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Transmutation of ^{129}I, ^{237}Np, ^{238}Pu, ^{239}Pu, and ^{241}Am using neutrons produced in target-blanket system 'Energy plus Transmutation' by relativistic protons

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Abstract. Target-blanket facility 'Energy + Transmutation' was irradiated by proton beam extracted from the Nuclotron Accelerator in Laboratory of High Energies of Joint Institute for Nuclear Research in Dubna, Russia. Neutrons generated by the spallation reactions of 0.7, 1.0, 1.5 and 2 GeV protons and lead target interact with subcritical uranium blanket. In the neutron field outside the blanket, radioactive iodine, neptunium, plutonium and americium samples were irradiated and transmutation reaction yields (residual nuclei production yields) have been determined using γ-spectroscopy. Neutron field's energy distribution has also been studied using a set of threshold detectors. Results of transmutation studies of ^{129}I, ^{237}Np, ^{238}Pu, ^{239}Pu and ^{241}Am are presented.

Keywords. Neutron production; radioactive targets; reaction rates; accelerator driven systems; transmutation.

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1. Introduction

Accelerator driven systems (ADS) are recent projects, which give new ideas about the problems related to the fuel cycle. Such systems can be used to incinerate long-lived fission products and minor actinides produced by conventional fission reactors. They could also prove to be useful to burn out large amount of plutonium from

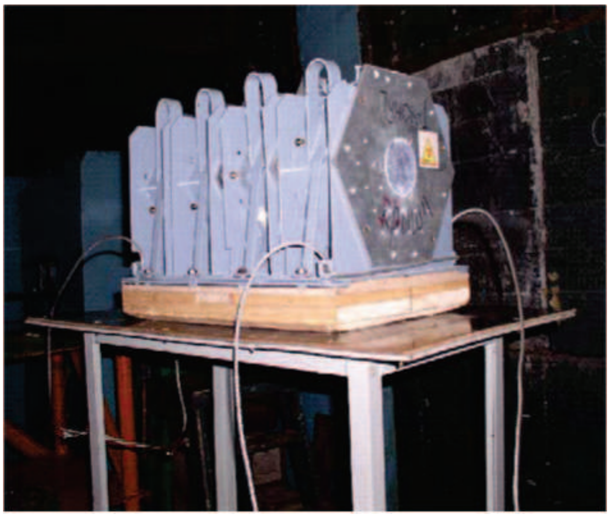

Figure 1. Photograph of the 'Energy + Transmutation' Pb/^{nat}U assembly outside the shielding before fixing the detectors.

nuclear weapons. Inside the system, within thick heavy metal target, spallation reactions generate high neutron fluxes and transmutation of selected isotopes takes place in the subcritical blanket. The neutron field is measured by means of different radiochemical sensors and activation detectors. In figure 1, a photograph of the 5-section assembly 'E + T with ^{nat}U' blanket and spallation solid target Pb has been shown without mounting of shielding has been used in the last few experiments during the years 2001–03.

2. Experimental set-up specification

'E + T' is a lead–uranium target blanket system [1], constructed from a lead target with 84 mm diameter, 480 mm length and with 43 kg total mass. The blanket contains four sections (figure 2). Each section is fuelled by 30 uranium rods in Al shell with 36 mm diameter, 104 mm length and 1.720 kg mass. Total mass of each section is 51.6 kg of natural uranium. So the whole blanket mass is 206.4 kg. Between each two sections are slits into which experimental instruments and detectors are inserted. The side from where the beam enters is covered with aluminum beam monitor and other activation or solid-state nuclear track detectors (SSNTD). The other detectors may lie on top of the blanket for different experiments.

A set of radioactive samples (^{129}I, ^{238}Pu, ^{239}Pu, ^{237}Np, ^{241}Am, and natural iodine ^{127}I) has been placed on top of the second section. Natural iodine is irradiated to subtract its influence in ^{129}I sample, which contains 17.1% of natural ^{127}I. Sample properties are listed in tables 1 and 2. The purity of other isotopes were 100% for ^{127}I, ^{239}Pu, and ^{241}Am. The ^{238}Pu targets contain 72.9% of ^{238}Pu, 16.8% of ^{239}Pu and the rest, a small mixture of ^{240,241,242}Pu. At the same place as RA-samples there is a set of threshold detectors (^{197}Au, ^{27}Al, ^{63}Cu, ^{58}Ni, ^{59}Co,

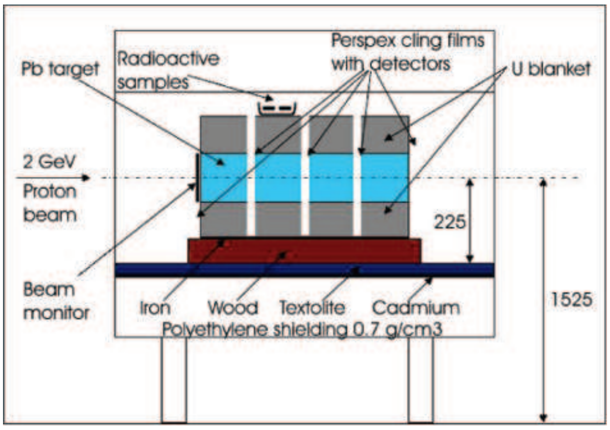

Figure 2. Simplified design of the 'Energy + Transmutation' assembly inside its shielding as used in the experiment (side view).

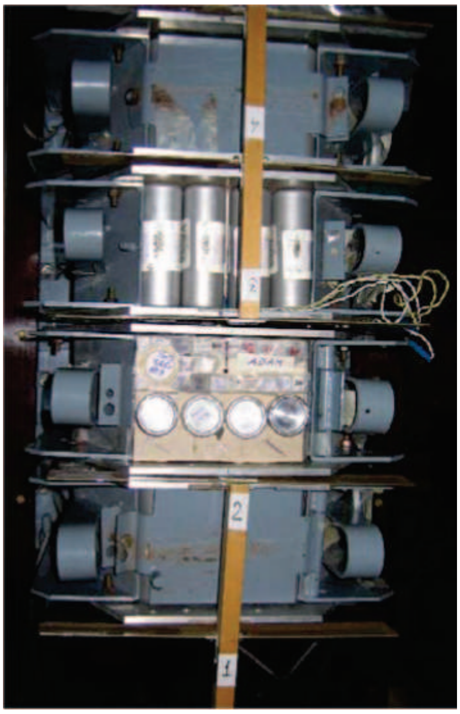

Figure 3. Top view of E + T set-up with radioactive samples (RA-samples) on it.

^{181}Ta, ^{115}In, ^{209}Bi) to determine neutron energy spectra at the place of RA-samples transmutation (see figure 3).

Facility 'E + T' was irradiated by the Nuclotron accelerator's proton beam with different energies. A list of basic experimental conditions is given in table 3. Total

Table 1. Samples properties for 0.7 and 1 GeV experiments.

	0.7 GeV		1 GeV	
Nuclei	Weight (g)	Activity (mCi)	Weight (g)	Activity (mCi)
^{127}I	1.439	stab.	1.439	stab.
^{129}I	0.591	0.10	0.591	0.10
^{237}Np	1.015	0.78	0.987	0.690
^{238}Pu	0.0517	879	0.0517	879
^{239}Pu	0.511	31.68	0.511	31.68

Table 2. Samples properties for 1.5 and 2 GeV experiment.

	1.5 GeV		2 GeV	
Nuclei	Weight (g)	Activity (mCi)	Weight (g)	Activity (mCi)
^{127}I	1.439	stab.	1.439	stab.
^{129}I	0.772	0.131	0.521	0.092
^{237}Np	1.011	0.745	1.011	0.710
^{238}Pu	–	–	0.0516	877
^{239}Pu	0.446	276	0.446	27.65
^{241}Am	–	–	0.186	638

Table 3. Basic experimental conditions.

Energy of protons (GeV)	Irradiation start	Time of irradiation (min)	Integral number of protons on Pb target
0.7	27th June '04	530.60	0.88(4) E+13
1.0	30th Nov. '03	423.40	2.93(13) E+13
1.5	11th Dec. '01	722.92	1.10(5) E+13
2.0	27th June '03	463.13	1.18(15) E+13

number of protons captured by the target were obtained from aluminum monitors and processed by radiochemical methods of γ-spectroscopy. For such monitoring purposes the reaction ^{27}Al$(p, 3pn)^{24}$Na was applied. The values of cross-sections [2] $\sigma(^{24}\text{Na})$=11.08(20) mb, 10.51(17) mb, 9.93(17) mb and 9.58(17) mb were used for the calculation of integral number of protons with energies 0.7, 1.0, 1.5 and 2.0 GeV, correspondingly. These values have been used in all the calculations.

Gamma-measurement has been performed by the high-purity germanium detectors (properties are given in table 4). Almost all measurements have been carried out with different filters of Pb, Cd and Cu. First measurements of the sample have been started after 6 h after the irradiation was stopped. Measurement duration

Table 4. Characteristics of HPGe detectors used for γ-measurement.

HPGe detector	CANBERRA GR1819	ORTEC GMX-23200	ORTEC GMX-20190-P
Relative efficiency	18.9%	27.7%	28.3%
Resolution ($E_\gamma = 1332$ keV)	1.78 keV	1.86 keV	1.80 keV
Amplifier	ORTEC 973	CANBERRA 2024	CANBERRA 2026
ADC	ORTEC 921 SPECTR. MASTER	ORTEC 919 SPECTR. MASTER	ORTEC 919 SPECTR. MASTER

Table 5. Number of γ-spectra measured during each experiment.

GeV	0.7	1	1.5	2
RA samples (+ ^{127}I)	55	80	31	76
Monitors ^{27}Al	12	16	12	13
Monitors ^{139}La	40	33	29	34
HPGe calibration	144	11	60	50
Threshold foils	106	–	208	60
Background and RA-samples before irradiation	11	18	16	11
Total number	368	158	356	244

times were from 0.5 to 48 h; all measurements have been usually done within 12 days. Observation possibility of residual nuclei is in the range of half an hour to one month in their half-lives. Two measurement stations have been used in Laboratory of Nuclear Problems (LNP) of JINR.

Processing of the measured data was performed by interactive mode of the Deimos code [3]; energy calibration, background γ-lines subtracting, single and double escape peaks subtracting, efficiency calibration, experimental half-lives determination etc., were made by a system of codes [4,5]. Determination of other minor corrections (coincidence summing correction, non-point geometry etc.) is in progress now. A correction for non-constant beam intensity has also been made.

Total number of measured spectra is given in table 5; thousands of γ-lines were analyzed. Identification was made according to energy, half-lives and intensity of peaks agreement and special attention to multiplex peaks has been emphasized.

Total number of neutrons passing through 1 cm^2 per one incident 2 GeV proton is $9.70 \cdot 10^{-3}$. The data are obtained from the threshold detectors [6], so this value is valid on the top surface of the second blanket section.

The values of B and R factors have been calculated (eqs (1)–(3)), and the R factors are presented here. Determination of neutron energy spectra and the total number of neutrons with all energies near the position of RA-samples are

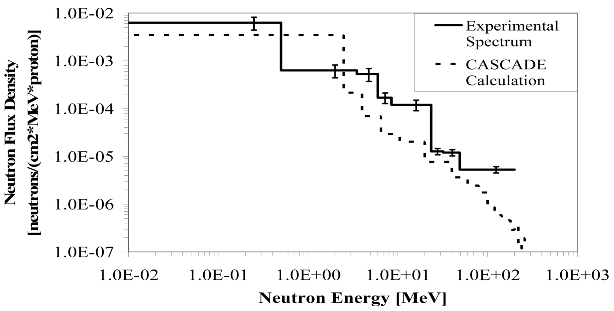

Figure 4. Neutron spectra results obtained from threshold detectors activation data for 2 GeV protons experiment.

investigated using activation foil kits and methods of neutron spectra unfolding. These activities are also still in progress.

$$B\left(\begin{matrix}A\\Z\end{matrix}\text{Res}\right) = \frac{\text{Number of produced nuclei } \begin{matrix}A\\Z\end{matrix}\text{Res}}{(1 \text{ g of target isotope}) \cdot (1 \text{ incident proton})}, \tag{1}$$

$$R\left(\begin{matrix}A\\Z\end{matrix}\text{Res}\right) = \frac{\text{Number of produced nuclei } \begin{matrix}A\\Z\end{matrix}\text{Res}}{(1 \text{ target isotope atom}) \cdot (1 \text{ incident proton})}, \tag{2}$$

$$R\left(\begin{matrix}A\\Z\end{matrix}\text{Res}\right) = B\left(\begin{matrix}A\\Z\end{matrix}\text{Res}\right) \cdot \frac{A_{\text{target}}}{N_A}. \tag{3}$$

3. Results

Residual nuclei observed in RA-samples using radiochemical and γ-spectroscopy methods are listed in tables 6–11.

After the irradiation of ^{129}I by secondary neutrons we observed four isotopes of iodine which are the products of the following reactions: $^{129}\text{I}(n,7n)^{123}\text{I}$, $^{129}\text{I}(n,6n)^{124}\text{I}$, $^{129}\text{I}(n,4n)^{126}\text{I}$ and $^{129}\text{I}(n,\gamma)^{130m,g}\text{I}$. The last reaction has the highest reaction rate compared to others. We measured cumulative yields from 84% decay of isomeric states ^{130m}I ($T_{1/2} = 9.0(1)$ min) and decay of ground state ^{130g}I ($T_{1/2} = 12.36(3)$ h). We started our measurements more than two hours after the end of irradiation and were still not able to see γ-rays 536.09(3) keV, whose intensity changed with half-life of 9.0 min. Then we determined the ratio $\sigma_m(n,\gamma)/\sigma_g(n,\gamma)$ for our spectrum of neutrons. For thermal neutron this ratio is 2.0(3) and for neutrons with $E_n < 100$ keV it is 1.58(14) (see ref. [3]). If we accept this last value we must increase our cumulative yield of ^{130m,g}I on 11% (see table 12).

Fission products and ^{238}Np were found in ^{237}Np targets after their irradiation by secondary neutrons. Reaction rate for fission can be established by means of

Table 6. Residual nuclei produced and observed in ^{238}Pu sample, R factor results.

		R [E27]		
Residual nuclei	$T_{1/2}$ (h)	1 GeV	1.5 GeV	2 GeV
Zr-97	16.91	3.2(3)	–	17.7(22)
Ru-105	4.44	–	–	15.0(13)
Sb-129	4.40	0.66(34)	–	–
I-132	2.295	3.24(39)	–	23.6(59)
I-133	20.8	2.28(54)	–	7.5(31)
Xe-135	9.14	2.58(52)	–	6.5(24)
TNoF[a]	–	20(7)	–	20(7)
TNoF[b]	–	23(8)	–	23(8)

[a] TNoF (total number of fission per one target nucleus) is calculated using cumulative yields for $E_n = 0.025$ eV.

[b] For $E_n = 500$ keV (400 keV for Pu-238).

[c] For $E_n = 14$ MeV.

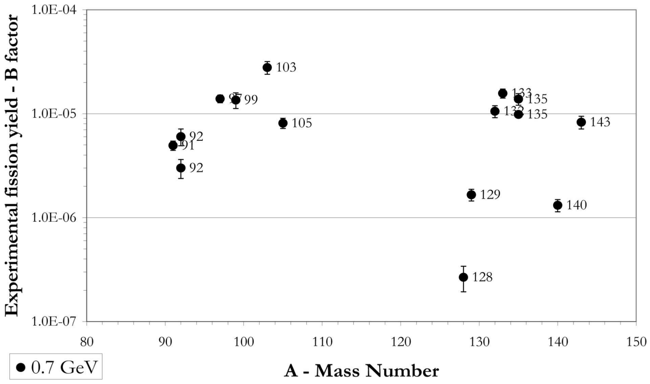

Figure 5. Experimental fission yields for ^{239}Pu, $E_p = 0.7$ GeV.

cumulative yields of fission products. These are known in literature for three energy of neutrons: thermal (0.025 eV), $E_n = 500$ keV, and $E_n = 14$ MeV. We try to establish effective energy of neutrons for the fission of ^{237}Np. But we observed just seven (experiment with $E_p = 1.5$ GeV) or less fission products which lie on (or nearby on) a maximum of A-distribution and from that we can suggest that effective energies of neutrons are between 500 keV and 14 MeV. We calculate R for fission (TNoF = total number of fission per one target nucleus) for different assumption about energy of neutrons. It is evident (tables 6–8) that TNoF does not differ by more than 20–30%. Ratio of reaction rate for (n, γ)/(fission) changes with energy of protons from 1.0 to 1.9 for ^{237}Np target.

Higher number of fission products was observed in Pu-239 sample. The same procedure as in Np-237 was adopted to determine the average/total number of

Table 7. Residual nuclei produced and observed in ^{239}Pu sample, R factor results.

	R [E27]			
Residual nuclei	0.7 GeV	1 GeV	1.5 GeV	2 GeV
Sr-91	1.96(8)	2.14(8)	4.52(18)	5.57(64)
Sr-92	1.19(19)	2.2(3)	4.11(64)	4.14(59)
Y-92	2.39(24)	5.91(58)	7.52(76)	12.12(12)
Zr-97	5.52(51)	3.24(30)	8.82(80)	11.35(10)
Mo-99	5.37(22)	5.38(22)	9.73(39)	13.71(18)
Ru-103	11.03(52)	5.26(25)	10.54(50)	19.57(37)
Ru-105	3.22(13)	4.9(2)	9.08(37)	13.76(25)
Sb-128	0.106(11)	0.129(16)	0.213(27)	0.37(6)
Sb-129	0.66(7)	1.19(12)	2.08(21)	3.2(9)
Te-132	4.18(17)	3.45(14)	4.59(19)	13.81(40)
I-131	–	2.8(1)	–	12.8(8)
I-132	–	2.67(63)	–	9.5(16)
I-133	6.23(21)	6.08(21)	11.04(38)	16.94(15)
I-135	3.90(10)	4.86(12)	8.78(22)	12.2(4)
Xe-135	5.48(62)	7.67(89)	11.5(13)	38.99(48)
Ce-143	3.29(13)	3.23(13)	7.47(30)	11.4(13)
Ba-140	0.52(16)	4.2(13)	7.4(22)	12.2(18)
La-140	–	0.588(70)	–	1.3(2)
TNoF[a])	52(8)	46(3)	52(2)	68(5)
TNoF[b])	55(7)	48(5)	54(3)	71(5)
TNoF[c])	62(8)	52(4)	60(3)	78(7)

[a] TNoF (total number of fission per one target nucleus) is calculated using cumulative yields for $E_n = 0.025$ eV.

[b] For $E_n = 500$ keV (400 keV for Pu-238).

[c] For $E_n = 14$ MeV.

fission per one target plutonium nucleus. Summary and library data comparison for Pu are presented as graphs in figures 5–8. Unfortunately, only 5 fission products of ^{238}Pu were observed due to small sample mass, different cross-sections and also the γ-spectral shape (see figures 9 and 10). Ratios $R(^{241}\mathrm{Am})/R(^{239}\mathrm{Pu})$ are given in the last column of table 11. For ^{238}Pu and ^{239}Pu these ratios are approximately equivalent for different residual nuclei. If we suppose that this conclusion is true also for ratios in table 11 then we can assume that

$$\sigma(^{241}\mathrm{Am}(n,f)) < 1.3\sigma(^{239}\mathrm{Pu}(n,f)).$$

4. Conclusions

All these results give good opportunity to make quality ADS benchmark. Coupled spallation target and fissionable blanket experiments are real challenges for codes

Table 8. Residual nuclei produced and observed in ^{237}Np sample (R factor results); fission and (n, γ) reactions results.

	R [E27]			
Residual nuclei	0.7 GeV	1 GeV	1.5 GeV	2 GeV
Sr-92	2.2(3)	–	1.50(19)	0.86(18)
Zr-97	0.80(24)	2.10(9)	2.12(7)	1.59(8)
Ru-105	–	–	1.97(28)	–
Sb-129	–	–	1.65(31)	1.11(32)
Te-132	0.56(12)	1.79(18)	1.77(28)	1.47(11)
I-133	0.83(40)	2.14(21)	2.01(24)	1.82(28)
I-135	1.52(22)	1.35(26)	2.36(28)	1.96(18)
TNoF[a]	42(8)	62(12)	85(13)	61(10)
TNoF[b]	42(8)	61(11)	80(11)	58(9)
TNoF[c]	53(7)	81(7)	93(7)	69(6)
Np-238	56.1(24)	151(5)	140(3)	133(3)
(n,γ)/fiss.[a]	1.32(25)	2.43(47)	1.65(25)	2.17(36)
(n,γ)/fiss.[b]	1.35(24)	2.46(44)	1.74(24)	2.27(34)
(n,γ)/fiss.[c]	1.06(13)	1.87(16)	1.51(12)	1.91(16)

[a] TNoF (total number of fission per one target nucleus) is calculated using cumulative yields for $E_n = 0.025$ eV.

[b] For $E_n = 500$ keV (400 keV for Pu-238).

[c] For $E_n = 14$ MeV.

Table 9. Residual nuclei produced and observed in ^{129}I sample, R factor results.

		R [E29]		
Residual nuclei	$T_{1/2}$	1 GeV	1.5 GeV	2 GeV
I-123	13.270 h	2.5(5)	–	3.9(7)
I-124	4.176 d	3.6(10)	7.7(7)	4.0(5)
I-126	13.110 d	10.8(5)	27.4(24)	22.5(44)
I-130	12.300 h	388(15)	696(36)	809(33)

like MCNPX calculation and these experimental results are great for validation. 'Energy + Transmutation' is a unique facility and its potential has been used to make as much experimental work as possible. Using the obtained data we calculated transmutation of the radioactive samples (see table 12), in which we have taken 10 mA current of accelerator and 30 days of irradiation. Experimental results presented are the first results of plutonium transmutation at 'Energy + Transmutation' target-blanket system. Comparable experimental results with deuterons and E + T are soon expected.

Table 10. Residual nuclei produced and observed in ^{127}I sample, R factor results.

		R [E29]		
Residual nuclei	$T_{1/2}$	1 GeV	1.5 GeV	2 GeV
In-111	2.805 h	0.103(26)	–	0.38(10)
Sb-118m	5.000 h	0.191(4)	–	0.65(150
Te-119	16.030 h	0.46(12)	–	1.31(27)
Te-119m	4.700 d	0.37(12)	–	1.03(12)
Sb-120m	5.760 d	0.215(6)	–	0.60(8)
I-120	1.350 h	1.90(4)	1.8(3)	–
I-121	2.120 h	1.09(4)	5.9(15)	3.31(23)
Te-121	16.780 d	1.39(21)	–	4.0(8)
I-123	13.270 h	3.92(26)	17.3(21)	13.0(10)
I-124	4.176 d	6.19(30)	29.5(21)	19.0(10)
I-126	13.110 d	25.1(12)	105(11)	81(4)
I-128	0.417 h	775(45)	–	–

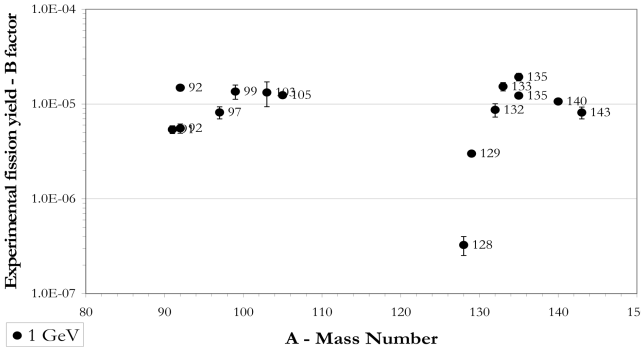

Figure 6. Experimental fission yields for ^{239}Pu, $E_p = 1.0$ GeV.

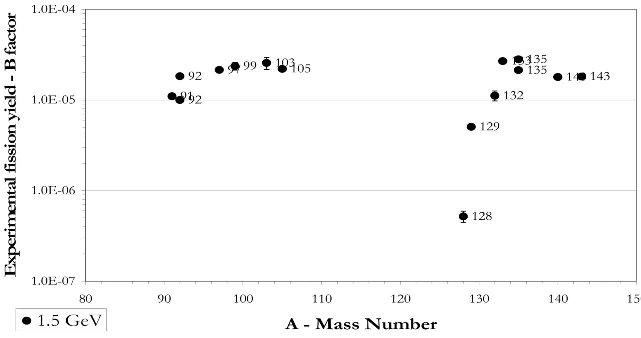

Figure 7. Experimental fission yields for ^{239}Pu, $E_p = 1.5$ GeV.

Table 11. Upper limits computed for residual nuclei expected as most probably produced in ^{241}Am sample, R factor estimation for 2 GeV experiment.

Residual nuclei	$T_{1/2}$	2 GeV R (Upper limit) [E27]	$B(^{241}\mathrm{Am})/B(^{239}\mathrm{Pu})$
Ru-103	39.260 d	<547	<36
Ru-105	4.440 h	<46.9	<3.1
Sb-129	4.400 h	<40.1	<12
Te-132	3.204 d	<50.0	<4.0
I-133	20.800 h	< 57.6	<2.7
I-135	6.570 h	< 39.1	<2.8
Ce-143	33.040 h	< 550	<28
Sr-91	9.630 h	< 37.0	<5.5
Sr-92	2.710 h	< 19.0	<3.7
Zr-97	16.900 h	< 24.9	<1.3
Mo-99	2.748 d	< 340	<2.1

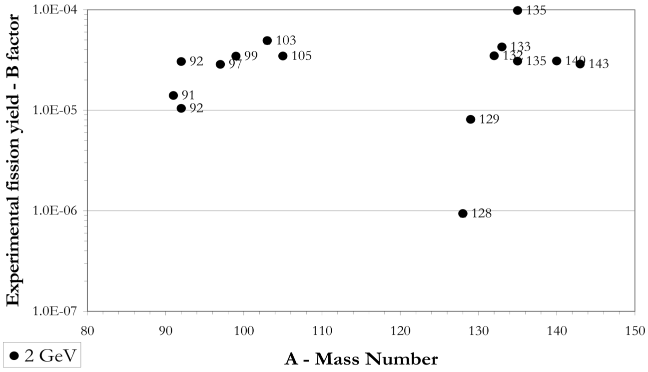

Figure 8. Experimental fission yields for ^{239}Pu, $E_p = 2.0$ GeV.

Table 12. Incineration of radioactive nuclei (%) with secondary neutrons produced in 'E + T' set-up using protons with various energies and hypothetical current 10 mA.

	Incineration 30 days (%)			
Radio-isotope	0.7 GeV	1.0 GeV	1.5 GeV	2.0 GeV
129-I	–	0.075	0.132	0.153
237-Np	1.64	3.53	3.66	3.18
238-Pu	–	0.34	–	0.34
239-Pu	0.91	0.79	0.90	1.17

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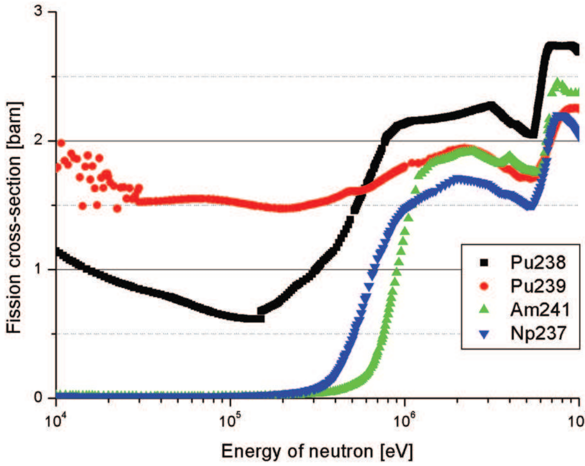

Figure 9. Neutron fission cross-section comparison for two isotopes of plutonium (taken from ENDF/B-VI.8 library).

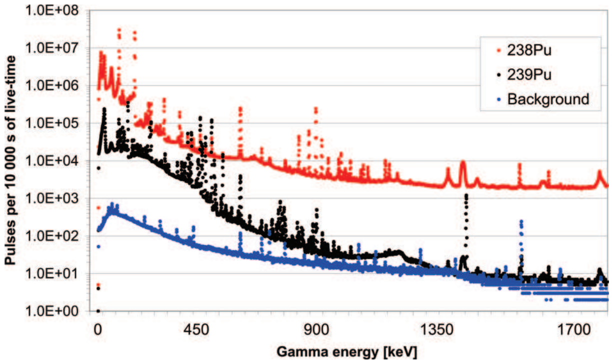

Figure 10. Gamma spectra comparison for two isotopes of plutonium.

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