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Production of plutonium tracers in ^{237}Np nuclear reactions with ^{3}He-ions in the energy range from 26 to 60 MeV

J. Aaltonen[a,*], P. Dendooven[b], E.A. Gromova[c], V.A. Jakovlev[c], W.H. Trzaska[b]

[a]*Laboratory of Radiochemistry, Department of Chemistry, University of Helsinki, P.O. Box 55, FIN-00014 Helsinki, Finland*
[b]*Department of Physics, University of Jyväskylä, P.O. Box 35, FIN-40351 Jyväskylä, Finland*
[c]*Laboratory of Nuclear Reactions and Nuclear Medicine, V.G. Khlopin Radium Institute, 28 2nd Murinski Av., 194021 St. Petersburg, Russia*

Abstract

Cross-sections for the reaction ^{237}Np+^{3}He→^{236,237,238}Pu at ^{3}He bombarding energies from 26 to 60 MeV were measured. Thick-target yields, based on the measured and previously known cross-sections were constructed. The results are discussed and compared with other reactions leading to the formation of the same final nuclei. © 1998 Elsevier Science S.A.

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1. Introduction

Due to the strongly ionizing properties of alpha particles (large energy loss within a short range), biological tracers are required to be free of alpha-emitters such as, e.g., ^{236}Pu or ^{238}Pu. The nucleus ^{237}Pu ($T_{1/2}$=45.13±0.07 d [1]), which decays mainly by electron capture rather than by alpha emission, is, for that reason, used as tracer to study biological pathways of plutonium in the environment [2]. However, alpha-emitters can and are used as nonbiological tracers. For example ^{236}Pu ($T_{1/2}$=2.851±0.008 y [1]) is mainly used as a tracer for the control of ^{239}Pu release into the environment from the nuclear fuel cycle.

An important aspect in the use and choice of a tracer is its production method. For the nuclide ^{237}Pu the following reactions have been used:

$$^{237}\mathrm{Np(p,n)}^{237}\mathrm{Pu}, \quad (1)$$

$$^{237}\mathrm{Np(d,2n)}^{237}\mathrm{Pu}, \quad (2)$$

$$^{238}\mathrm{U}(^{3}\mathrm{He,4n})^{237}\mathrm{Pu}, \quad (3)$$

and

$$^{235}\mathrm{U}(\alpha,2\mathrm{n})^{237}\mathrm{Pu}. \quad (4)$$

Of these, the most productive reactions (1 and 2) require the irradiation of neptunium with 40 MeV protons [3,4] or 25 MeV deuterons [5]. With such a choice of projectiles and bombarding energies, a number of competing reaction channels lead to other plutonium isotopes. For example, the activity ratio $A(^{236}\mathrm{Pu}+^{238}\mathrm{Pu})/A(^{237}\mathrm{Pu})$ is 0.125 for reaction (1) [3] and 0.07–0.16 for reaction (2) [5]. Reactions (3) [6,7] and (4) [8] are more selective for the production of ^{237}Pu than reactions (1) and (2) at the price of substantially lower yields.

Alternatively, ^{236}Pu and ^{237}Pu can also be produced in the reactions:

$$^{237}\mathrm{Np}(^{3}\mathrm{He,p3n})^{236}\mathrm{Pu}, \quad (5)$$

and

$$^{237}\mathrm{Np}(^{3}\mathrm{He,p2n})^{237}\mathrm{Pu}. \quad (6)$$

In this case, the contamination by ^{238}Pu comes from the reaction:

$$^{237}\mathrm{Np}(^{3}\mathrm{He,pn})^{238}\mathrm{Pu}. \quad (7)$$

Until now, reactions (5), (6) and (7) were investigated only at ^{3}He-ion energies up to 27 MeV [9,10]. The present work is devoted to the study of these cross-sections in the energy range from 26 to 60 MeV.

*Corresponding author. Tel.: 358 9 19140134; fax: 358 9 19140121.

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Table 1
Decay parameters [1] used for identification of the studied nuclei

Nuclide	Emission	Half-life	Energy (keV)	Intensity (%)
^{237}Np	Alpha	$2.14 \cdot 10^6$ y	4768	100
^{236}Pu	Alpha	2.851 y	5753	100
^{237}Pu	$K_{\alpha 1}$ X-ray	45.17 d	101.06	21.8
^{238}Pu	Alpha	87.74 y	5487	100

2. Experimental

The neptunium targets were prepared by multiple thermal decomposition of neptunium nitrate on 10 μm thick aluminum backings. The thickness of the ^{237}Np layer on each target was about 100 μg cm^{-2}. Two sets of eight targets, each separated by 10 μm thick aluminum foils, were irradiated in two irradiations with 40 MeV $^3He^+$ and 60 MeV $^3He^{2+}$ ion beams during 12 and 4 h, respectively. The irradiations were performed with the K-130 cyclotron at the Department of Physics of the University of Jyväskylä. The beam current in both irradiations was about 3 μA (3 and 1.5 particle μA, respectively).

The alpha-activities of ^{236}Pu and ^{238}Pu from reactions (5) and (7) were measured with surface-barrier silicon detectors with a sensitive area of 200 mm^2 and an energy resolution of 30 keV at 5767 keV. The activity of ^{237}Pu formed in the reaction (6) was determined from gamma-ray spectra measured with a high purity Ge-detector with a resolution of 2.0 keV at 1332 keV. The spectra were analyzed with the GANAAS (gamma activity and neutron activation analysis system) computer program distributed to the V.G. Khlopin Radium Institute by the IAEA.

In order to determine the activity of ^{237}Pu in the irradiated Np targets, a chemical separation of plutonium from neptunium, protactinium and fission products had to be made. The separation of plutonium from neptunium and fission products was carried out by using a novel extraction chromatographic resin — TRU.Spec resin made by Eichrom Industries, USA — according to the following method:

The neptunium targets were dissolved in concentrated nitric acid and then treated as described in the work of Horwitz et al. [11]. In the present work, the initial sample solutions consisted of 3 ml of 2 M nitric acid where 0.5 ml of 1 M solution of ferrous sulfaminate and 50 mg of sodium nitrite were added.

The elution scheme included the following four steps:

1. Loading of the sample solution onto the column (bed height 5 cm×7.6 mm I.D.: 2.26 cm^3 bed volume, 1.6 cm^3 free column volume) and rinsing with 20 ml of 2 M nitric acid. Rinsing of the column with 3.4 ml of 9 M hydrochloric acid (to convert the resin into chloride form).
2. Elution of the americium fraction with 20 ml of 4 M hydrochloric acid.
3. Elution of the plutonium fraction with 20 ml of 4 M hydrochloric acid–0.1 M hydroquinone.
4. Elution of the neptunium fraction with 20 ml of 1.0 M hydrochloric acid–0.03 M oxalic acid.

In order to make plutonium samples for alpha- and gamma-spectrometry, the following procedures were used:

1. Evaporating the solution from step 4 of the elution process to dryness.
2. Treating the residue with a mixture of concentrated nitric acid and concentrated hydrochloric acid.
3. Evaporating the solution to dryness.
4. Dissolving the residue in 1 M nitric acid.
5. Coprecipitation of plutonium with neodymium fluoride [12].
6. Filtering the solution through a Millipore filter.

Millipore filters were used as samples for alpha- and gamma-spectrometry. Ten samples were made by this method. The samples contained ^{236}Pu, ^{237}Pu, ^{238}Pu and a residual amount of ^{237}Np. The purification factor of plutonium from neptunium was about 10^3 and the chemical yield of plutonium about 80%.

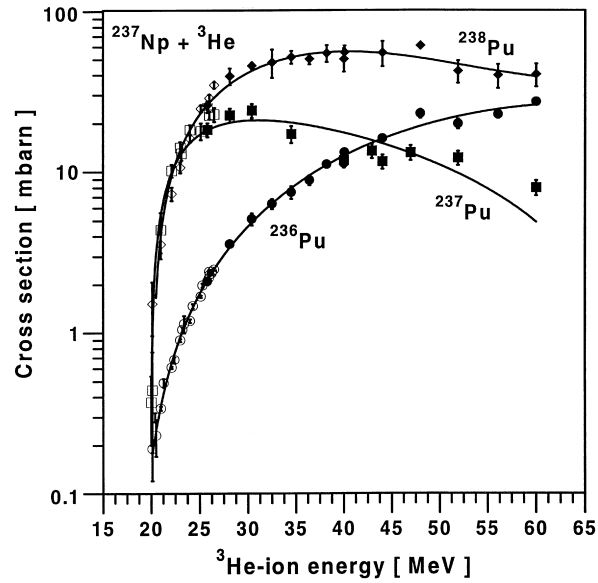

Fig. 1. Cross-sections of the ^{237}Np(^{3}He,pxn)$^{239-x}$Pu reactions. The open symbols are from Refs. [9,10]; the closed symbols are from this work. Solid lines are polynomial fits of the experimental cross-sections.

Table 2
Cross-sections of ^{236}Pu, ^{237}Pu and ^{238}Pu formation in the ^{237}Np+^{3}He reactions

E_{He-3} (MeV)	σ_{Pu-236} (mb)	σ_{Pu-237} (mb)	σ_{Pu-238} (mb)
25.8	2.10±0.10	18.3±1.8	26.0±1.5
28.1	3.57±0.16	22.5±2.3	39.4±4.7
30.4	5.1±0.4	24.1±2.6	45.7±2.1
32.5	6.4±0.5		48.1±9.6
34.5	7.5±0.7	17.2±2.1	51.8±4.7
36.4	8.9±0.6		50.4±3.8
38.2	11.1±0.4		54.8±6.6
40.0	11.3±0.3	11.8±1.2	50.3±8.4
40.1	13.1±0.7		55.4±5.5
42.9		13.4±1.4	
44.0	16.1±0.9	11.5±1.2	55.0±9.9
47.0		13.1±1.3	
48.0	23.0±1.5		61.0±2.4
51.9	19.8±1.4	12.2±1.2	42.3±6.8
56.1	22.7±1.2		39.8±6.8
60.0	27.2±0.8	7.99±0.86	40.4±6.7

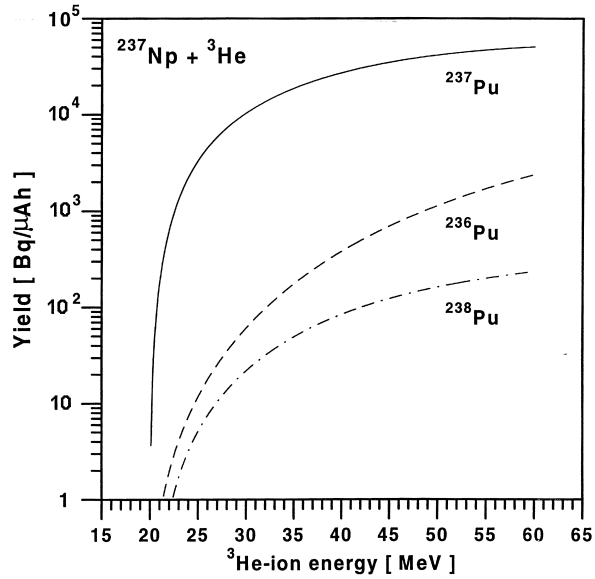

Fig. 2. Thick-target yields of ^{236}Pu, ^{237}Pu and ^{238}Pu in the ^{3}He-induced reactions on ^{237}Np calculated on the basis of the cross-sections measured in this work and in Refs. [9,10].

3. Results and discussion

The decay parameters used in the calculations of the experimental cross-sections were taken from Ref. [1] and are presented in Table 1. The cross-sections of the reactions ^{237}Np(^{3}He,*xnyp*) calculated from the measurements are presented in Fig. 1 and Table 2. The data of our earlier experiments [9,10] are also presented in Fig. 1.

The calculated thick-target yields of ^{236}Pu, ^{237}Pu and ^{238}Pu are shown in Fig. 2. It should be mentioned that the yields were calculated for the case of $^{3}He^{2+}$ ions. In order to obtain the yields for the case of $^{3}He^{+}$ ions, the values in Fig. 2 should be multiplied by two.

In Table 3, the yield of ^{237}Pu produced in the reaction (6) as well as the combined contamination by the alpha emitting isotopes ^{236}Pu and ^{238}Pu are compared with the respective values calculated for previously used reactions. It can be seen that the ^{237}Np+^{3}He reaction, while using comparable projectile energies, has an optimal set of the most important characteristics required for the use of ^{237}Pu as a tracer. For example, the yield of ^{237}Pu from the ^{237}Np+^{3}He reaction is about ten times higher than that from the ^{238}U+^{3}He reaction.

In order to obtain the best isotopical purity of the ^{237}Pu sample, the chemical separation of plutonium from neptunium has to be carried out as soon as possible after the end of bombardment (EOB). Otherwise, additional formation of ^{236}Pu and ^{238}Pu will take place by the decay of ^{236s}Np (^{236s}Np is the short-lived isomeric state of ^{236}Np with $T_{1/2}=22.5\pm0.4$ h) and ^{238}Np which are produced in the ^{237}Np+^{3}He irradiation as well. In this work, as seen in Table 3, the impurity factor of the final ^{237}Pu tracer will be among the best achievable via a direct irradiation.

Table 3
Yields and radionuclide purity of ^{237}Pu in various reactions

Reaction	Projectile energy (MeV)	Yields (kBq/μA h)	Composition at EOB[a]	Reference
^{235}U(α,2n)^{237}Pu	30	2	0.002	[8]
^{238}U(^{3}He,4n)^{237}Pu	26	0.4	0.001	[7]
	38	2	0.039	[6]
^{237}Np(p,n)^{237}Pu	17.5	24	0.125	[3]
	40.0	160	0.33	[4]
^{237}Np(d,2n)^{237}Pu	15	14	0.07	[5]
	25	48	0.16	
^{237}Np(^{3}He,p2n)^{237}Pu	26.7	6.1	0.002–0.005[b]	[9,10]
^{237}Np(^{3}He,p2n)^{237}Pu	60	51	0.01–0.05[b]	This work

[a] Ratio $A(^{236}Pu+^{238}Pu)/A(^{237}Pu)$.
[b] The minimum value of the activity ratio can be obtained when the chemical purification of plutonium is done immediately after EOB.

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