}U . This maximum is reached 27 days after the end of irradiation via ^{230}Pa β - decay and corresponds to $\approx 2.6\%$ of the ^{230}Pa activity initially produced.

Table 3: Thick-target yield (TTY) of different production routes

Reaction	Max. cross section (mb)	TTY (MBq/ $\mu\text{A.h}$)	Ref.
$^{232}\text{Th}(\text{d},4\text{n})^{230}\text{Pa} (\beta^-) ^{230}\text{U}$	296 ± 45	0.096 (29.9 MeV)	This work
$^{232}\text{Th}(\text{p},3\text{n})^{230}\text{Pa} (\beta^-) ^{230}\text{U}$	353 ± 15	0.24 (33.5 MeV)	[6]
$^{231}\text{Pa}(\text{d},3\text{n})^{230}\text{U}$	27.8 ± 3.4	0.119 (20.0 MeV)	[6]
$^{231}\text{Pa}(\text{p},2\text{n})^{230}\text{U}$	33.2 ± 5.3	0.245 (24.0 MeV)	[6]

Whatever the production route, direct or indirect, proton beams always give higher ^{230}U production values than deuteron beams. Both routes using protons are in the same order of magnitude. The indirect production route required a cyclotron with proton energy beam higher than 30 MeV and an elution of a $^{230}\text{Pa}/^{230}\text{U}$ generator. But in this case, ^{232}Th target is easier to obtain and handle than ^{231}Pa , which facilitates the routine production.

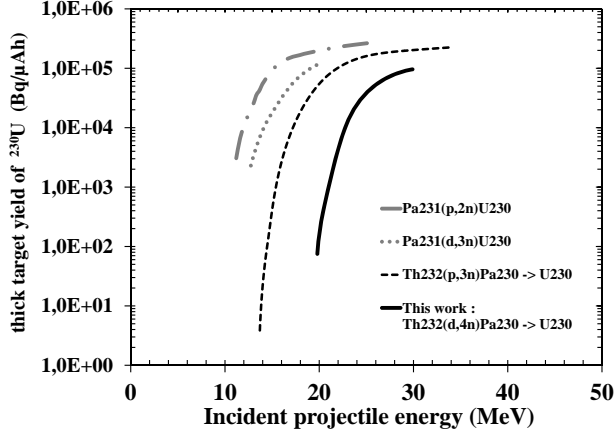


Figure 4: Thick-target yield of ^{230}U via different production routes

5. Conclusion

In this work, new data sets concerning the $^{230,232,233}\text{Pa}$ 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 $^{232,233}\text{Pa}$ 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. The $^{232}\text{Th}(d,4n)^{230}\text{Pa} (\beta^-) ^{230}\text{U}$ thick target yield has been calculated from the ^{230}Pa experimental cross section values previously obtained. This value comes as a complement of the work of A.Morgenstern et al [6] and confirms that the use of proton beam to produce ^{230}U is the best choice.

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