



COMPLEX RADIOCHEMICAL AND ACTIVATION ANALYSIS OF LONG-LIFE NUCLEAR WASTE TRANSMUTATION IN FAST REACTORS AND IN THE BEAMS OF HIGH ENERGY ACCELERATORS

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Content :

1. ISTC #1372 project results:

- ***Stage I: Transmutation of MA in BN-350 reactor***
- ***Stage II: Experimental study of MA transmutation of MA in the “Energy + transmutation” facility - JINR (Dubna)***
- ***Project products***

2. Six new project-proposals



The ISTC project # 1372

Objectives:

experimental and theoretical research of transmutation effectiveness of the most important minor actinides and fission long-lived products in BN-350 reactor and in beams of high energy accelerators

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Stage I: Transmutation of MA in BN-350 reactor

- **Fabrication and calibration of samples**
- **In-reactor irradiation**
- **Post-irradiation radio-chemical analysis**
- **Computer and theory analysis**

FABRICATION OF SAMPLES FOR IN-REACTOR IRRADIATION

~~*The method of needle detectors*~~, irradiated in inter-pin space of standard fuel assemblies of BN-350 reactor, *was applied*.

Samples:

- *Capillaries*, made of stainless steel ($\varnothing 1.2 \times 0.1 \text{mm}$), filled with studied composition
- *Two samples* of each isotope were fabricated:
 - *One was intended for irradiation in BN-350,*
 - *The other one was kept as a sample of the input material.*
- *Isotopic compositions* of both initial and irradiated materials in capillaries were analyzed

RADIOCHEMICAL WORKS FOR EXTRACTION AND PURIFICATION OF INDIVIDUAL ISOTOPES

Starting materials were purified from radionuclide and radiochemical impurities

Samples and targets intended for irradiation in BN-350 reactor and in beams of “Energy + transmutation” facility were made

RADIOCHEMICAL WORKS FOR EXTRACTION AND PURIFICATION OF INDIVIDUAL ISOTOPES

Np-237: isotopic composition (% of mass):

Pu-238 concentration - about $1.0 \pm 0.3\%$ in alpha-radiation, or $4 \times 10^{-7}\%$ of mass

Pu-238:

Pu-239 – $79,92 \pm 0,16\%$;

Pu-239 – $16,75 \pm 0,14\%$;

Pu-240 – $2,87 \pm 0,06\%$;

Pu-241 – $0,35 \pm 0,04\%$;

Pu-242 – $0,11 \pm 0,01\%$.

Pu-240:

Pu-239 – $9,14 \pm 0,09\%$;

Pu-240 – $89,09 \pm 0,10\%$;

Pu-241 – $1,59 \pm 0,03\%$;

Pu-242 – $0,18 \pm 0,01\%$.

Pu-239:

Pu-239 – $94,25 \pm 0,06\%$;

Pu-240 – $5,59 \pm 0,06\%$;

Pu-241 – $0,11\%$;

Pu-242 – $0,05\%$.

Am241:

total concentration ~0.14 mass % vs. *Am* mass.

isotopic composition:

Cm-243+244:

Cm-244 - 86.32±0.18%

Cm-245 - 9.36±0.15%

Cm-246 - 4.32±0.08%

Am:

Am-241 - 39.40±0.20%

Am-243 - 60.60±0.02%

(continue)

The isotopic composition was determined after measuring of sample start material alpha-spectrum:

Cm-244+243	- 97.824%
Cm-245	- 0.09%
Am-243	- 0.111%
Pu-239+240	- 0.203%
Am-241+Pu-238	- 1.663%

U-235:

U-234 - $(0.96 \pm 0.04)\%$,
U-235 - $(89.89 \pm 0.25)\%$,
U-236 - $(0.34 \pm 0.03)\%$,
U-238 - $(8.79 \pm 0.20)\%$.

Iodine

(TRANSFORMED INTO SOLID SODIUM IODIDE):

I-129 - 82.9%,
I-127 (STABLE) - 17.1%.

Weights of the isotopes loaded into each sample:



Np-237: (122.6 ± 0.2) mg of NpO₂
(corresponds to 108.6 ± 0.2 mg of Np)

Pu-238: (125.1 ± 0.2) mg of PuO₂
(corresponds to 110.0 ± 0.2 mg of Pu)

Pu-240: (170±0.2) mg of PuO₂
(corresponds to 150.3±0.2 mg of Pu)

Am-241:

sample #1: (161.0 ± 0.2) mg of AmO₂
(corresponds to 142.0 ± 0.2 mg of Am-241)

sample #2 (165.0±0.2) mg of AmO₂
(corresponds to 146.0±0.2 mg of Am-241)

Cirium (Cm-243+244) – quantity in the sample was 0.067 g.

Burn-up monitor – Uranium dioxide (90% U-235),
(0.1149±0.0002) g of U.



Nuclide composition of the samples irradiated in the reactor cell #89 , mg

Sample	^{241}Am	^{237}Np	^{238}Pu
^{237}Np		108.6 ± 0.2	
^{238}Pu	0.0011 ± 0.0005	$(3.0 \pm 1.0) E-5$	87.8 ± 0.2
^{239}Pu	0.15 ± 0.07	0.0017 ± 0.0006	18.5 ± 0.2
^{240}Pu	0.05 ± 0.02	0.00054 ± 0.00018	3.180 ± 0.07
^{241}Pu	0.02 ± 0.01	-	0.39 ± 0.04
^{242}Pu	0.005 ± 0.002	-	0.123 ± 0.011
^{241}Am	141.78 ± 0.20	-	
Total, mg	142.0 ± 0.2	108.6 ± 0.2	110.0 ± 0.2



STUDY OF SAMPLES IRRADIATED IN BN-350 REACTOR

The reference samples were irradiated in three regular FA of the BN-350 reactor during 5 micro-campaigns.

FA cell #89: Samples with *U-235 (burn-up monitor)*, and *Np-237, Pu-238, and Am-241* isotopes

FA cell #110: Samples with *U-235 and Pu-240*

FA cell #243: Samples with *Pu-240, Am-241 and Cm-244*

Burn-up levels of the primary nuclides in the reference samples were measured, and ***isotopic compositions*** of the nuclides, formed after in-reactor irradiation, were determined.



Loading map of BN-350 reactor during the 46th micro campaign



Fuel subassembly (FSA) – low enrichment zone (LEZ), 17% enrichment

Fuel subassembly – moderate enrichment zone (MEZ), 21% enrichment

Fuel subassembly – high enrichment zone (HEZ), 26% enrichment

Fuel subassembly of the inner blanket

Fuel assembly of the outer blanket

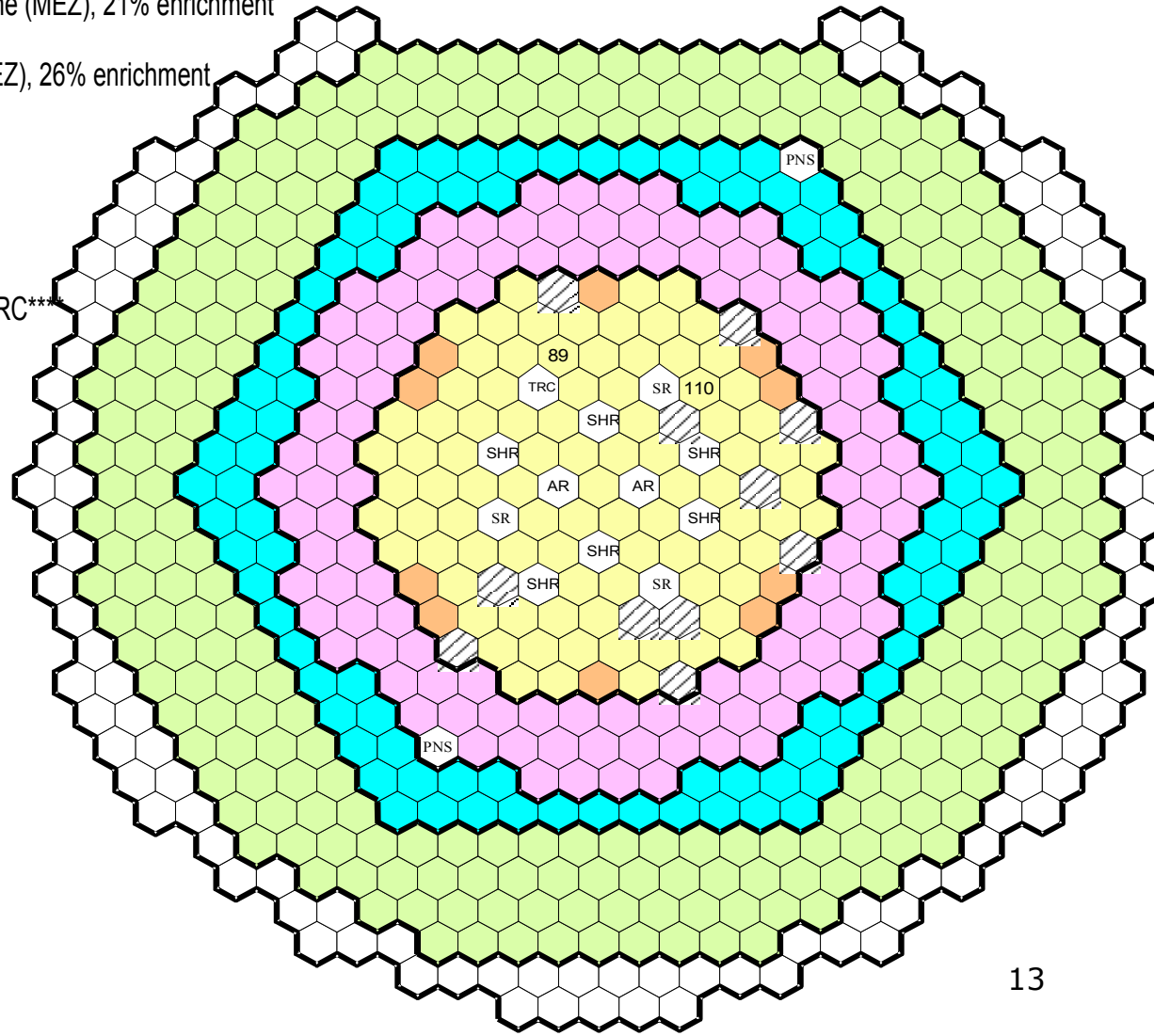
FSA in a shroud made of steel EP-450

Control rod assemblies: SR*, AR**, SHR***, TRC****

Photoneutron source assemblies

FSA with capillaries


FSA in the in-vessel storage





The integral neutron fluence in the samples

<i>FA Cell</i>	<i>Sample</i>	<i>Fluence, 10^{23} n/ cm²</i>
#89	Am41	2.229
	U-235 (monitor)	2.351
	Np-237	2.278
	Pu-238	2.012
#110	Pu-240	1.927
	U-235 (monitor)	2.262
#243	Cm – upper (1)	1.526
	Cm – lower (2)	1.968
	Am-241	2.264
	Pu-240	2.378



**On the basis of
the Russian system of neutron multi-
group data “BNAB-93” the neutron
fields were calculated over BN-350
reactor for all micro-campaigns,
taking into account changes of the
neutron fluxes (spectrum) due to the
fuel burn-up.**

The detailed calculations and theoretical study of transmutation of the main isotopes of the samples were made.



Example of *nucleon composition changes*, caused by both reactions with neutrons and radioactive decays (*calculations*):

<i>Main isotope</i>	<i>Burn-up of main isotope, %</i>	<i>Accumulation of secondary nuclides, %</i>	<i>Total burn-up of actinides, %</i>
<i>Np-237</i>	35	25	10
<i>Pu-238</i>	34	12	11
<i>Pu-240</i>	20	9	22
<i>Am-241</i>	35	25.5	9.5
<i>Cm-243+244</i>	27	17	10



Example of *nucleon composition changes for Np-237 sample after irradiation in BN-350 (radiochemical analysis and the calculations):*

<i>Isotope</i>	<i>Experiment</i>		<i>The calculated final quantity of nuclei</i>	<i>Calculation/ Experiment</i>
	<i>Initial quantity of nuclei</i>	<i>Final quantity of nuclei</i>		
<i>Cs137</i>	-	0.0130 ± 0.0004	0.0148	1.14 ± 0.03
<i>Ce144</i>	-	$(2.09 \pm 0.16) \cdot 10^{-5}$	$2.42 \cdot 10^{-5}$	1.16 ± 0.08
<i>Np-237</i>	2.760 ± 0.005	-	1.78	-
<i>Pu-236</i>	-	$(7.5 \pm 3.7) \cdot 10^{-6}$	$7.07 \cdot 10^{-6}$	0.94 ± 0.47
<i>Pu-238</i>	$(4.4 \pm 1.5) \cdot 10^{-5}$	0.583 ± 0.016	0.626	1.07 ± 0.03
<i>Pu-239</i>	$(1.4 \pm 0.5) \cdot 10^{-5}$	0.0319 ± 0.0010	0.0384	1.20 ± 0.04
<i>Pu-240</i>	-	0.00111 ± 0.00007	0.00153	1.38 ± 0.06



The sample nuclide compositions were calculated with using of modern versions of neutron data libraries:

- **BNAB-93 – Russia,**
- **BROND – Russia,**
- **JEF-3.3 - European Union,**
- **ENDF/B-6 (version 7) – USA, and**
- **JENDL-3.3 – Japan.**

The results demonstrate that discrepancies between calculations and experiment are less than uncertainty, caused by both cross-section errors, or by use of different neutron data libraries.



Stage II: Experimental study of MA transmutation of MA in the “Energy + transmutation” facility - JINR (Dubna).

- **Fabrication of radioactive targets for the proton accelerator**
- **Fabrication of specimens of track detectors**
- **Irradiation and control measurements**
- **Post-irradiation analysis**
- **Calculation and theory analysis**

Radioactive targets for experiments with the proton accelerator as well as specimens of track detectors were used in JINR (Dubna).


Targets:

**made of aluminum AD-1,
warranty for 5 years of operation
dimensions:**

- **diameter – 34mm,**
- **height – 14mm, thickness – 1 mm,**

The following radio-nuclides were included in the target compositions:

- ***I-129* – in form of NaI powder - 4 targets**
- ***Np-237* – in form of NpO_2 - 4 targets**
- ***Pu-238* – in form of PuO_2 - 3 targets**
- ***Pu-239* – in form of PuO_2 - 3 targets**
- ***Am-241* – in form of AmO_2 - 1 target**

At the “Energy + transmutation” facility (JINR)  *the secondary neutron spectra* of the facility at the incident proton energies of 1 GeV and 2 GeV were measured.

The neutron flux fluences in the target are:
0.00970 n/p×cm² (1 GeV) and
0.00304 n/p×cm² (2 GeV).

I-127, I-129, Pu-238, Pu-239, Np-237, and Am-241 targets were irradiated.

Proton integral fluxes :
(3.28+.14)10¹³ (1 GeV) and
(1.07+.14)10¹³ (2 GeV).



The parent nuclide burn-up,

measured in the secondary neutron field of U-Pb target, and referred to the proton beam current of 10 mA
(irradiation duration - 30 days and 730 days):

<i>Isotope</i>	<i>30 days</i>		<i>720 days</i>	
	<i>$E_p = 1$ GeV</i>	<i>$E_p = 2$ GeV</i>	<i>$E_p = 1$ GeV</i>	<i>$E_p = 2$ GeV</i>
<i>I-129</i>	0.06	0.152	1.44	3.65
<i>Np-237</i>	1.2	2.8	28.3	67
<i>Pu-238</i>	0.19	1.44	4.63	27.4
<i>Pu-239</i>	1.1	4.28	26.4	>100



Project products
(Prof. Yu. Shubin and his Lab):

The complete files of neutron data for Pu-240, Np-237, and Am-241 isotopes for incident neutron energies up to 150 MeV have been created on the basis of the evaluated neutron cross-sections.

The format and physical testing of the files was performed on the basis on CHEKER, FIZCON, and NJOY codes.

NEW PROJECT PROPOSALS:



***BASED ON RADIOCHEMICAL STUDIES BY
THE SPECIMEN METHOD WITH USE OF
ACCELERATORS AND REACTORS***

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Proposal # 1. Radiochemical preparation of compositions



for fabrication of targets and reference specimens/targets,

intended for irradiation in beams of accelerating complexes and reactors

Content:

- 1. Production, purification, analysis of initial isotopic material (minor actinides) by nuclear-physical and chemical-spectral methods and the material certification.***
- 2. Determination of optimum weight and size specifications of specimens/targets on the basis of the calculated data on fluxes and spectra in the places of location of the specimens/targets.***
- 3. Discussion of the research program and development of recommendations on arrangement of specimens/targets and their irradiation time.***
- 4. Development of Statements of Work (SOW) and Technical Specifications (TS) for the production of specimens/targets.***
- 5. Fabrication of specimens/targets from individual minor actinides and execution of all accompanying documents.***



The study of the irradiated specimens/targets using radiochemical and nuclear and physical methods.

Content:

- 1. Adjustment of methods and instruments for the study of specific irradiated specimens/targets.***
- 2. Development of working standards and performance of calibration measurements of activities of the irradiated and non-irradiated specimens/targets.***
- 3. Radiochemical analysis for measuring concentrations of fission fragments, isotopes of heavy nuclides and a residual concentration of a starting nuclide in the irradiated specimens/targets.***
- 4. Measurements of concentrations of an initial isotope and produced isotopes in the specimens/targets by nuclear and physical methods.***
- 5. Comparative analysis of the calculated and experimental data***

Proposal # 3: Development and fabrication of neutron sources for commissioning fast neutron reactors on the basis of current experience.

Proposal # 4:

Development of production technology and fabrication of neutron sources with required parameters for equipping instruments intended for control of fission material concentrations in various products.

Proposal # 5:

Studies of different irradiated fuel compositions by the reference specimen method.



Proposal # 6:



Developing a benchmark for testing nuclear data and calculation methods based on experiment on irradiation of actinides samples in the BN-350 reactor.

1. IPPE is ready to perform a work on developing a benchmark in a framework of an ISTC project.

1. Project duration: 1 year.

• Estimated project costs: 60,000 \$

Dr. Anatoly TSYBULIA, Director, Institute of Nuclear Reactors of IPPE, Obninsk –

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International benchmarks:

Setting test calculation models of experiment;

▪

Evaluation of uncertainties (models, FA) composition, geometry, temperatures, control rods position and the others;

▪

Experimental data;

▪

Detail description and evaluation of the benchmark experiments;

▪

Calculations with Russian and foreign codes and data;



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