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^{232}Th(d,4n)^{230}Pa cross-section measurements at ARRONAX facility for the production of ^{230}U

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ABSTRACT

Introduction: ^{226}Th ($T_{1/2}$ = 31 min) is a promising therapeutic radionuclide since results, published in 2009, showed that it induces leukemia cells death and activates apoptosis pathways with higher efficiencies than ^{213}Bi. ^{226}Th can be obtained via the ^{230}U α decay. This study focuses on the ^{230}U production using the ^{232}Th(d,4n)^{230}Pa($\beta-$)^{230}U reaction.
Methods: Experimental cross sections for deuteron-induced reactions on ^{232}Th were measured from 30 down to 19 MeV using the stacked-foil technique with beams provided by the ARRONAX cyclotron. After irradiation, all foils (targets as well as monitors) were measured using a high-purity germanium detector.
Results: Our new ^{230}Pa cross-section values, as well as those of ^{232}Pa and ^{233}Pa contaminants created during the irradiation, were compared with previous measurements and with results given by the TALYS code. Experimentally, same trends were observed with slight differences in orders of magnitude mainly due to the nuclear data change. Improvements are ongoing about the TALYS code to better reproduce the data for deuteron-induced reactions on ^{232}Th.
Conclusions: Using our cross-section data points from the ^{232}Th(d,4n)^{230}Pa reaction, we have calculated the thick-target yield of ^{230}U, in Bq/μA·h. This value allows now to a full comparison between the different production routes, showing that the proton routes must be preferred.

1. Introduction

The ARRONAX cyclotron [1], is a new facility installed in Nantes, France. A dedicated program has been launched on the production of innovative radionuclides for PET imaging and for β^- and α targeted therapy 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. This cascade conducts to the ^{210}Pb long half-life. The ICRP publication n° 68 [3] permits to obtain the ^{210}Pb committed effective dose in Sv·Bq^{-1}, leading to a calculated admissible value of 0.7 mSv for 10 mCi of ^{226}Th injected. 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 → ^{230}U, ^{232}Th(d,4n) ^{230}Pa → ^{230}U. Twelve data sets are published concerning the ^{230}Pa cross section induced by protons [4] 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 section is measured using the stacked-foil technique [5], as well as contaminants created during irradiation. Our new set is compared with the only existing one [6], with other production routes emerging, and with the TALYS code calculations [8].

2. Materials and methods

Our experiment was carried out 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 of thickness) and the stack was located about 7 cm downstream. 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 (>99.5%). Each thin foil has been weighed before irradiation using an accurate scale ($\pm 10^{-5}$ g) and scanned to precisely determine their area. From these values and assuming that the thickness is homogeneous over all surfaces, the thickness has been deduced (around 45 μm for ^{232}Th and 10 μm for ^{nat}Ti). 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

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Table 1
Nuclear decay data and contributing reactions of ^{48}V and ^{230,232,233}Pa [4,15].

Radionuclide	$T_{1/2}$ (days)	E_γ (keV)	I_γ (%)	Reaction(s)	$E_{threshold}$ (MeV)
^{48}V	15.9735 (25)	944.104	7.870 (7)	^{47}Ti(d,n)	0.00
		983.517	99.98 (4)	^{48}Ti(d,2n)	7.32
		1312.096	98.2 (3)	^{49}Ti(d,3n)	15.79
				^{50}Ti(d,4n)	27.16
^{230}Pa	17.4 (5)	951.95	29.1 (14)	^{232}Th(d,4n)	16.01
^{232}Pa	1.31 (2)	969.315	41.6 (19)	^{232}Th(d,2n)	3.54
^{233}Pa	26.967 (2)	312.17	38.6 (4)	^{232}Th(d,n) + (d,p) decay	0.00
				^{232}Th(d,p) decay	0.00

IAEA [9–12]. In each foil, the ^{48}V activity value has been determined after the complete decay of ^{48}Sc ($T_{1/2}$ = 43.67 h). Nuclear data associated to ^{48}V are summarized in Table 1.

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 using the Faraday cup 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 [13]. Energy losses in the kapton foil and air were taken into account. Typical irradiations were carried out with about 100 nA for 30 min. After some cooling time, measurements were done using a high-purity germanium detector with low-background lead and copper shield from Canberra[ered]. 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 γ ray). The ^{230}Pa activity values were derived from the spectra and the nuclear decay data given in Table 1, using the Fitzpeak spectroscopy software package [14]. The dead time during the counting was always lower than 10% in order to reduce the effect of sum peaks.

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

$$\sigma = \frac{\mathrm{Act}\cdot A}{\chi\cdot\Phi\cdot N_a\cdot\rho\cdot e_f\left(1-e^{-\lambda t}\right)} \tag{1}$$

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) and its purity (χ), 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 Eq. (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 ^{230}Pa.

$$\sigma = \sigma'\cdot\frac{\chi'\cdot\mathrm{Act}\cdot A\cdot\rho'\cdot e_f'\left(1-e^{-\lambda' t}\right)}{\chi\cdot\mathrm{Act}\cdot A'\cdot\rho\cdot e_f\left(1-e^{-\lambda t}\right)} \tag{2}$$

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 h. The cross-section uncertainty is estimated with a propagation error calculation. Since all the parameters of Eq. (2) are independent, the total error is expressed as a quadratic sum (Eq. (3)). 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.

$$\frac{\Delta\sigma}{\sigma} = \sqrt{\left(\frac{\Delta\sigma'}{\sigma'}\right)^2 + \left(\frac{\Delta\mathrm{Act}}{\mathrm{Act}}\right)^2 + \left(\frac{\Delta\mathrm{Act}'}{\mathrm{Act}'}\right)^2 + \left(\frac{\Delta e_f}{e_f}\right)^2 + \left(\frac{\Delta e_f}{e_f}\right)^2} \tag{3}$$

Using the cross-section values obtained in this work, we have calculated the thick-target yield (TTY) in Bq/μA·h as a function of the projectile energy, using the relation (4).

$$\mathrm{TTY} = \Phi.\chi.\frac{N_a\cdot\rho}{A}\left(1-e^{-\lambda.t}\right)\int_{E_{\min}}^{E_{\max}}\frac{\sigma(E)}{\frac{\mathrm{d}E}{\mathrm{d}x}}\mathrm{d}E \tag{4}$$

where χ is the target enrichment and $\frac{\mathrm{d}E}{\mathrm{d}x}$ is the specific energy loss of the projectile in the target material (MeV·cm^{-1}). In a thick target, the incident particle energy decrease with the penetration depth. E_{max} corresponds to the incident projectile energy when it enters into the target whereas E_{min} corresponds to its energy when it leaves the target.

3. Results

After the irradiation of a thorium target at these energies by deuterons, two other protactinium isotopes than ^{230}Pa are produced: ^{232}Pa with a short half-life of 1.3 days and ^{233}Pa with a longer half-life of 27 days. The production cross sections of these two contaminants have been obtained and compared with those in Ref. [6]. A different magnitude between both series is observed, resulting, especially for the contaminants, from the nuclear data value change since 1986.

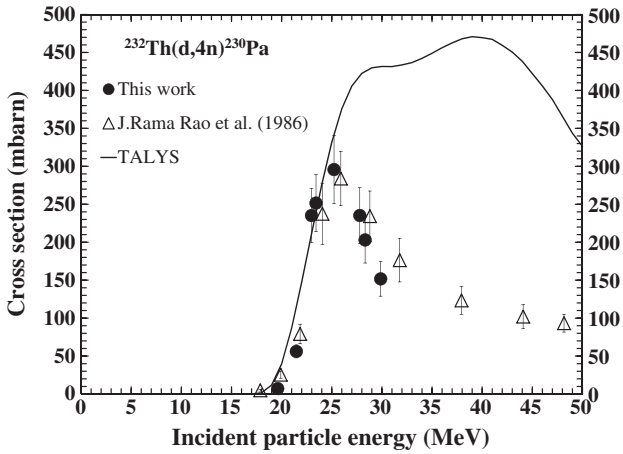

Fig. 1. Experimental cross section of ^{232}Th(d,4n)^{230}Pa.

Table 2
Measured cross section for the Pa isotopes.

E (MeV)	^{230}Pa	^{232}Pa	^{233}Pa
19.61 ± 0.39	7.18 ± 1.16	61.62 ± 8.27	153.83 ± 19.71
21.47 ± 0.32	56.22 ± 8.60	52.34 ± 7.33	126.81 ± 15.29
22.98 ± 0.46	235.50 ± 35.49	52.46 ± 8.46	133.68 ± 15.74
23.43 ± 0.52	251.86 ± 37.81	50.19 ± 5.97	114.24 ± 13.35
25.23 ± 0.45	296.05 ± 45.13	48.96 ± 6.26	107.28 ± 12.86
27.79 ± 0.33	235.44 ± 36.75	43.80 ± 5.90	91.14 ± 11.42
28.33 ± 0.38	203.23 ± 30.53	37.93 ± 6.64	83.19 ± 9.74
29.88 ± 0.32	151.86 ± 22.81	35.01 ± 5.02	71.87 ± 8.43

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 another 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 and the numerical values are reported in Table 2. Our data points are presented as full circles whereas data from Rama Rao et al. [6] are plotted as empty triangles. TALYS 1.4 [8] calculation values, performed using default parameters, are plotted as a solid line.

Our new data set is consistent with the energy threshold associated to ^{232}Th(d,4n)^{230}Pa and show a maximum of 296 mb at 25.23 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. Making the calculation with the older branching ratio used by Rama Rao et al. (28%), our values increase by 5%, coming closer to the existing series. The TALYS calculation shows that neither the shape nor the maximum value of the cross section is reproduced.

3.1.1. Production of the ^{232}Pa contaminant

^{232}Pa ($T_{1/2}$ = 1.31 days) emits many detectable γ lines. In their work, Rama Rao et al. [6] chose to use the 894-keV γ line (22%). In our case, looking at the second counting taken when ^{232}Pa had totally disappeared, a peak at this energy is still present. We found that this is coming from a sum peak between Pb x-rays (75 keV) from our shielding and the ^{136}Csγ line (819 keV), a fission fragment produced during the irradiation. We have preferred to use the 969-keV γ line with a higher branching ratio of 42.3% (Table 1), subtracting the contribution of ^{228}Ac (Eγ = 969 keV, I = 15.8%). Recently, we have performed a new experiment, adding 3 mm of copper shield to attenuate, by a factor 11, the 75-keV x-ray emission from lead. In this experiment, there is no longer the peak in the second measurement, indicating that the source was the one identified. The ^{232}Pa cross-section data are presented in Fig. 2. Due to the ^{230}Pa energy range of interest, we only measured cross sections in the tail of the curve. The trend is consistent with the existing data set from Ref. [6]. Our values are slightly higher, mainly due to the different γ line used and the updated nuclear decay data. In fact, since 1986, the 894-keV γ line branching ratio used by Rama Rao et al. [6] decreased from 22% [6] to 19.8% [15]. TALYS results using default parameters are not in agreement with the data even though the shape is not too bad.

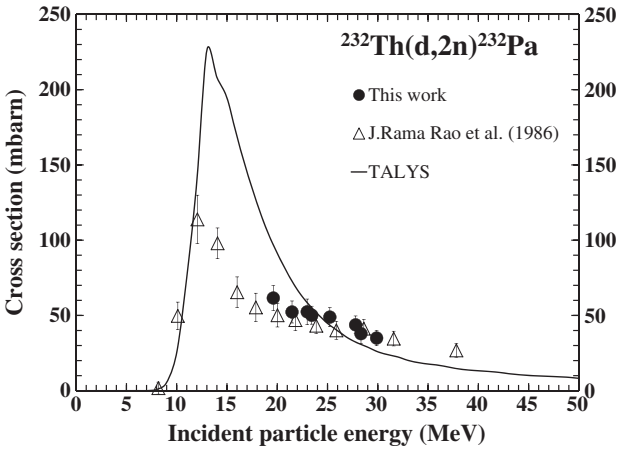

Fig. 2. Experimental cross section of ^{232}Th(d,2n)^{232}Pa.

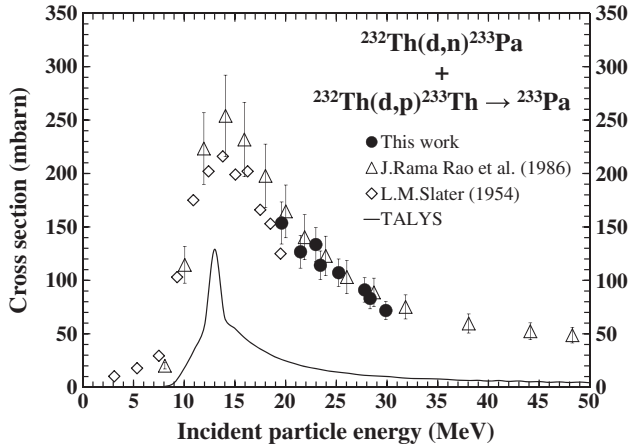

Fig. 3. Experimental cumulative production cross section of ^{233}Pa.

3.1.2. Cumulative production of the ^{233}Pa contaminant

^{233}Pa is produced directly through ^{232}Th(d,n) but also indirectly by the decay of ^{233}Th produced 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 are plotted and compared to Ref. [6] and TALYS in Fig. 3. We used the 312-keV γ line to follow the decay of ^{233}Pa ($T_{1/2}$ = 26.967 days). Its high branching ratio (Table 1) leads to a small uncertainty associated with the activity value (around 2.7%). Our data are very similar to those of Ref. [6]. The small difference can be accounted to the branching ratio they used (I = 37%), which is lower than the current recommended value (I = 38.6%) listed in the databases [4,15]. The TALYS results underestimate the amplitude and the peak width is poorly reproduced.

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

The thick-target yield (TTY) of ^{230}U after the deuteron irradiation of ^{232}Th has been calculated using the ^{230}Pa cross section, previously determined. In this case, ^{230}U is obtained from the $\beta-$ decay (branching ratio of 7.8%) of ^{230}Pa. For the TTY calculation, different scenarios can be imagined, depending on the strategy applied to collect ^{230}U. The first one is to consider that 7.8% of the ^{230}Pa atoms

Table 3
^{230}U Thick-target yield (TTY) of different production routes.

Reaction	Max. cross section (mb)	TTY (MBq/μA·h)	Ref.
^{232}Th(d,4n)^{230}Pa ($\beta-$) ^{230}U	296 ± 45	0.101 (29.9 MeV)	This work
^{232}Th(p,3n)^{230}Pa ($\beta-$) ^{230}U	353 ± 15	0.24 (33.5 MeV)	[7]
^{231}Pa(d,3n)^{230}U	27.8 ± 3.4	0.119 (20.0 MeV)	[7]
^{231}Pa(p,2n)^{230}U	33.2 ± 5.3	0.245 (24.0 MeV)	[7]

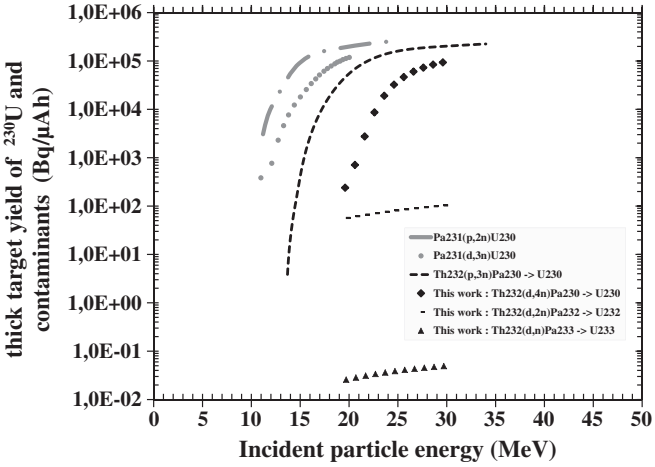

Fig. 4. Thick-target yield of ^{230}U via different production routes and contaminants.

produced at the end of the irradiation will become ^{230}U atoms. This is a theoretical value that cannot be reached because it corresponds to an infinite number of processing. The second one corresponds to only one process performed when ^{230}U activity is at maximum. The best elution condition is probably between these two processes but, in order to compare our TTY values with those from other production routes, we have decided to show the results of the second case. The maximum yield value is summarized in Table 3 with other production routes and the results are plotted in Fig. 4. The TTY of the indirect production routes (this work and Ref. [7]) corresponds to the maximum activity of ^{230}U reached 27 days after the end of irradiation via ^{230}Pa $\beta-$ decay and corresponds to ≈2.6% of the ^{230}Pa activity initially produced.

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}Pa/^{230}U generator. But in this case, ^{232}Th target is easier to obtain and handle than the radioactive and non-natural ^{231}Pa target, which facilitates the routine production. As a complement to this work, the ^{232}U and ^{233}U TTY have been calculated and are plotted in Fig. 4. At 29.9 MeV, their significance is, respectively, one thousand and 2 million times lower than the ^{230}U production.

4. Conclusion

In this work, new data set concerning the ^{230}Pa production cross section induced by deuterons has been obtained, in agreement with the existing one. The TALYS 1.4 code is not yet able of making good predictions for deuteron-induced Pa isotopes on ^{232}Th and further developments, especially for break-up reactions, are ongoing (TALYS version 1.6). In addition, in our study fission fragments are produced in large quantities, especially from 25 MeV, which could affect the cross-section prediction for activation products. The ^{232}Th(d,4n)^{230}Pa ($\beta-$) ^{230}U thick-target yield has been calculated from the ^{230}Pa experimental cross section. This value comes as a complement of the work of Morgenstern et al. [7] and confirms that the use of proton beam to produce ^{230}U is the best choice.

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