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Production of ^{235}Np, ^{236}Pu and ^{237}Pu *via* nuclear reactions on ^{235,236,238}U and ^{237}Np targets

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Proton and ^{3}He *induced nuclear reactions / Radioisotopes of neptunium and plutonium / Excitation functions / Thick-target yields / Chemical separation*

Summary. A summary of methods for cyclotron production of ^{235}Np (396.1 d), ^{236}Pu (2.858 y) and ^{237}Pu (45.2 d) *via* nuclear reactions with protons and ^{3}He-ions on ^{235,236,238}U and ^{237}Np targets in wide energy ranges is given. Methods for the chemical separation and purification of these nuclides from the irradiated uranium and neptunium targets are described. Cross sections, yields and radionuclidic purities of ^{235}Np, ^{236}Pu and ^{237}Pu are presented and compared with literature data on the nuclear reactions leading to these radionuclides. The nuclear reactions with the so far highest known yields of ^{235}Np, ^{236}Pu and ^{237}Pu are determined: ^{236}U$(p, 2n)^{235}$Np, ^{237}Np$(p, 2n + pn\beta^-)^{236}$Pu and ^{237}Np$(p, n)^{237}$Pu, respectively. The highest radionuclidic purity of ^{235}Np, ^{236}Pu and ^{237}Pu tracers can be reached with the ^{236}U$(p, 2n)^{235}$Np, ^{236}U$(p, n\beta^-)^{236}$Pu and ^{237}Np$(^3$He$, t)^{237}$Pu reactions, respectively. In addition new cross sections and yield data of the ^{236}U$(^3$He$, p3n)^{235}$Np reaction in the energy range 42.4–60 MeV are given.

1. Introduction

The nuclides ^{236}Pu (2.858 y), ^{237}Pu (45.2 d) and ^{235}Np (396.1 d) are convenient tracers for studying the distribution and biological pathways of plutonium and neptunium in the environment [1]. Since 1976 methods for the production of these radionuclides *via* nuclear reactions with 18 MeV protons and 27 MeV ^{3}He-ions on ^{235,236,238}U and ^{237}Np targets have been studied using the Åbo Akademi 103 cm AVF cyclotron (MGC-20). In 1992 the new K-130 cyclotron of the University of Jyväskylä extended the energy range of the studied nuclear reactions up to 40 MeV of protons and 60 MeV of ^{3}He-ions. Besides the investigations also commercial productions of plutonium and neptunium tracers have been performed with these cyclotrons.

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Plutonium-236

The alpha-emitter ^{236}Pu ($T_{1/2} = 2.858 \pm 0.008$ y [2]) is mainly used as a tracer for the control of ^{238,239,240}Pu release into the environment from the nuclear fuel cycle. The following ^{236}Pu production reactions have been studied [3–16]:

$$^{235}\mathrm{U}(^3\mathrm{He}, 2n + pn\beta^-)^{236}\mathrm{Pu}\,, \quad (1)$$

$$^{236}\mathrm{U}(p, n)^{236\mathrm{m}}\mathrm{Np} \xrightarrow{\beta^-} {}^{236}\mathrm{Pu}\,, \quad (2)$$

$$^{236}\mathrm{U}(^3\mathrm{He}, 3n)^{236}\mathrm{Pu}\,, \quad (3)$$

$$^{236}\mathrm{U}(^3\mathrm{He}, t)^{236\mathrm{m}}\mathrm{Np} \xrightarrow{\beta^-} {}^{236}\mathrm{Pu}\,, \quad (4)$$

$$^{238}\mathrm{U}(p, 3n)^{236\mathrm{m}}\mathrm{Np} \xrightarrow{\beta^-} {}^{236}\mathrm{Pu}\,, \quad (5)$$

$$^{238}\mathrm{U}(^3\mathrm{He}, 5n + p4n\beta^-)^{236}\mathrm{Pu}\,, \quad (6)$$

$$^{237}\mathrm{Np}(p, 2n)^{236}\mathrm{Pu}\,, \quad (7)$$

$$^{237}\mathrm{Np}(p, pn)^{236\mathrm{m}}\mathrm{Np} \xrightarrow{\beta^-} {}^{236}\mathrm{Pu}\,, \quad (8)$$

$$^{237}\mathrm{Np}(^3\mathrm{He}, p3n)^{236}\mathrm{Pu}\,, \quad (9)$$

$$^{237}\mathrm{Np}(^3\mathrm{He}, \alpha)^{236\mathrm{m}}\mathrm{Np} \xrightarrow{\beta^-} {}^{236}\mathrm{Pu}\,. \quad (10)$$

In environmental samples the detection limits for ^{238,239,240}Pu will depend on the levels of these nuclides introduced with the ^{236}Pu tracer. The main impurity in the ^{236}Pu tracer produced is ^{238}Pu which is formed in the following reactions studied [3–16]:

$$^{236}\mathrm{U}(^3\mathrm{He}, n)^{238}\mathrm{Pu}\,, \quad (11)$$

$$^{236}\mathrm{U}(^3\mathrm{He}, p)^{238}\mathrm{Np} \xrightarrow{\beta^-} {}^{238}\mathrm{Pu}\,, \quad (12)$$

$$^{238}\mathrm{U}(p, n)^{238}\mathrm{Np} \xrightarrow{\beta^-} {}^{238}\mathrm{Pu}\,, \quad (13)$$

$$^{238}\mathrm{U}(^3\mathrm{He}, 3n + t\beta^-)^{238}\mathrm{Pu}\,, \quad (14)$$

$$^{237}\mathrm{Np}(^3\mathrm{He}, pn)^{238}\mathrm{Pu}\,, \quad (15)$$

$$^{237}\mathrm{Np}(^3\mathrm{He}, 2p)^{238}\mathrm{Np} \xrightarrow{\beta^-} {}^{238}\mathrm{Pu}\,. \quad (16)$$

Plutonium-237

The ^{237}Pu tracer ($T_{1/2} = 45.2 \pm 0.1$ d [2]) was mainly produced with the following reactions [3–16]:

$^{235}U(^{3}He, n)^{237}Pu$, (17)

$^{236}U(^{3}He, 2n)^{237}Pu$, (18)

$^{238}U(^{3}He, 4n)^{237}Pu$, (19)

$^{237}Np(p, n)^{237}Pu$, (20)

$^{237}Np(^{3}He, t)^{237}Pu$. (21)

^{237}Pu decays mainly by electron capture. It is used as a biomedical research tracer for studying the biological pathways of plutonium in the environment [1]. Therefore it should be free of alpha-emitters, such as ^{236}Pu or ^{238}Pu.

Neptunium-235

The nuclide ^{235}Np ($T_{1/2} = 396.1 \pm 1.2$ d [2]), which decays mainly by electron capture, is used as a biomedical research tracer for studying the biological pathways of neptunium in the environment [1]. The following ^{235}Np production reactions have been studied [4, 5, 13–16]:

$^{235}U(p, n)^{235}Np$, (22)

$^{236}U(p, 2n)^{235}Np$, (23)

$^{236}U(^{3}He, p3n)^{235}Np$, (24)

$^{238}U(p, 4n)^{235}Np$. (25)

So far there has been no systematic compilation of data for the production of ^{235}Np, ^{236}Pu and ^{237}Pu. Fragmentary data in narrow energy ranges are not convenient and enough for choosing the optimal production parameters. The present work gives a summary of the methods of cyclotron production of ^{235}Np, ^{236}Pu and ^{237}Pu. It is based on our results obtained in this research field during 1980–2001 and literature data. Since 1988 the work was carried out as a collaboration of Laboratory of Radiochemistry of University of Helsinki (HURL), Accelerator Laboratory of Åbo Akademi University (ÅAAL), Accelerator Laboratory of University of Jyväskylä (JYFL) and Laboratory of Nuclear Reactions and Nuclear Medicine of the V.G. Khlopin Radium Institute (RI). As a result of the performed studies the nuclear reactions with the so far highest known yields of ^{235}Np, ^{236}Pu and ^{237}Pu are determined. In addition new data of the cross section and yield of the $^{236}U(^{3}He, p3n)^{235}Np$ reaction in the energy range 42.4–60 MeV are given.

2. Experimental

2.1 Preparation and irradiation of uranium- and neptunium-oxide targets

Targets of uranium- and neptunium-oxide ($^{236,238}U_3O_8$ and $^{237}NpO_2$) have been prepared in the Radium Institute by multiple thermal decomposition of uranium or neptunium nitrate cellulose painted on 10 μm, 50 μm and 100 μm thick aluminum backings. The transformation was ascertained using alpha-spectrometry and direct weighing. The average thickness of the actinide layer was 60–120 $\mu g/cm^2$ for the thin targets and 1.5–2 mg/cm^2 for the thick targets. The uniformity of the target layers was checked with alpha spectrometry through a 1 mm diameter diaphragm for several points of the target. The thickness variations were within 3% for all types of targets used.

To vary projectile energy the degrading foil stack technique was used. The proton or ^{3}He-ion energy at each target was calculated from the range-energy relationships in aluminum and in the target material [17]. Irradiations of the targets were carried out with 18 MeV protons and 27 MeV $^{3}He^{2+}$-ions of the external beam of the Åbo Akademi 103 cm AVF cyclotron (MGC-20), and with 40 MeV protons, 40 MeV $^{3}He^{+}$-ions and 60 MeV $^{3}He^{2+}$-ions of the K-130 cyclotron of the University of Jyväskylä.

In the irradiations at the Åbo Akademi cyclotron, stacks of targets and degrading foils were mounted in a target holder which was cooled with water. The target surface was cooled with helium gas, streaming between the stack of targets and the 12.5 μm stainless steel foil against the cyclotron vacuum. The parts of the target holder formed a Faraday cup which was connected to a current integrator in order to determine the charge collected during the irradiation. The integrated charge of projectiles passing through the targets during the irradiation was determined with an accuracy of 5%.

A multiple target system (Fig. 1) was used for irradiation of targets at the K-130 cyclotron of the University of Jyväskylä. The target holders and energy degrading aluminum foils were mounted on two water cooled aluminum plates, respectively. The stacks of targets and foils were cooled with helium gas (pressure 0.8–0.9 bar), streaming between the target holders. The target system was electrically insulated from the cyclotron beam line. The integrated charge on the mutiple target system was measured with an ORTEC 439 digital current integrator. The integrated charge error was less than 1%. A collimator placed in front of the insulator ensured that the particle beam entering the chamber was going through the targets and the energy degrading foils. The beam intensity in different irradiations was varied from 1 to 10 μA and the irradiation time from 1 to 10 hours.

2.2 Chemical separation procedures

Different chemical separation procedures were used in our experimental works. According to our experience [14, 16] the use of TRU Resin (Eichrom Technologies Inc., USA) [18] provides a good separation between uranium, neptunium, plutonium and fission products (FP) with high yields of uranium, neptunium and plutonium in one separation cycle. The use of TRU Resin was restricted to the

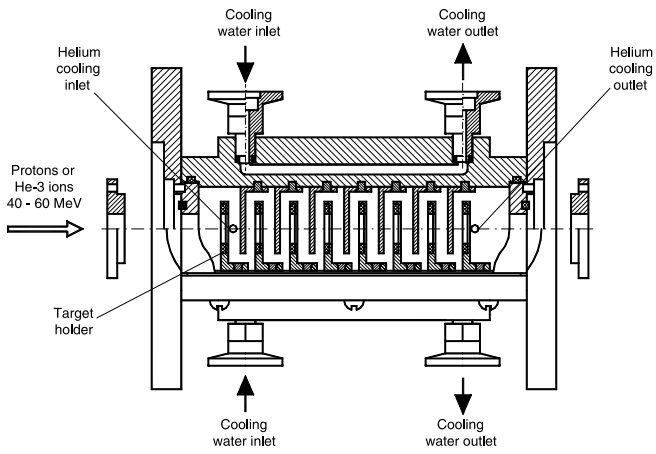

Fig. 1. Plot of multiple target system.

chemical procedures with thin uranium or neptunium targets weighing not more than four milligrams [18]. Anion exchange and extraction chromatography were used for the separation of plutonium or neptunium from thick uranium and neptunium targets.

2.2.1 Separation of plutonium from uranium target

Plutonium in nanogram quantities was isolated from the irradiated uranium target (0.7 g) with a chemical method including two separation steps: anion exchange and extraction chromatography in di-2-ethylhexylphosphoric acid (D2EHPA)/hydrochloric acid solution. These are schematically shown in Figs. 2a and 2b.

In the sample made from the stock solution of plutonium only alpha-peaks of plutonium isotopes were observed. The total yield of plutonium was about 80%.

2.2.2 Separation of plutonium from neptunium target

The sample solution of the neptunium target is prepared the same way as in the case of the uranium target (Sect. 2.2.1 above). In the sample solution plutonium is adjusted in tetravalence state and neptunium in hexa-valence state by the following procedure:

- 0.1 ml of ~ 50% $N_2H_4 \cdot HNO_3$ solution is added.
- The sample solution is heated at 95–98 °C on a hot plate.
- After complete destruction of $N_2H_4 \cdot HNO_3$ the sample solution is heated again for about 30 minutes until the colour of the solution changes quickly from green to brown. At this moment neptunium oxidizes to Np(VI)-valence form. The separation and purification of plutonium from this solution is carried out by the chemical method described above (Sect. 2.2.1).

2.2.3 Separation of neptunium from uranium target

The uranium target is dissolved in a mixture of concentrated nitric and hydrochloric acids by heating. This sample solution is evaporated to dryness and then treated with concentrated nitric acid. The dry residue is dissolved in 3 ml of 1.5 M nitric acid. For stabilisation of neptunium in Np(IV) and plutonium in Pu(III) 0.5 ml of 1 M Fe(II)-sulfaminate is added. Ascorbic acid of 0.8 M is then added dropwise into the solution until the blue colour does not appear any more.

The separation and purification of ^{235}Np from uranium and plutonium were made by using two chromatographic columns filled with teflon powder impregnated with methyl-three-octyl-ammonium nitrate (MTOA). The method is described in [15] and schematically shown in Fig. 3. The gamma activity of the fission products in the final solution was less than 0.01% and the alpha activity of the other nuclides less than 0.001% of that of ^{235}Np.

2.3 Cross sections

Plutonium-236

Cross sections of the nuclear reactions (1)–(16) were determined by measurement of alpha activity of ^{236}Pu and ^{238}Pu

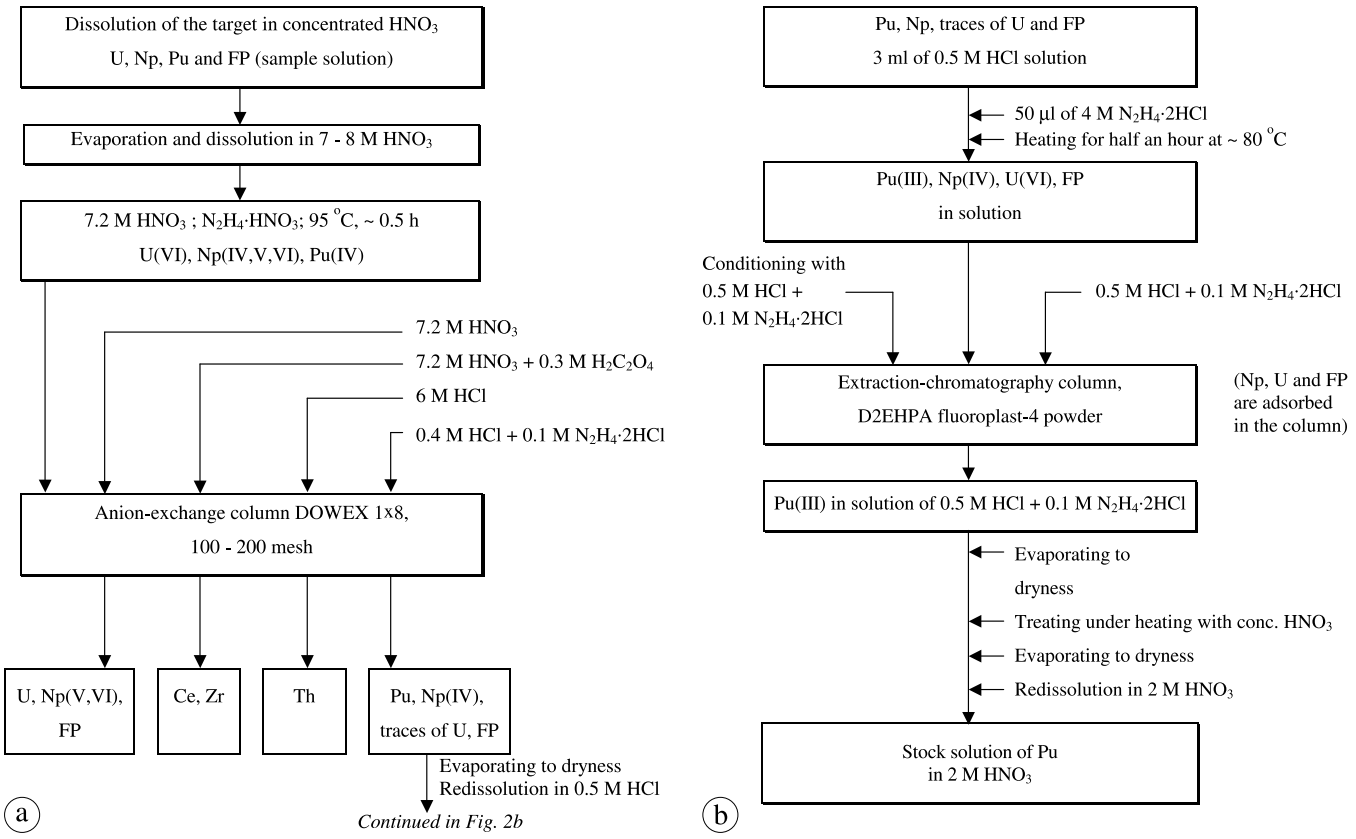

Fig. 2. Scheme of anion-exchange separation used in the separation procedure of plutonium from uranium and neptunium targets (FP = fission products, D2EHPA = di-2-ethylhexylphosphoric acid).

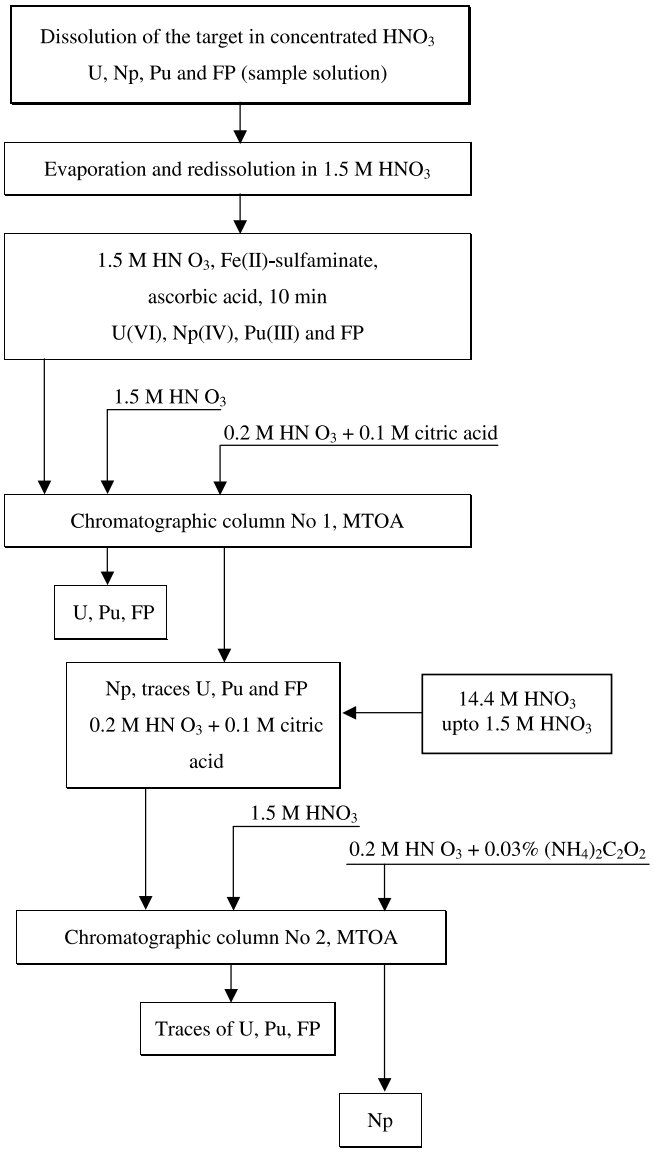

Fig. 3. Scheme of extraction-chromatography separation used in the separation procedure of neptunium from uranium target (MTOA = methyl-three-octhyl-ammonium nitrate).

in the irradiated thin targets with surface-barrier silicon detectors. The energy resolution of the alpha spectrometers was 20–35 keV at 5767 keV. Up to four alpha detectors were used simultaneously with outputs on an IBM PC computer.

A new data processing essentially decreased the errors of the final results. For all measurements the induced alpha activities were normalized with the target material alpha emission. The uncertainties of the detector surface area and the distance between the detector and the sample give the main contribution to the efficiency error. Determination of the ratio of induced and target material alpha activities at almost the same geometry of the detector system essentially decreases the error contributions of the detector surface area and the distance. This is important because the measurements were carried out with different detector assemblies in four laboratories. Another reason for the determination of the ratio of induced and target alpha activities was correction of the dead time which was caused by high activity of fission products. The dead time immediately after the end of bombardment (EOB) was up to 50% and it slowly decreased over the first days of measurements. Because the alpha counts of the target material were collected at the same high count rates as the induced ones, the dead time correction was automatically taken into account with this normalization procedure. In all cases the target was covered with a diaphragm with an accurately measured hole diameter (11–16 mm with error ±0.01 mm) in order to decrease the influence of uncertainties at the thin-target edges. Autoradiography was used to determine the shape and position of the beam spot on the irradiated targets. The diameter of the beam spots was 2–6 mm. The efficiency of alpha registration was calculated by the numerical integration of the exact analytical expression of the differential solid angle derived in [19] and/or by the Monte–Carlo method. The difference between registration efficiencies of alphas from the beam spot and the target material area defined by the diaphragm was less than 10% for all cases. The efficiency errors were 3%–6%, but as a result of the negative covariance coefficients the error of the efficiency ratio was less than 1% for all alpha detector assemblies.

For most studied targets ^{236}Pu and ^{238}Pu were formed both directly in nuclear reactions ((3), (7), (9), (11), (15)) and *via* β^--decay of ^{236m}Np ($T_{1/2} = 22.5$ h; reactions (4), (8), (10)) and ^{238}Np ($T_{1/2} = 2.117$ d; reactions (12), (16)), respectively. To determine separately the cross sections of these reactions the alpha-activity of each thin target was measured immediately after EOB for 4–5 days to observe the alpha-activity of ^{236}Pu and ^{238}Pu growing in the targets.

A typical time-dependence for the ratio of alpha-peak counts of ^{236}Pu to those of the target (for example ^{237}Np) is shown in Fig. 4. The time-dependence for each irradiated target was fitted with the formula given in [10]. Fig. 4 shows a good fit of data measured in the three laboratories HURL, JYFL and RI with different alpha detection systems. It demonstrates that the chosen measurement method and all corrections were done in the proper way. An example of error calculation can be found in [10]. The measurement of

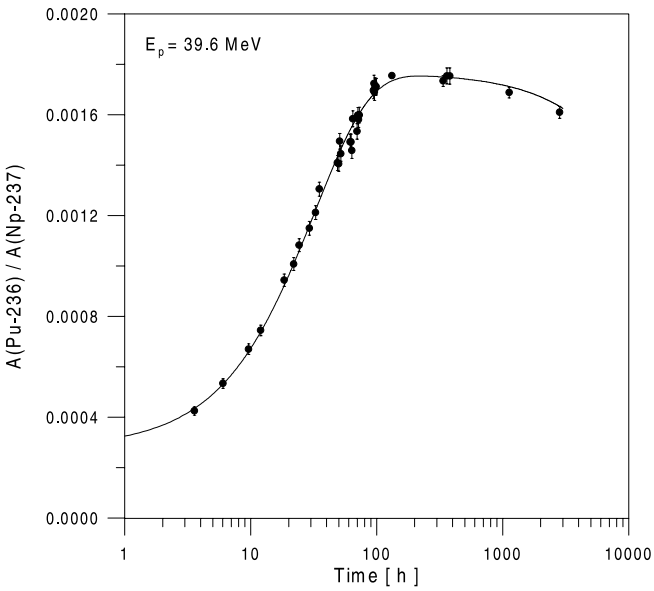

Fig. 4. Time-dependence of ratio of alpha-peak counts of ^{236}Pu to those of ^{237}Np for target irradiated at $E_p = 39.6$ MeV. Solid line is a fit with formula from [10].

the activity-ratio-time dependence gives information about growing of the impurities of ^{238}Pu in ^{236}Pu and (^{236}Pu + ^{238}Pu) in ^{237}Pu at the respective production time.

Plutonium-237 and neptunium-235

Cross sections of the reactions (17)–(25) were determined by measurement of X-ray activity of ^{235}Np and ^{237}Pu with Ge(Li)- and/or HPGe-detectors. The measurements of the ^{235}Np activities were done several times from 4 to 20 months after EOB in order to enable reliable identification on the basis of the half-life $T_{1/2} = 396.1$ d. In order to measure the gamma-activity of ^{237}Pu in the irradiated targets a chemical separation of plutonium from uranium or neptunium/protactinium and fission products had to be done. The separations of plutonium from uranium, neptunium and fission products were carried out according to the methods described above (Sects. 2.2.1 and 2.2.2). The HPGe-detector of HURL was permanently kept at stabilized high voltage in the conditioned underground measuring room for several years. Systematic measurements of the detector efficiency showed its high stability. Gamma spectra were measured with energy resolutions of 1.8 keV at 1332.5 keV. The error of the gamma detector efficiency was about 2%. The spectra were analyzed with the computer programs GANAAS (supplied by the IAEA) and *Micro Sampo* [20]. *Micro Sampo* is known as one of the best gamma spectroscopy codes providing good multiplet separation.

3. Results and discussion

Plutonium-236

The yields of ^{236}Pu and the impurities of ^{238}Pu from different nuclear reactions are presented in Table 1 and Fig. 5. The respective cross sections are shown in Fig. 6a–g. As can be seen in Table 1, the reaction ^{237}Np$(p, 2n + pn\beta^-)^{236}$Pu is the most productive one with a high radionuclidic purity of ^{236}Pu. The yield of this reaction is about three times higher than the earlier mainly used reaction ^{238}U$(p, 3n\beta^-)^{236}$Pu [25, 26] at the proton energy 40 MeV. The highest radionuclidic purity of ^{236}Pu can be obtained by the ^{236}U$(p, n\beta^-)^{236}$Pu reaction. Here it should be noticed that the impurity of ^{238}Pu for this reaction depends on the abundance of ^{238}U in the enriched ^{236}U target. In Table 1 the lower limit of ^{238}Pu impurity for reactions (7) and (8) is estimated from the ^{237}Np$(p, \gamma)^{238}$Pu reaction cross section of about 0.3 mbarn at its maximum [4, 21]. ^{237}Np always has contamination of ^{238}Pu. Therefore in the case of reactions (7)–(10) it is necessary to do careful chemical purification of neptunium from plutonium before the preparation of the target. For the reactions (1), (3)–(6), (9) and (10) the ratio of ^{238}Pu to ^{236}Pu can be decreased by choosing the time for the chemical separation as 3 to 4 days after EOB [22].

Plutonium-237

The yields of ^{237}Pu and the impurities of ^{236}Pu and ^{238}Pu in ^{237}Pu are presented in Table 2 and Fig. 5c–e. The respec-

Table 1. Thick target yields and radionuclidic purity of ^{236}Pu in various reactions.

Reaction	Projectile Energy [MeV]	Yield [kBq/μA h]	Radionuclidic Purity at EOB[b] [%]	Reference
^{235}U$(d, n\beta^-)^{236}$Pu	18, 2.5 mg/cm^2 [a]	0.022	0.08	23
^{235}U$(^3$He$, 2n)^{236}$Pu	27 0.36 mg [a]	∼ 0.1	–	3
^{235}U$(\alpha, 3n)^{236}$Pu	30–36, 5 mg/cm^2 [a]	0.035	2.0	24
^{236}U$(p, n\beta^-)^{236}$Pu	17.5	1.1	≥ 0.005[c]	4, 5
^{236}U$(^3$He$, t\beta^-)^{236}$Pu	42.4	2.4	0.8	6
	60	4.2	1.0	14
^{238}U$(p, 3n\beta^-)^{236}$Pu	17.5	1.8	3.6	7
	30	15	1.6	8
	40	16	2.0	25
	50	22	2.4	26
^{238}U$(^3$He$, 5n)^{236}$Pu	24.5	0.4	2300	3
	27	0.7	1200	Ibid.
^{237}Np$(\gamma, n\beta^-)^{236}$Pu	25 (e$^-$), 10 mg [a]	0.050	< 0.02	27
^{237}Np$(p, 2n + pn\beta^-)^{236}$Pu	17.5	3.0	≥ 0.06[d]	9
	40	53	≥ 0.025[d]	10
^{237}Np$(d, 3n)^{236}$Pu	15	1.0	40	28
	25	7.7	20	
^{237}Np$(^3$He$, p3n)^{236}$Pu	26.7	0.04	40	4, 5, 11
	60	4.4	10	12

a: The target thickness or total mass for thin target irradiations;
b: Activity impurity of ^{238}Pu in ^{236}Pu;
c: This value is an estimation of ^{238}Pu formation in reaction (13) for an abundance of about 0.1% ^{238}U in the ^{236}U target;
d: Lower limit of ^{238}Pu impurity is estimated from the suggestion that the ^{237}Np$(p, \gamma)^{238}$Pu reaction cross section is about 0.3 mbarn in its maximum.

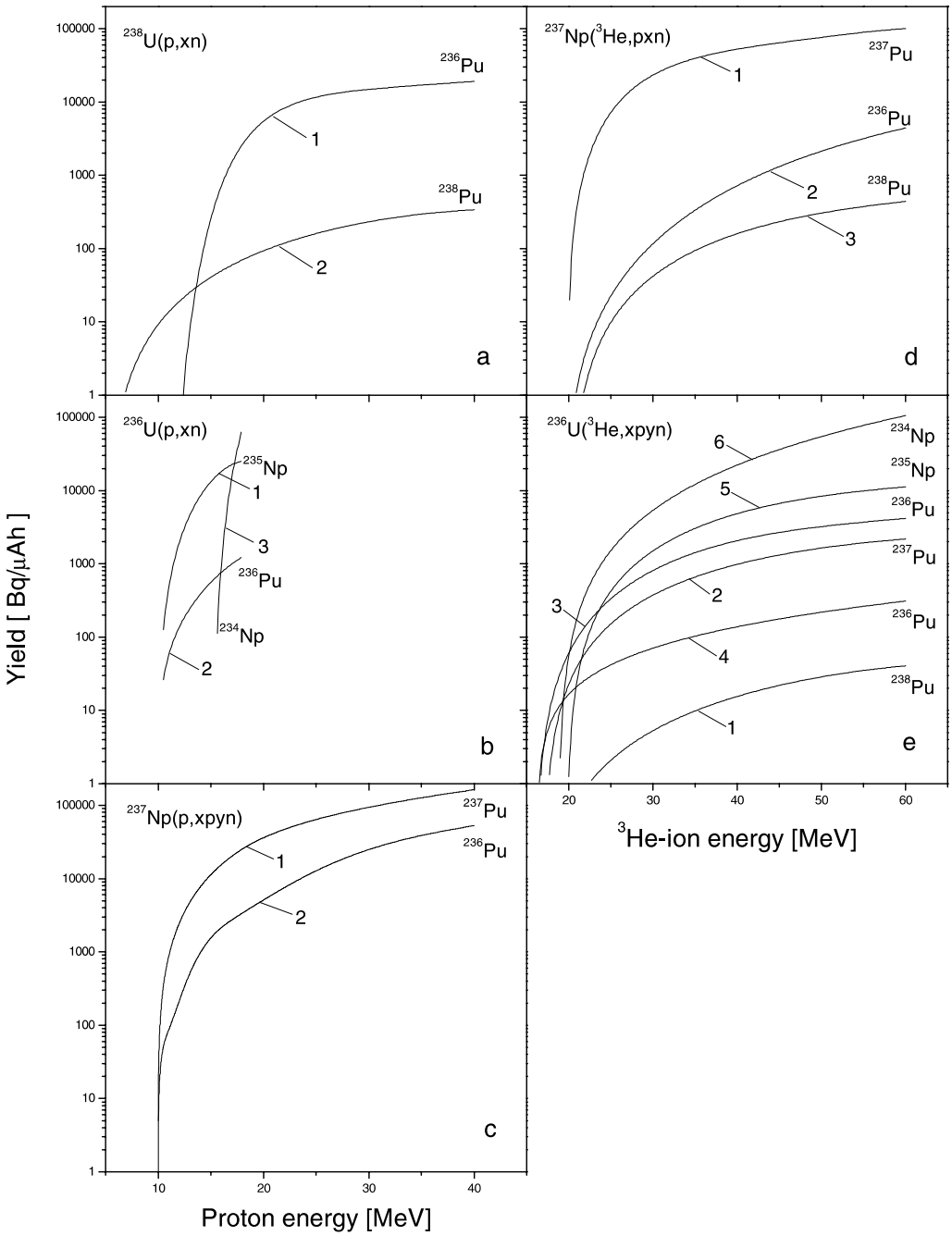

Fig. 5. Thick-target yields for following nuclides: (a) ^{236}Pu and ^{238}Pu in the proton-induced reactions on ^{238}U calculated on the basis of the cross sections measured in works [7, 8], (b) ^{235}Np (1), ^{236}Pu (2) and ^{234}Np (3) in the proton-induced reactions on ^{236}U calculated on the basis of the cross sections measured in works [4, 5], (c) ^{236}Pu and ^{237}Pu in proton-induced reactions on ^{237}Np [9, 10]: ^{237}Np$(p,n)^{237}$Pu (1) and ^{237}Np$(p,2n+pn\beta^-+d\beta^-)^{236}$Pu (2), (d) ^{237}Pu (1), ^{236}Pu (2) and ^{238}Pu (3) in the ^{3}He-induced reactions on ^{237}Np calculated on the basis of the cross sections measured in [4, 5, 11, 12], (e) ^{234}Np, ^{235}Np, ^{236}Pu, ^{237}Pu and ^{238}Pu in reactions induced by ^{3}He ions on ^{236}U [6, 14 and this work]: ^{236}U$(^3\mathrm{He}, n+p\beta^-)^{238}$Pu (1), ^{236}U$(^3\mathrm{He}, 2n)^{237}$Pu (2), ^{236}U$(^3\mathrm{He}, p2n\beta^-)^{236}$Pu (3), ^{236}U$(^3\mathrm{He}, 3n)^{236}$Pu (4) (upper limit), ^{236}U$(^3\mathrm{He}, p3n)^{235}$Np (5) and ^{236}U$(^3\mathrm{He}, p4n)^{234}$Np (6).

tive cross sections are shown in Fig. 6d,e,h. Formation of the impurities ^{236}Pu and ^{238}Pu was determined as described above. It can be seen in Table 2 that the most productive reaction is ^{237}Np$(p,n)^{237}$Pu. The yield of this reaction is about fifty times higher than the earlier mainly used reaction ^{235}U$(\alpha,2n)^{237}$Pu (Table 2). A good impurity ratio and a high yield are provided by the ^{237}Np$(^3\mathrm{He}, p2n)^{237}$Pu reaction. It should be noticed that the minimum value of the $(^{236}\mathrm{Pu}+{}^{238}\mathrm{Pu})/{}^{237}\mathrm{Pu}$ activity ratio can be obtained when the chemical purification of plutonium is done immediately after EOB.

Extensive technical efforts have been made to obtain ultra-pure ^{237}Pu by combining radiochemical and mass separation techniques [23, 24]. The use of the mass separation technique is necessary if ^{237}Pu is to be used for *in vivo* studies in humans. This technique was not used in the present work.

Neptunium-235

The yields of ^{235}Np and the impurities of ^{234}Np and/or ^{237}Np in ^{235}Np are presented in Table 3 and Fig. 5b,e. The re-

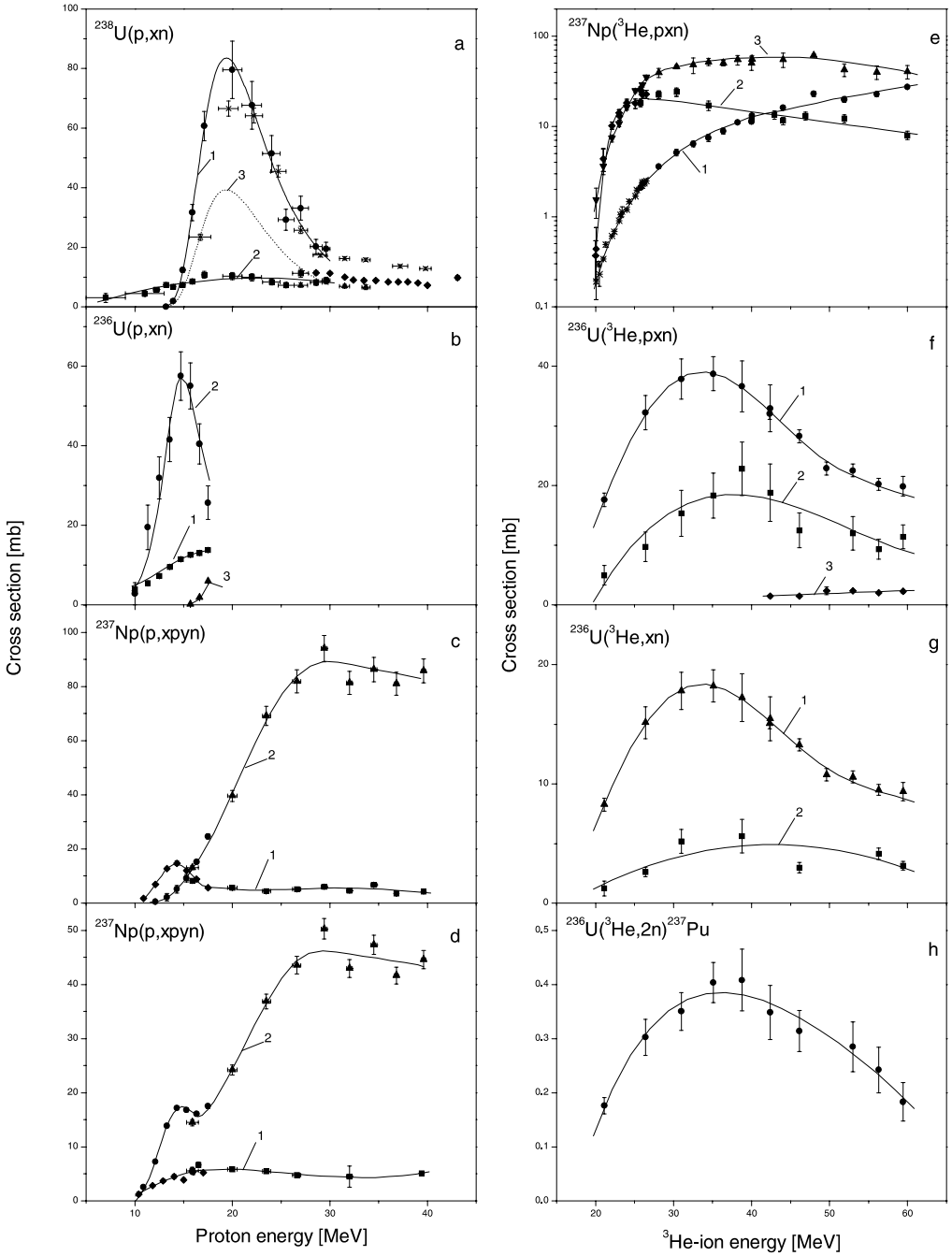

Fig. 6. Cross sections of the following reactions: a) ^{238}U$(p,3n)^{236m}$Np (1), ^{238}U$(p,n)^{238}$Np $\xrightarrow{\beta^-}$ ^{238}Pu (2) and ^{238}U$(p,3n)^{236m}$Np $\xrightarrow{\beta^-}$ ^{236}Pu (3) [7, 8]; solid and dashed lines are polynomial fits of the experimental cross sections; crosses and triangles are from [25], diamonds – from [33]; b) ^{236}U$(p,n)^{236m}$Np (1), ^{236}U$(p,2n)^{235}$Np (2) and ^{236}U$(p,3n)^{234}$Np (3) [4, 5]; solid lines are results of the theoretical calculations from [4]; c) ^{237}Np$(p,2n)^{236}$Pu (1) and ^{237}Np$(p,pn+d)^{236m}$Np (2); circular and rombic points were derived in work [9], $E_p = 10.9$–17.5 MeV; squared and triangular points are from [10], $E_p = 15.9$–39.6 MeV; solid lines are spline fits of experimentally determined cross sections; d) ^{237}Np$(p,n)^{237}$Pu (1) and ^{237}Np$(p,2n+pn\beta^-+d\beta^-)^{236}$Pu (2); circular and rombic points were derived in work [9], $E_p = 10.9$–17.5 MeV; squared and triangular points are from [10], $E_p = 15.9$–39.6 MeV; solid lines are spline fits of experimentally determined cross sections; e) ^{237}Np(^{3}He, $4n\varepsilon + p3n + 2p2n\beta^-)^{236}$Pu (1), ^{237}Np(^{3}He, $3n\varepsilon + p2n)^{237}$Pu (2) and ^{237}Np(^{3}He, $2n\varepsilon + pn + 2p\beta^-)^{238}$Pu (3) [4, 5, 11, 12]; solid lines are polynomial fits of the experimental cross sections; f) ^{236}U(^{3}He, $t)^{236m}$Np (1), ^{236}U(^{3}He, $p3n)^{235}$Np (2) and ^{236}U(^{3}He, $p4n)^{234}$Np (3) [6, 14 and this work]; solid lines are polynomial fits of the experimental cross sections; g) ^{236}U(^{3}He, $n+p\beta^-)^{238}$Pu (1) and ^{236}U(^{3}He, $p2n\beta^-)^{236}$Pu (2) [6, 14]; solid lines are polynomial fits of the experimental cross sections; h) ^{236}U(^{3}He, $2n)^{237}$Pu; solid line is polynomial fit of the experimental cross sections [6, 14].

spective cross sections are shown in Fig. 6b,f and Table 4. The cross sections, yields and impurities of ^{234}Np in ^{235}Np of the ^{236}U(^{3}He, $p3n)^{235}$Np reaction in the energy range 42.4–60 MeV presented in Tables 3 and 4 are determined with the same method as in [4, 5, 14]. It can be seen in Table 3 that the reaction ^{236}U$(p,2n)^{235}$Np is the most productive one (thick-target yield = 24 kBq/μA h) with the highest radionuclidic purity of ^{235}Np in the energy range up to 17.5 MeV. It could be suggested that the yield of ^{235}Np in the ^{236}U$(p,2n)^{235}$Np reaction would essentially increase at higher proton energy and become higher than in the ^{238}U$(p,4n)^{235}$Np reaction. The lower limit of ^{237}Np impurity for the reaction ^{236}U$(p,2n)^{235}$Np is estimated from the ^{236}U$(p,\gamma)^{237}$Np reaction cross section of about 0.3 mbarn at

Table 2. Thick target yields and radionuclidic purity of ^{237}Pu in various reactions.

Reaction	Projectile Energy [MeV]	Yield [kBq/μA h]	Radionuclidic Purity at EOB[b] [%]	Reference
^{235}U(^{3}He, n)^{237}Pu	26.5–27	∼ 0.6	15	3
^{235}U(α, $2n$)^{237}Pu	30	2	0.2	29
	25	1.5	0.03	30
	5 mg/cm^2[a]			
	27	3.1	0.8	24
	5 mg/cm^2[a]			
^{236}U(^{3}He, $2n$)^{237}Pu	42.4	1.1	14–220[c]	6
	60	2.2	< 14–190[c]	14
^{238}U(^{3}He, $4n$)^{237}Pu	24.5	0.2	4.0	3
	27	0.7	3.5	Ibid.
	38	0.6	2.7	31
	38	2	3.9	32
^{237}Np(p, n)^{237}Pu	17.5	23	9–13[c]	9
	40	160	5–33[c]	10
^{237}Np(d, $2n$)^{237}Pu	15	14	3–7[c]	28
	25	48	8–16[c]	
^{237}Np(^{3}He, $p2n$)^{237}Pu	26.7	12	0.2–0.5[c]	4, 5, 11
	60	100	1–5[c]	12

a: The target thickness for thin target irradiations;
b: Activity impurity of ^{236}Pu and ^{238}Pu in ^{237}Pu;
c: The minimum value of the activity impurity can be obtained when the chemical purification of plutonium is done immediately after EOB.

Table 3. Thick target yields and radionuclidic purity of ^{235}Np in various reactions.

Reaction	Projectile Energy [MeV]	Yield [kBq/μA h]	Radionuclidic Purity at EOB[b] [%]	Reference
^{235}U(p, n)^{235}Np	12–16	1–2	10^5[c]	13
^{235}U(d, $2n$)^{235}Np	18	0.35	No data	23
	2.5 mg/cm^2[a]			
	15	3	No data	34
	25	11	No data	
^{236}U(p, $2n$)^{235}Np	16.2	19	$< 10^{-7}$[b]; 10[c]	4, 5
	17.5	24	150[c]	ibid.
^{236}U(^{3}He, $p3n$)^{235}Np	42.4	5.8	500[c]	14
	60	11.2	940[c]	this work
^{238}U(p,4n)^{235}Np	27–30	15	2×10^{-4}[b]	15
	320 mg/cm^2[a]			
	50	41	2×10^{-5}[b]	26
	50	40	1.9×10^{-4}[b]	25

a: The target thickness for thin target irradiations;
b: Activity impurity of ^{237}Np in ^{235}Np;
c: Activity impurity of ^{234}Np in ^{235}Np. At the cooling time about three months after EOB this value will be about 10^6 times lower.

Table 4. Cross sections of the ^{236}U(^{3}He, $p4n$)^{234}Np and ^{236}U(^{3}He, $p3n$)^{235}Np reactions.

$E_{\text{He-3}}$ [MeV]	$\sigma_{\text{Np-234}}$ [mb]	$\sigma_{\text{Np-235}}$ [mb]	Reference
21.1 ± 0.4	–	4.9 ± 1.6	14
26.4 ± 0.3	–	9.7 ± 2.4	ibid.
31.0 ± 0.3	–	15.3 ± 3.7	ibid.
35.1 ± 0.2	–	18.3 ± 3.7	ibid.
38.8 ± 0.2	–	22.8 ± 4.3	ibid.
42.3 ± 0.2	1.41 ± 0.20		this work
42.4 ± 0.2		18.8 ± 4.7	14
46.3 ± 0.3	1.42 ± 0.16	12.5 ± 2.9	this work
49.6 ± 0.2	2.32 ± 0.65		this work
53.0 ± 0.2	2.30 ± 0.18	12.0 ± 2.8	this work
56.3 ± 0.2	2.03 ± 0.18	9.3 ± 1.7	this work
59.4 ± 0.2	2.22 ± 0.20	11.4 ± 2.0	this work

its maximum [4, 21]. For the reaction ^{238}U(p, $4n$)^{235}Np the ^{237}Np impurity was estimated according to the theoretically calculated cross section of the ^{238}U(p, $2n$)^{237}Np reaction [8].

4. Conclusion

The present work compiles data for cyclotron production of ^{235}Np, ^{236}Pu and ^{237}Pu. It is based on our results obtained in this research field during 1980–2001 and literature data. About twenty nuclear reactions were studied in wide energy ranges of the cyclotron projectiles. The nuclear reactions with the so far highest known yields of ^{235}Np, ^{236}Pu and ^{237}Pu tracers were determined: ^{236}U(p, $2n$)^{235}Np, ^{237}Np(p, $2n + pn\beta^-$)^{236}Pu and ^{237}Np(p, n)^{237}Pu, respectively. The highest radionuclidic purity of ^{235}Np, ^{236}Pu and

^{237}Pu tracers can be reached with the ^{236}U(p, $2n$)^{235}Np, ^{236}U(p, $n\beta^-$)^{236}Pu and ^{237}Np(^{3}He, t)^{237}Pu reactions, respectively. New cross sections and yield data of the ^{236}U(^{3}He, $p3n$)^{235}Np reaction in the energy range 42.4–60 MeV are given. The chemical procedures developed provide good separation and high yields of plutonium and neptunium tracers from the different target materials used.

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