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ON THE POSSIBILITIES OF EXPERIMENTS WITH ACTINIDE TRANSMUTATION

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Decontamination of nuclear wastes by transmutation of aktinides and fission products to stable or shot-lived nuclides is one of the urgent problem of the nuclear energy industry.

Different solutions to the problem are being studied. Accelerator-based installations and special reactor-burners of actinides on fast and thermal neutrons seem most. promising today.

Evaluation of transmuting concepts requires comprehensive experimental activities which can be combined into special programms of main scientific centres.

This report discusses possibilities to obtain the nuclear data neeled for evaluation of different targets and blankets for accelerator-based transmuting systems.

This concept,where the nuclear flux results from interaction of a proton beam with a thick target,requires a solution of some problems outside the scope of problems existing in conventional reactor physics and technology.

It is,first of all,because of nuclear reactions in the target, which result in production of neutrons in a broader energy range compared to the fission neutron spectra.

In accelerater-based transmuting system one has to follow the changes with time in the nuclide content of the target and accumulation of a long-lived nuclear reaction products.

Blankets transmuting systems can have different moderator materials(heavy water,molted salts) and ,because of,the neutron energy spectra of broad range also.

Measurements of general characteristics of systems for transmutation of Np,Am,Cm can be performd in functioning reactors with different neutron spectra. The need for such experiments is caused by poor experimental data on differential cross-sections for actinides available now.in these cases the libraries do not provide consistent calculated values of constants.

EXPERIMENTS WITH PROTON BEAMS.

Partially,the data on the total cross-section for actinides, production cross-section in reaction of protons with taget nuclei,neutron yields from the target per one proton of the beam,as well as,absolute rates of nuclear reactions in specific samples resulting from secondary interactions,could be obtained using activation techniques with semiconductor and scintillation detectors of high resolution.

Irradiation experiments in IHEP(Protvino) will be perfomed at different values of kinetic energy of proton beam -1200,1000, 800,103,73,37,24 MeV).
The reactions ^{27}Al(p,α)^{24}Na and ^{63}Cu(n,p)^{63}Zn will be used to monitoring of the primary beam intensity, The weight of the irradiated targets will up to 100 mg.

The study of interactions of protons with Pb-target requires that the target samples were enrichment to 95% of each of the four isotops -^{201,206,207,208}Pb ,

Correctness of the applied methods for calculation of the target behavior can be verified by measurements of space distribution of neutron flux.In such experiments the samples of high enrichment can be used ^{12}C(98,99%), ^{19}F(100%), ^{27}A1 (100%), 59CO(1009A), 63CU(100%) ^{65}Cu(98,7%), ^{64}Zn(99,4), ^{90}Nb(100%), ^{115}In(99,99%), ^{197}Au(100%).
The reactions of interest have threshold and thus rather informative for comparison with the calculation predictions.

Determination of the absolute neutron yields from targets under proton bombardment is performed using cylindrical targets surrounded by thick layer of moderator. The intensity of the primary beam is monitored. The distribution of the thermal neutrons on the space of layer is measured by conventional detectors. Absolute neutron flux is determined at several points. Numerical integrations of the measured relative distribution and normalisation to the absolute neutron flux at several points will result in determination of the number of neutron per one proton of the beam interacting with the target nuclei.

Calibration of detectors for measurement of relative and absolute values of thermal neutron flux will be done in thermal columns of functioning reactors.

EXPERIMENTAL STUDIES AND EVALUTION OF BLANKET PARAMETERS

The development and design of large scale blanket requires a lot experimental studies covering all aspects of the problems and may include experiments with full stalled subsystems.

Multiplying properties of the blanket can be successfully studied using critical systems of negligible(zero)power or subcritical systems with external neutron source.

In the Institute Theoretical and Experimental Physics(ITEP) a reactor of zero power MAKET has been recently commissioned (Shvedov O.V.,.. Preprint ITEP-167,1986). It provides unique capabilities for precise studies of the neutron physics,maintenance and safety problems of the heavy water lattices of various designs. MAKET has specific design features,different experimental techniques with automated data processing and model calculation.
The scope of studies can include determination of very specific parameters of lattice,as well as full scaled core model and blankets of heavy water nuclear installations of different type and purpose. Table 1 shows design parameters of MAKET which prove its unique capabilities for experimental studies:

- core vessel is 19 m^3.It can be early increased to 35 m^3.This will allow experiments with full stalled cores of research and powerfull reactors,as well,as with blankets of transmuting systems.
-the core vessel is not less 2 m distant from the floor and the walls of the building of its placing. It has a removable neutron absorption cover and tube supports,to make minimal the effects of neutron back-scattering from construction elements on critical dimensions of studied lattices.
-the shielding of the reactor allows to work at 1 kW power.
-the core vessel has containment preventing the contact of heavy water with environment of the working area,decreasing thus the losses and depletion of heavy water.
-continuous measurements of heavy water levels in the vessels of hydraulic system and safety systems prevent leakage of heavy water into the core vessel.The full heavy water balance can be determinatedat any instant with a precision of 15 kg of total quantity 10.000 kg

-the controlled temperature in the core can be changed. This makes possible studies of the temperature effects on reactivity.

-the experiments on MAKET,except regular systems,are also supported with standard and specific automated electronic mesurement systems.

The design feature and available equipment of MAKET provide possibilities for reactor neutron physics studies,as well as studies of maintenance parameters and safety problems of multiplying lattices of nuclear installations of different type.

The results of the studies can be used for validation of computer codes applied for model calculation and for the correction of the nuclear constants.

The studies of blankets of transmuting systems can be successfully carried out at MAKET equipped with external neutron source.

This subcritical system has a remarkable safety feature, an uncontrollable increase of power in such system is excluded.

It is planned to use the shield of the dismouted reactor HWR ITEP for installation intended for the blanket neutron physics studies.The installation will have external neutron source produced in Be or C target by a beam of accelerated protons or deutrons.

The neutron source will be surrounded by subcritical blanket on thermal or fast neutrons with fuel and transmuted materials as solids or molted salts.

The parameters of such system will be sufficient to perform the required studies.For instance,heavy water blankets with flux 10^{12} n cm^{-2} can be obtaind with deutron beam of 30 MeV kinetic energy and turret 1 mA.

It is worth noting that ITEP has its one know-how on construction of accelerator having required beam parameters.

SOME PROBLEMS OF ACTIVATION EXPERIMENTS

It is worth repeating that while evaluting available nuclear data for actinides,one has to remember,that blankets of transmuting sistems have different neutron spectra. This requires reliable nuclear data in the whole range of neutron spectrum.

In the following, the estimates of uncertainties of neutron cross-sections for actinides are obtained.The requirements for characteristics of samples and experimental apparatus are discussed.The products from (n,y)reactions on actinides have very long life-times.Therefore,activation experiments in the low neutron fluxes,such it is in MAKET,become difficult.Only the reactions, ^{237}Np(n,y) and ^{242}Pu(n,y) seem practical for activation technique studies on MAKET.The sample quantities for activation and measurements of y-spectra with Ge(Li) detector for those nucleides are 0,1-2 mg.

For measurements of 84 KeV y-radiation from ^{243}Pu the planar germanium detector of the 10 -10^{-3} -ef^{-1}iciency depending on its size will be used.The studies of transmuting fission reactions are accompanied by measurements of fission products of ^{143}Ce and ^{140}La on the yields of which there exist reliable nuclear data.

The measurements are advisable under the following conditions:

-sufficient neutron fluence for reliable detection of y-spectra of fission products in the presence of natural background from actinides.

-the material of samples must be pure enough to have an acceptable background from reaction on other isotopes end elements.

-there must be available precise nuclear data on fission product yields for the nucleides under study.

Let's apply the stated requirements to ^{237}NP ; ^{238,241,242}Pu, ^{241,242m,243}Am; ^{244,245}Cm isotopes :

Table 2 shows γ-ray intensities from ^{143}Ce and ^{140}La:

$A(^{143}Ce)/N_{nucl}$ and $A(^{140}La)/N_{nucl}$

as well as intensity of the background from sample irradiation for 30 hours in the flux of 10^{12}n cm^{-2}s.The intensity of y-rays from ^{143}Ce is an order of magnitude larger compared to ^{140}La.

Besides,the measurements efficiency with a germanium detector having a large volume,is 10 times higher in case of ^{143}Ce compared ^{140}La. Therefore,activation studies must be based on measurements of ^{143}Ce,first of all.

Such measurements,as the data in Table 2 show,are possible for ^{238,241,242}Pu; ^{241,242}Am; ^{245}Cm.

However,fission cross-section measurements for ^{237}Np and ^{243}Am are only possible with ^{140}La due to high background radiation near 300 KeV.

It seems that measurements of ^{242m}Am and ^{245}Cm cross-sections are beneficially to perform with ^{140}La because of necessity to suppress its low-energy γ-radiation by the filtres which significantly reduce the ^{143}Ce radiation. Fission cross-section of ^{244}Cm can't be measured by the irradiation of the sample in such a low neutron flux due to domination of the spontaneous fission product.

Table 3 shows the evalution results for necessary quantities of nuclides in samples for the event rate accumulation 1 imp/s in maximum.

It is worth to note that the production of highy enriched isotopes ^{238}Pu-99,6%; ^{241}Pu-99,998%; ^{242}Pu-99.96%; ^{243}Am-73,6%; ^{244}Am-99,949%; ^{244}Cm-99,9%; ^{245}Cm-99,998% is routine practise.

Isotopes are delivered as oxides,nitrides,chlorides in ampouls or thin layers on support of different thickness. In case of difficulties with a production of large samples for measurements on ^{140}La one can try also scintillator spectrometers. In this case the samples may have 20-50 times smaller amounts of material.

The data on neutron fission yields were reported in CINDA-90 and "Review of Fission product yields",NEANOC-300-V, 1990(Table 4). The most complete data on fission yields exist for ^{237}Np; ^{241,242}Pu ^{241}Am; ^{245}Cm.

In fact,there are two types of experiments to measure cumulative yields of ^{140}La ^{143}Ce. The first one is the mass-spectrometric measurements determining total cumulative yield of the isobaric chains: Y^{140}Ch, Y^{143}Ch. The second type of experiments based on γ-spectrometry determines a cumulative yield of specific products. As the yields of ^{140}La and ^{143}Ce are negligible,the cumulative yields of isobaric chains and ^{140}La, ^{143}Ce should practically be the same. This fact can be used a criterion for reliability of nuclear data.

The data for actinides well fissioned by thermal neutrons exist for the thermal neutron energy range only. The data on fission products from ^{238}pu; ^{243}Am; ^{244}Cm are not published, So,the available data about the yields of fission products don't make possible activation y-spectrometric method for measurements of fission cross-sections for ^{238}Pu; ^{243}Am; ^{244}Cm. Measurements of cross-sections for ^{241}Pu; ^{241}Am; ^{245}Cm may be performed with a 5% precision, whereas for ^{237}Np; ^{242}Pu; ^{242}Am-with 10%. The cross-section for reaction having no threshold can be determined using standarting thermal neutron fluxes. The method could be suggested for ^{238}Pu; ^{243}Am; ^{244}Cm.

REACTOR EXPERIMENTS -

One of the possibilities to obtain the needed nuclear data is buning up of actinides in the screen of fast reactor BN-350. The neutron spectrum is essentially changing with the distance from the core, as it changes in the blankets of transmuting electronuclear installations.

Therefore, the needed experimental data on reaction cross-sections can be obtained for characteristic intermediate spectra.

The experimental samples placed into the screen can be irradiated in fluence up to 10^{17}n cm^{-2}. The measurements of cross-section of ^{244}Cm become possible.

The transmutation studies based on burning up of actinides in the high(10^{16}n cm^{-2}s) flux neutron fields with soft spectrum, require the data on cross-section for ^{238}Np and ^{242g}Am (Nuclear Energy Generation Waste Transmutation using Accelerator Driven Intense Thermal Neutron Souse-C. D. Bouman et. al., LA-UR-91-91-2601). The presently available data on these nuclides are shown in Table 5.

The quantities of ^{238}Np and ^{242g}Am sufficient for the experiment can be obtained by irradiation of samples in reactor CM-2(НИИАР); ^{238}Np and ^{242}gAm can be used in experiments aiming at validation data on thermal cross-section and resonanse integrals for these actinides or getting data of better precision, The data can also be used for comprasion with the neutron code calculations for some spectra multiplied lattices.

One-day irradiation of ^{237}Np and ^{241}Am samples in CM-2 core can provide the following ratios:

$$\frac{^{238}\mathrm{Np}}{^{237}\mathrm{Np}} \cdot 2.5*10^{-2} \qquad \frac{^{242}\mathrm{Am}}{^{241}\mathrm{Am}} = 8.7*10^{-2}$$

(neutron flux in CM-2 core is $2*10^{15}$ n cm^{-2}s)

The accumulated quantity of ^{238}Np nuclei may be measured by y-spectrometry method using ^{238}Np y-radiation or ^{238}Pu a-radiation after cooling. Production of ^{242}gAm may be measured using ^{242}Cm a-radiation.

Correlations between the nuclide activities will be:

Neptunium sample: Americium sample:

$$\frac{A_a(^{238}\mathrm{Pu})}{A_a(^{237}\mathrm{Np})} = 600; \qquad \frac{A_\gamma(^{238}\mathrm{Np})}{A_\gamma(^{233}\mathrm{Pa})} = 10^{7}; \qquad \frac{A_\alpha(^{242}\mathrm{Am})}{A_a(^{241}\mathrm{Am})} = 85;$$

On the second stage the repeated irradiation of samples is performed and the fission quantity is measured by the track method relative to fissions in ^{235}U sample. If the repeated irradiation is performed in thermal spectrum with hardness, equals 0.2, for example, the following ratio of partial contributions to the total fission quantity is espected:

$$\frac{N_f(^{238}\mathrm{Np})}{N_f(^{237}\mathrm{Np})} = 50; \qquad \text{in cadmium screen - 6;}$$

$$\frac{N_f(^{242}\mathrm{Am})}{N_f(^{241}\mathrm{Am})} = 40; \qquad \text{in cadmium screen - 1.2;}$$

So, there exists the real possibility for experimental measurements of fission cross-section for ^{238}Np and ^{242}gAm with suffisiently high precision (better then 5%).

CONCLUSION

1. The experimental program going in ITEP is aimed at studies of neutron-physics parameters of targets of transmuting systems, irradiated by a proton beam of 24-1200 MeV kinetic energy. It is planned to obtain the estimates of the absolute yields, neutron spectra, changes in chemical composition of target under irradiation.
2. The experimental program will include measurements of the neutron cross-sections for actinides with samples irradiated in appropriate neutron fields of reactor БН-350. Similar experiments will be carried out in neutron fluxes with soft energy spectrum.
3. Validated and more precise nuclear data for ^{238}Np and ^{242g}Am will be obtained from experiments using the high flux reactor CM-2.
4. Analisis of existing data on fission products yields for actinides shows that available data is either not sufficient or does not provide the reqired precision of activation experiments. New measurements of fission products yields are planned.
5. Zero power reactor MAKET with external neutron source (^{252}Cf or neutron source of (d,t) reaction spectrum) will be used for neutron physics studies of different blanket designs.
6. In a pit of the dismounted reactor HWR ITEP a subcritical model of a blanket of transmuting sistem with external neutron source will be installed. The neutron flux will originate from a target (Be or C) irradiated by the beam of protons or deutrons (current 1 mA) of 10-30 MeV linear accelerator. The flux in the blanket equal to 10^{12} n cm^{-2} s^{-1} is expected.
7. The planned experimental programme with subcritical transmuting sistem will make it possible to verify the developed in ITEP computer codes for reactor calculations, such as TREC, a few groups code for geterogeneous lattices.

TABLE 1

BASIC PARAMETERS OF THE ZERO POWER REACTOR MAKET

1. Core vessel B-1	diameter	2.6 m
	height	3.6m
	volume	19 m^3
2. Safety vessel B-O	diameter	3.4m
	height	3.7 m
	volume	33 m^3
3. Conrol rods		
automatic control rods		1
compensation and safety rods		2
4. Automatic heavy water level meters in vessel B-1		2
measurement accurency of absolute level		1.5 mm
of relative level		0.5 mm
5. Limitater of heavy water level in vessel B-1		1
measurement accurancy of absolute level		1.5 mm
6. Signaller of heavy water level in vessel B-1		1
measurement accurancy Of absolute level		1.5 mm
7. Maximum rate of heavy water pumping into vessel B-1		0.7 l/s
8. Channals of control and safety system		11
9. Channels of radiation control system		
thermal neutrons		2
intermediate neutrons		2
fast neutrons		2
y-radiations		30
10. Maximal power of the reactor		1 kW

Table 2

Radiation intensity ^{143}Ce и ^{140}La after the sample irradiation

Nuclide	$\frac{143\ Ce}{N\ nucl}$	$\frac{140\ La}{N\ nucl}$	Main background gamma-lines		σ_f (barn)
			$\frac{A\phi}{N\ nucl}$	E_γ ,кeV	
237_{NP}	5,0X10-$^{;7}$	5,0X1O-'^{s}	4,0x10^{-1} '	311,9 (^{233}Pa)	0,33
238_{Pu}	3,0x10$^{-1\ 7}$	3,0X1O-'^{s}	2, 4x10$^{-1\ 5}$ l,0XI0-^{17}	152 201	0,2
241_{Pu}	4,0X10-^{1}6	4,0X1 o-^{i7}	4, 4X1 0-' ^{6} 9,0X10-'3	332,3 59,5 (241 $_{Am}$)	2,5
242_{Pu}	3, 0x10$^{-1\ 7}$	3, 0x10^{-1s}	4,5X10-^{18}	103	0,2
241 Am	4,0X10-'7	4,0X10-^{1s}	1, 8x10-' ^{i} 7, 6x10-i ^{7} 3,0x10$^{-1\ 9}$	5,95 322 955	0,3
$242m_{Am}$	1, 5x10$^{-1\ e}$	1 ,5X1O-'7	3,2X10-1 ^{2} 3,0x10$^{-1\ 4}$	117 163	1
243_{Am}	3,0x10$^{-1\ 7}$	3,0X1O-'8	4,3X10-1 ^{3} 3, 4x10^{-17}	277 (^{239}Np) 662	0,2
244_{Cm}	7,0x10$^{-1\ 7}$ spent f 9, 5x10^{-17}	7,0X1O-'8 spent f. 9,0x10$^{-1\ 7}$	3,0x10$^{-1\ 3}$ 1 ,2X10-14 8,0X10-i ^{6}	42,8 152,6 817-937	0,45
245_{Cm}	1, 5x10$^{-1\ 6}$	1 ,5x10^{-17}	2,4x10 $^{-,3}$	174	1

Table 3

Necessary nuclides quantities in sample,mg
Flux of fast neutrons 10^{10}n cm^{-2}s^{-1};time of irradiation 30 h

Monitor	^{143}Ce	140~a
Half-life	33,0 h	12,789 d - 40,22 h
Energy of detected gamma-ray Kev	293	1596
Emission probability %	42,9	95,4
^{237}Np	1,5	40
^{238}Pu	2	50
^{241}Pu	0,2	5
^{242}Pu	2,5	80
^{241}Am	1,5	40
^{242m}Am	0,4	10
^{243}Am	2,5	80
^{244}Cm	1	30
^{245}Cm	0,4	10

Table 4

Fission products yields for the transmutation
(CINDA-90;Review of Fission Product Yields. .NEANDC-300-V. 1990.)

Nuclid	Neutron spectrum	Fission products (%) ^{140}La	Fission products (%) ^{143}Ce	Note Average from
^{237}Np	fast	6,08 ±0,27	4,36 ±0,91	3 works
	fast.	(Ch)5,48 ±0,027		6 works.
			(Ch)4,71 ±0,02	3 works
		(^{140}Ba)5,899±0,290		4 works
^{241}Pu	fast	5,36 ±0,05	4,56 ±0,05	
^{242}Pu	fast	6,49 ±0,78		
		(^{140}Ba)6,7 ±0,7	3,95 ±0,3	
		(Ch)5,56 ±0,067	(Ch)4,598±0,032	2 works
^{241}Am	fast	5,32 ±0,21	3,57 ±0,11	5 works
		(Ch) 5,23 ±0,1	(Ch)3,96 ±0,7	
^{242m}Am	therm.	5,26 ±0,51	4,11 ±0,6	2 works
^{245}Cm	therm.	5,37 ±0,07		

Table 5

Fission cross-sections of ^{238}Np and 242gA$_m$
(C. D. Bouman, E. D. Arthur et al. LA-OP-91-91-2601)

Nuclide	238N$_p$	242gA$_m$
T 1/2, час	50,81	16
σ_f^{th} ,барн	2200±200	2900±100
I_f, барн	1454 ±150	300