A THEORETICAL STUDY OF HEAVY WATER - MINOR ACTINIDE BLANKETS FOR PROTON ACCELERATOR TRANSMUTATION

V. D. KAZARITSKY, P. P. BLAGOVOLIN, A. D. GALANIN

Institute of Theoretical and Experimental Physics
MOSCOW, 117259, RUSSIA

ABSTRACT

Nuclear fission is the only reaction leading to actinide transmutation. In an accelerator based assembly, employing heavy water solution circulation actinides are almost permanently exposed to a high neutron flux. The actinide supply from the spent fuel elements of nuclear plants is balanced with removing fission products from the transmutation system. In the process of incineration in the hybrid heavy water assembly there occurs a state when actinide concentrations do not change in time. It is in a steady state that the likely regimes of irradiation and transmutation performance of the system are studied. The fissions of the short-lived nuclide ^{238}Np dominate in high level neutron fluxes, while in routine level fluxes it is the fissions of ^{239}Pu that are dominant. It is shown that the transmutation performance depends on an acceptable energy deposition density rather than the type of the loaded actinide waste.

INTRODUCTION

There is a great demand for reducing the risk of radioactive waste disposal, that encourages research into transmutation of long lived radioactive nuclei to short-lived and stables ones. At present it is acknowledged that there is a need for transmuting the nuclei Np, Am, Cm, ^{99}Tc, ^{129}I [1,2].

Advances in chemistry of partitioning, progress in accelerator technology and other factors make it possible to hope for the appearance of an accelerator based plant transmuting the radioactive nuclides effectively and safety [3].

If an electronuclear plant is designed on the principles of an intensive neutron source, besides an linear accelerator of protons it should include a liquid Pb-target (or Pb-Bi eutectic) surrounded by heavy water. A simplified plant layout is shown in Fig 1.

The minor actinides transmutation in the heavy water blanket of the plant driven by an intermediate proton accelerator proved to have essential advantages [4 1. Following the terms of [4 1, the plant is here referred to as a ATW (Accelerator Transmutation of Waste]. In the ATW the linear accelerator, operating in a continuous regime, should be a 0.8 - 1.6 GeV-proton class accelerator enabling beam current as high as 200 - 300 mA. In the liquid-metal target the protons are converted to neutrons. For the 1.6 GeV-energy beam it is possible to produce about 50 neutrons whose mean energy is in the low MeV range. Moderated by heavy water the neutrons form a high flux of thermal neutrons in the range of 10^{16} n/cm^{2}sec. If it should be necessary the proton beam can be parted between several targets.

The only nuclear reaction that follows the actinide transmutation (transformation to the short-lived and stable nuclides) is a fission reaction. At a plant of the ATW-type the actinides will circulate through the neutron field as long as they are not fissioned. The actinides are fed into the ATW from reprocessing power reactor fuel in quantities same as those of fission products that should be removed. The weight of fission products removed per the year may serve as a measure of transmutation performance.

After some operation time there must be attained an equilibrium when the actinide concentrations (in per-unit) will not change in time. The equilibrium concentrations will be different from the input concentration. Some input concentrations formed by the waste of thermal reactor fuel reprocessing are listed in Tab.1.

It is assumed that U and Pu are extracted without losses and are returned to the fuel cycle of power reactors. The minor actinides are those parts of actinides that arrive for incineration.

This paper presents the results of an investigation of connection between equilibrium blanket characteristics and the flux level of the neutron source.

USED SYMBOLS

σ_T - thermal cross section;
RI - resonance integral;
γ - epithermal index;
ρ_i - nuclear density of i-th nuclide;
σ_f - fission cross section;
σ_c - capture cross section;
$T_{1/2}$ - half-life;
λ = $\ln 2 / T_{1/2}$ - decay constant;
Φ - density of thermal neutron flux, n/см²sec;
S - density of neutro sources;
i-1 - index of isotope with A_i-1;
n - index of β^+ -predecessor;
m - index of B^- -predecessor;
k - index of α -predecessor;
M^2 - migration area;
L^2 - diffusion length square;
τ - age (for D_2O - 125см²);
D - diffusion coefficient (for D_2O - 0,96);
Σ_a - total macroscopic cross section of neutron capture;
Σ_A - the same for actinides;
Σ_D - the same for heavy water;
Σ_H - the same for light water;
Σ_{FP} - the same for fission products;
ν - number of fission neutrons produced per fission;
$\bar{\sigma}_f$ - the average fission cross section of actinides in equilibrium;
$\bar{\sigma}_a$ - the same for capture;
ρ_A - integral actinide concentration;
Q - power density, W/см³;
I - current of incidental protons, A;
η - number of spallation neutrons produced per beam proton;

ONE-GROUP STUDY OF EQUILIBRIUM STATE

The variation of isotope composition for the actinide mixture may be determined by the system of differential equations:

$$\frac{d\rho_i}{dt} = -\sigma_{f_i}\rho_i\Phi - \sigma_{c_i}\rho_i - \lambda_i\rho_i + \sigma_{c_{i-1}}\rho_{i-1}\Phi + \lambda_n\rho_n + \lambda_m\rho_m + \lambda_k\rho_k + S_i \quad (1)$$

Granting that the system is linear, normalization is to be taken as it is convenient. For instance, we take ^{237}Np concentration as high as I.The used totality of the actinides and their transmutation ways are represented in Fig 2. In case of equilibrium the time derivative is considered to be zero. The system may be solved consequently by substitutions following in the arrangement of Fig.2. numbers. This procedure was realized by computer code RAWNOW. Calculation series were provided for cases of several neutron flux levels, epithermal index values and input actinide concentration.

The cross sections are calculated in one-group approximation by formula:

$$\sigma = \sigma_T + \gamma RI \quad (2)$$

Several loadings of different origin are studied: those that are waste of the spent VVER-1000-fuel with 3-year cooling; the same of PWR-fuel with 10-year cooling; mononuclide ^{237}Np loading,

If equilibrium mixture is represented by an effective nuclide, its resultant cross sections are represented by formula:

$$\sigma = \Sigma \sigma_i \rho_i / \Sigma \rho_i \quad (3)$$

The contributions of some nuclides presented on the scheme of Fig 2 are listed in Tab. 2. The resultant cross sections of the effective nuclide proved to depend on the neutron flux. For high fluxes C more than 10^{15} n/cm^{2}sec) the cross sections are increased following the neutron flux. The resultant fission cross sections as functions of the neutron flux and epithermal index are plotted in Fig.3-5. The list of nuclides whose fissions dominate is given in Tab. 3. For a high flux it is the fissions of short-lived nuclides (such as ^{238}Np) that will be essential.

ESTIMATES OF TRANSMUTATION RATES

One of the chief features of the ATW type plant as regards the target-blanket is that it may operate in a subcritical regime. The neutrons, originated in the Pb-target under the action of beam particles, can be multiplicated in the blanket at the expense of fission. The multiplication is equal to $1/(1-K_{eff})$. In the one-group theory K_{eff} is determined by fraction:

$$K_{eff} = K_{inf} / K_{geo} \qquad (4)$$

T-he divisor takes account of the neutron leakage and is equal to the value at which the finite size assembly may be critical. If the diffusion and age models are taken into account, the characteristics, related to a slab CH cm) problem, are defined as:

$$\begin{aligned} K_{geo} &= 1 + \alpha^2 M^2 \\ \alpha^2 &= (\pi / H)^2 \\ M^2 &= L^2 + \tau \\ L^2 &= D / \Sigma_a \end{aligned} \qquad (5)$$

Taking account for all the materials to capture neutrons the capture cross sections for the irradiation zone are :

$$\Sigma_a = \Sigma_a^A + \Sigma_a^{D2O} + \Sigma_a^{H2O} + \Sigma_a^{FP} \qquad (6)$$

It is assumed that the content of light water in heavy water is 0. 4%. The equality (6) may be written as:

$$\begin{aligned} \Sigma_a &= \Sigma_a^A (1 + q_{D2O} + q_{H2O} + q_{FP} \\ K_{inf} &= \nu \bar{\sigma}_f \rho_A / \Sigma_a \end{aligned} \qquad (7)$$

The concentration of fission products in the transmuted mixture may be kept on a constant level depending on the regeneration rates and the neutron flux. It is considered that the relative

concentration is:

$$'_{FP} = 0,1 \qquad (8)$$

It should be kept in mind that the fission products do not spoil the neutron balance very much. The concentrations and cross sections used for water equal to:

$$\sigma_a^{D_2O} , 1,14 \text{ mб}$$
$$\sigma_a^{H_2O} , 664 \text{ mб} \qquad (9)$$
$$\rho_{D_2O} = 0,0331$$
$$\rho_{H_2O} = 0,0331 .0,004$$

It is possible to express the relative capture of neutrons in water by:

$$'_{D20} .3,77 \text{ " } 10\text{-}' / C\ \bar{\sigma}_a^A \rho_A)$$
$$'_{H20} \text{ } 8,8 \cdot 10\text{-}' / C\ \bar{\sigma}_a^A{}_{PA)} \qquad (10)$$

The number of transmutations (fission reactions) per a spallation neutron, originated in the Pb-target may be represented by:

$$K_{fis} = \frac{1}{1 - K_{eff}} \cdot \frac{\bar{\sigma}_f \text{ PA}}{\Sigma_a} \qquad (11)$$

This function gives an idea of the efficiency of the use initial neutrons . The function depends on the actinide concentration and average cross sections, which in their turn depend on the neutron flux.

There is a link between the actinide concentration and the neutron flux. The link results from setting an upper limit on power density:

$$\bar{\sigma}_f \rho_A \Phi < Q \qquad (12)$$

The value of Q depends on the target-blanket design. For the estimates, represented here, it is important that the limitation exists, hence it follows that the neutron flux varies inversely with the actinide concentration:

$$\Phi \sim \frac{1}{\rho_A} \qquad (13)$$

Transmutation efficiencies as functions of the neutron flux are represented in Fig.6-7 for the power density range from 18 to 150 W/cm^3. A simplified calculation of an average neutron flux gives the value of about $10^{15} n/cm^2 sec$. In this case efficiency is about 0.5.

The 5-target plant performance is determined by beam current, equal to $0.3 \cdot 6.24 \cdot 10^{18} p/sec$ and hence by $55 \cdot 1.87 \cdot 10^{18} n/sec$ originated in 5 targets and by neutron efficiency, that is taken as 0.5. In this case the transmutation rate would equal $1 \cdot 10^{19}$ fissions/sec and the thermal power deposited in blanket would equal 1.5 GW, which is equivalent to the transmutation of the 500 kg actinides per year.

One-group average cross sections per fission product pair are shown in Fig. 8. In the used actinide mixture it is 80 barn. Using relationship (10) and calculating ρ_A from (12), one may find the fission product concentration ρ_{FP}:

$$\rho_{FP} = 2,5 \cdot 10^{-6} \text{ pair}/\text{cm}^3 \qquad (14)$$

in 10^{24}-units, It is necessary that the fission product removal per target have 0.004 g/sec, but solution removal rate would be about 15 l/h , Actinide composition of the solution drawn for regeneration depends on the neutron flux. The calculated composition plotted against the neutron flux is shown in Fig. 9-12. The total actinide concentration of the D_2O solution as a function of the neutron flux is given in Fig. 13. In consequence, the transmutation plant may have the following characteristics:

energy of beam protons	1.6 GeV
mean proton current	300 mA
beam power	480 MW
accelerator efficiency	50 %
accelerator power supply	960 MW
target number per accelerator	5
heat power per target	96 MW
heat power per blanket	300 MW
actinide transmutation rate	500 Kg/year

the same in VVER-1000-exchange units	17
drawing the solution for recovery	75 l/hour
steam generation	2500 t/hour

CONCLUSION

The above analysis of plant performance is qualitative rather than quantitative. The general properties of a hybrid system based on D_2O-dissolved actinides were subjected to analysis.

The actinide transmutation rate is shown to depend on allowable power density in a water blanket. In an equilibrium regime performance does not largely depend on the input actinide composition (reactor type and cooling time). The neutron flux largely determines both the performance and the equilibrium actinide composition.

The average cross sections, calculated per one fissioned nucleus, increases rapidly with the neutron flux. In case of sufficiently large power density C50-100 W/cm^3), the transmutation performance reaches its peak at flux of 10"- $10^{16} n/cm^2 sec$. For small power densities the performance is comparatively flat.

The calculation shows that there may exist a regime of operating when the plant would incinerate the minor actinide waste of no less than ten 1000 MW-power stations.

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Table 1.

Minor actinide waste nuclei from power reactors
(in relative units)

Nuclides	VVER (3 year cooling]	LWR (10 year cooling)
^{241}Am	0,46	0,477
^{237}Np	1,0	0,422
^{242m}Am	1,2E-3	6, 0E-4
^{242}Cm	3, 1E-4	0
^{243}Cm	8, lE-4	3, 18E-4
^{243}Am	0,224	0,085
^{244}Cm	0,049	0,016
^{245}Cm	9, 4E-3	8, 7E-4
^{246}Cm	2, 4E-4	0

Table 2.

Fission and capture rates of the nuclei in equilibrium (neutron flux of 10^{16} n/cm²sec)

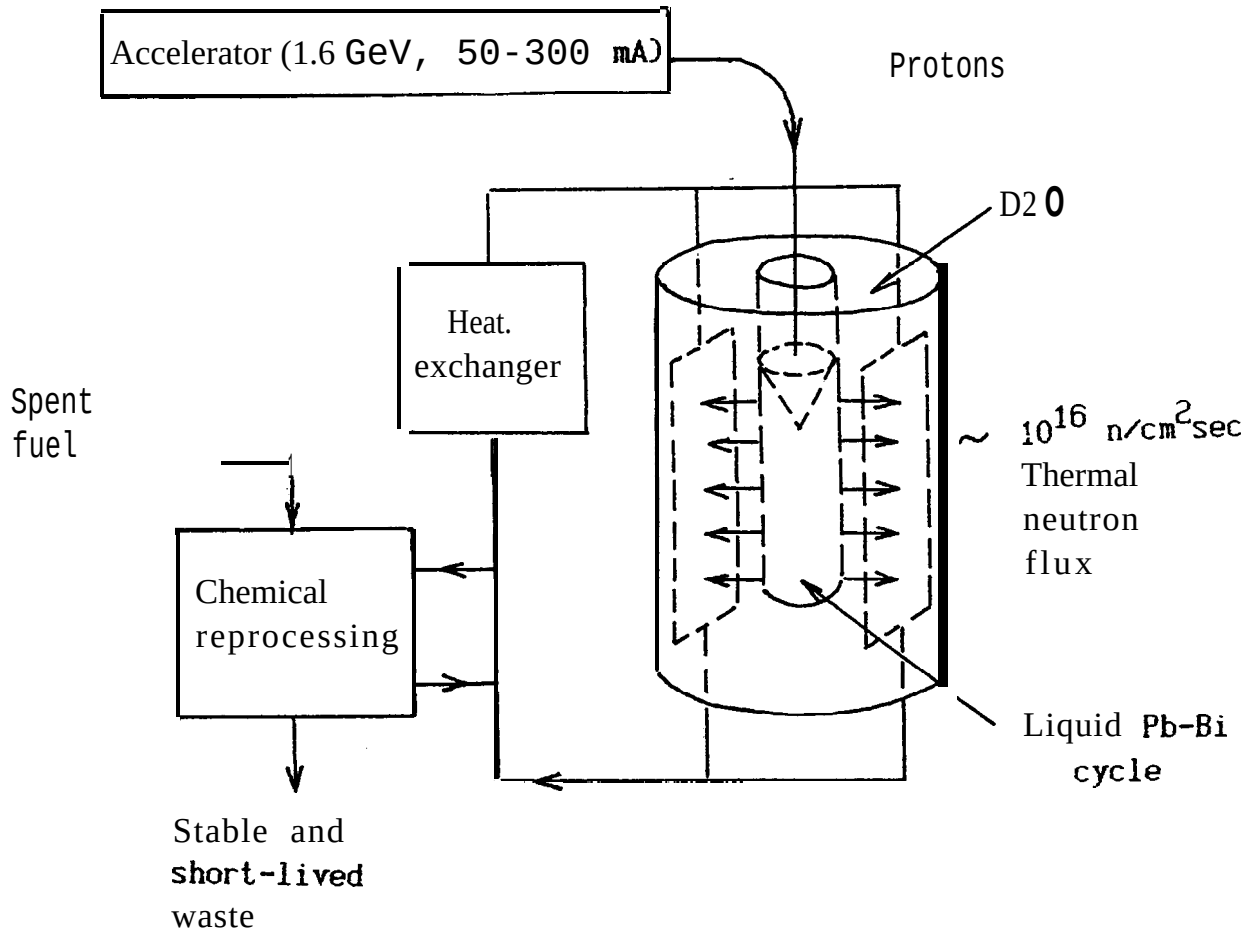
Nuclides	$\rho \sigma_f$, %	$\rho \sigma_c$, %
^{241}Am		17,5
^{237}Np		38,2
^{242m}Am	2,2	
^{242g}Am	15,2 "	
^{238}Np	48	
^{242}Cm	1,2	3,4
^{243}Cm	4,3	
^{238}Pu		6
^{239}Pu	7	2
^{240}Pu		2
^{241}Pu	2,3	
^{242}Pu		1,5
^{243}Am		10
^{244}Cm		11,7
^{245}Cm	15,5	1,7
^{246}Cm		1,6
^{247}Cm	1,4	

Table 3.

"The relative fission rates as neutron flux functions (dominant nuclei)

Flux $n/cm^2 sec$	Nuclides	Fission rates	Concentration relative to ^{237}Np
10^{16}	^{238}Np	0,481	0,0917
	^{245}Cm	0,155	0,0289
	^{242m}Am	0,152	0,0295
	^{239}Pu	0,070	0,0334
10^{15}	^{239}Pu	0,317	0,1492
	^{238}Np	0,210	0,0396
	^{245}Cm	0,203	0,0375
	^{241}Pu	0,098	0,0370
10^{14}	^{239}Pu	0,492	0,2305
	^{245}Cm	0,220	0,0404
	^{241}Pu	0,152	0,0572
	^{238}Np	0,032-	0,0059

Fig. 1. Operating scheme of plant for transmuting radioactive waste of power reactors



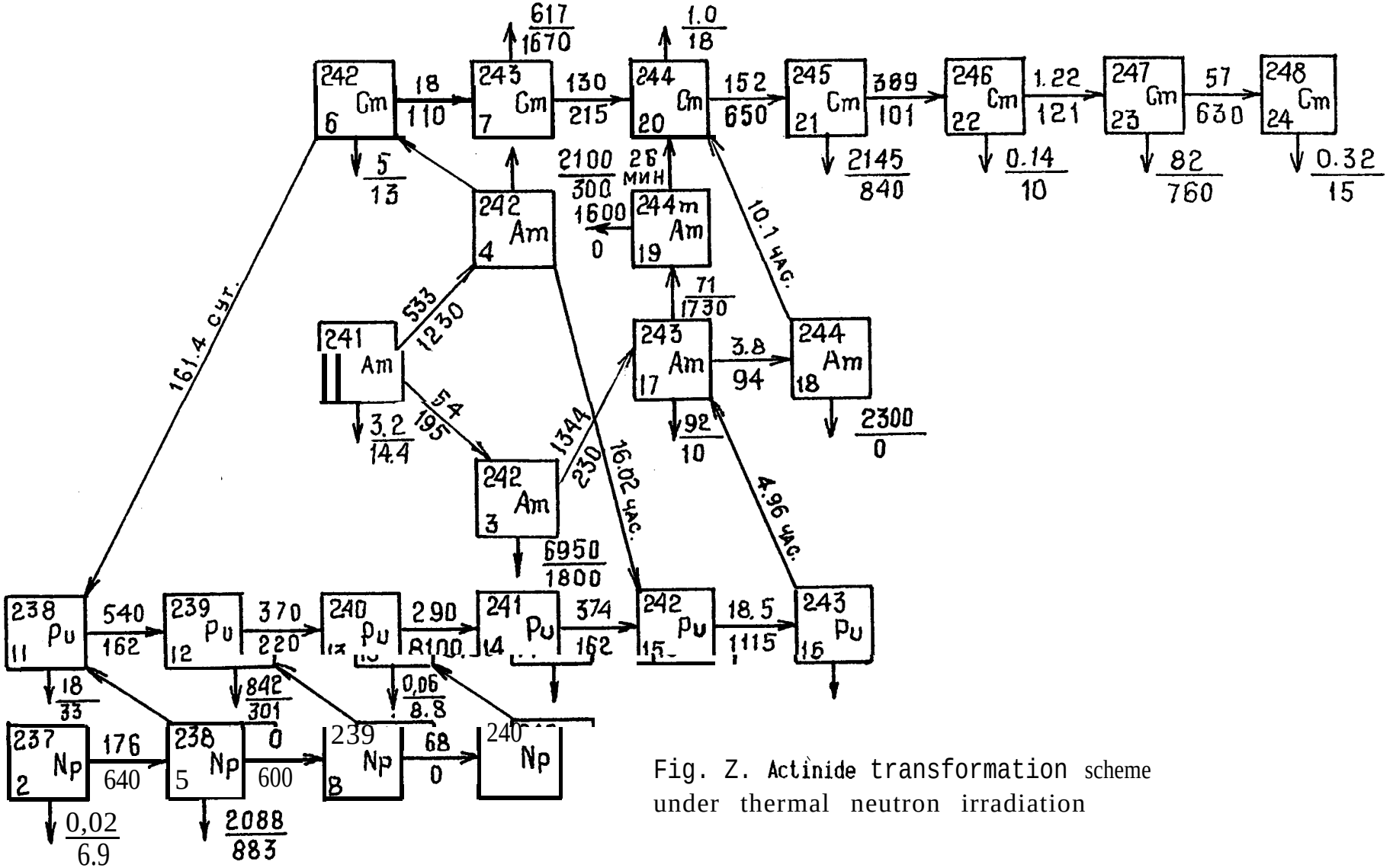
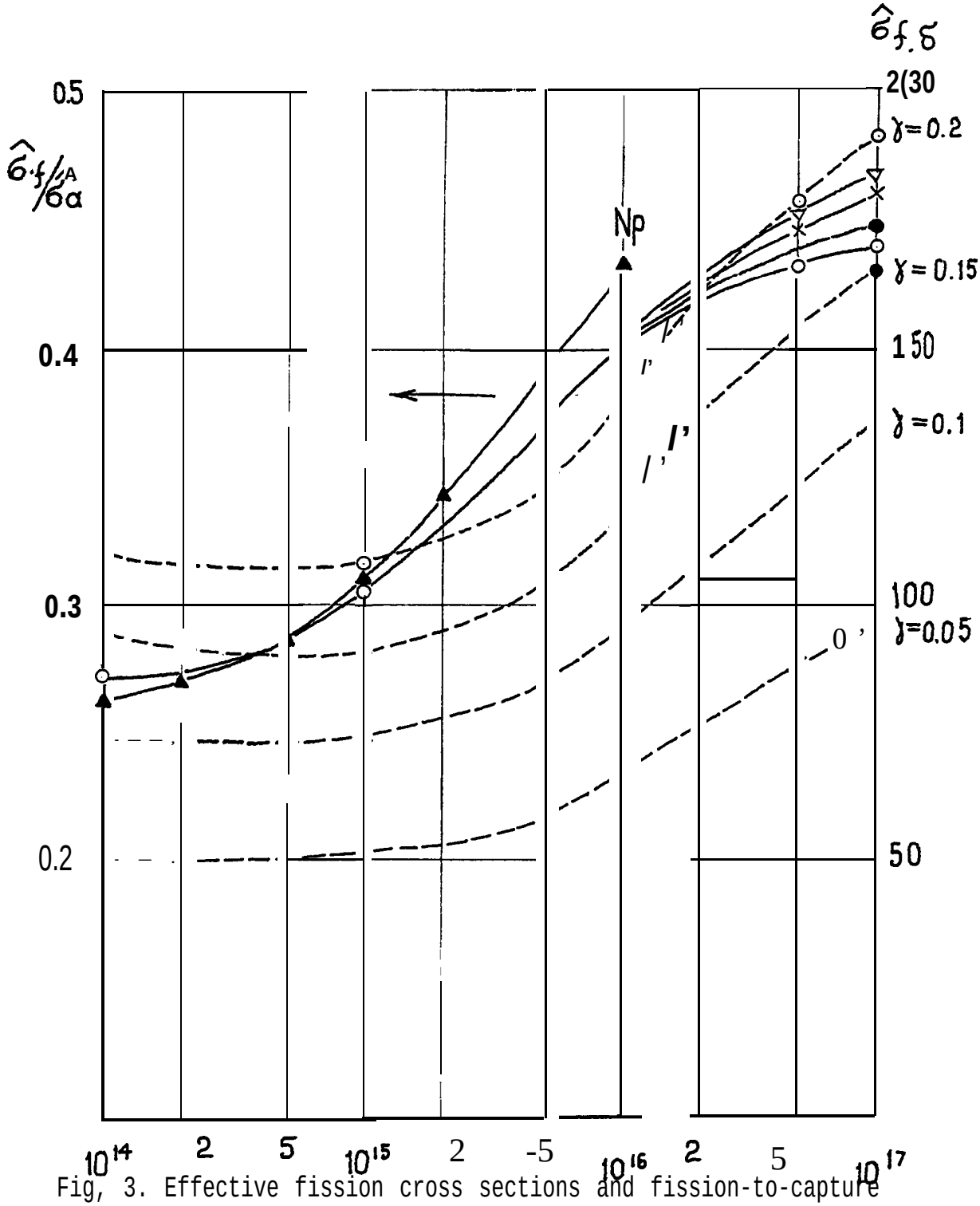


Fig. Z. Actinide transformation scheme under thermal neutron irradiation



Fig, 3. Effective fission cross sections and fission-to-capture cross sections ratios as functions of neutron flux and epithermal index Cthe VVER-1OOO waste for 3-year cooling): the same ratios for ^{237}Np loading.

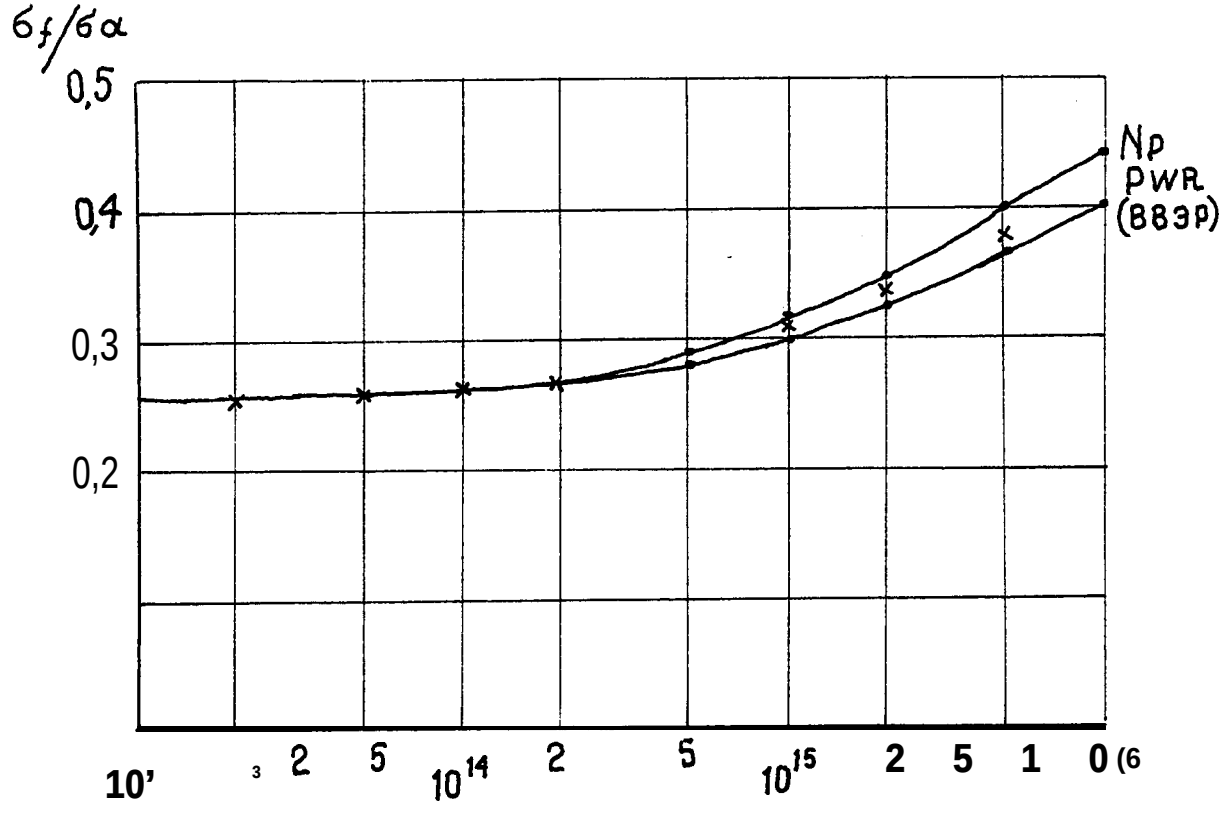


Fig. 4. Fission-to-capture cross section ratios as functions of neutron flux and waste loading type: PWR waste with 10 years cooling; VVER waste with 3-year cooling, ^{237}Np.

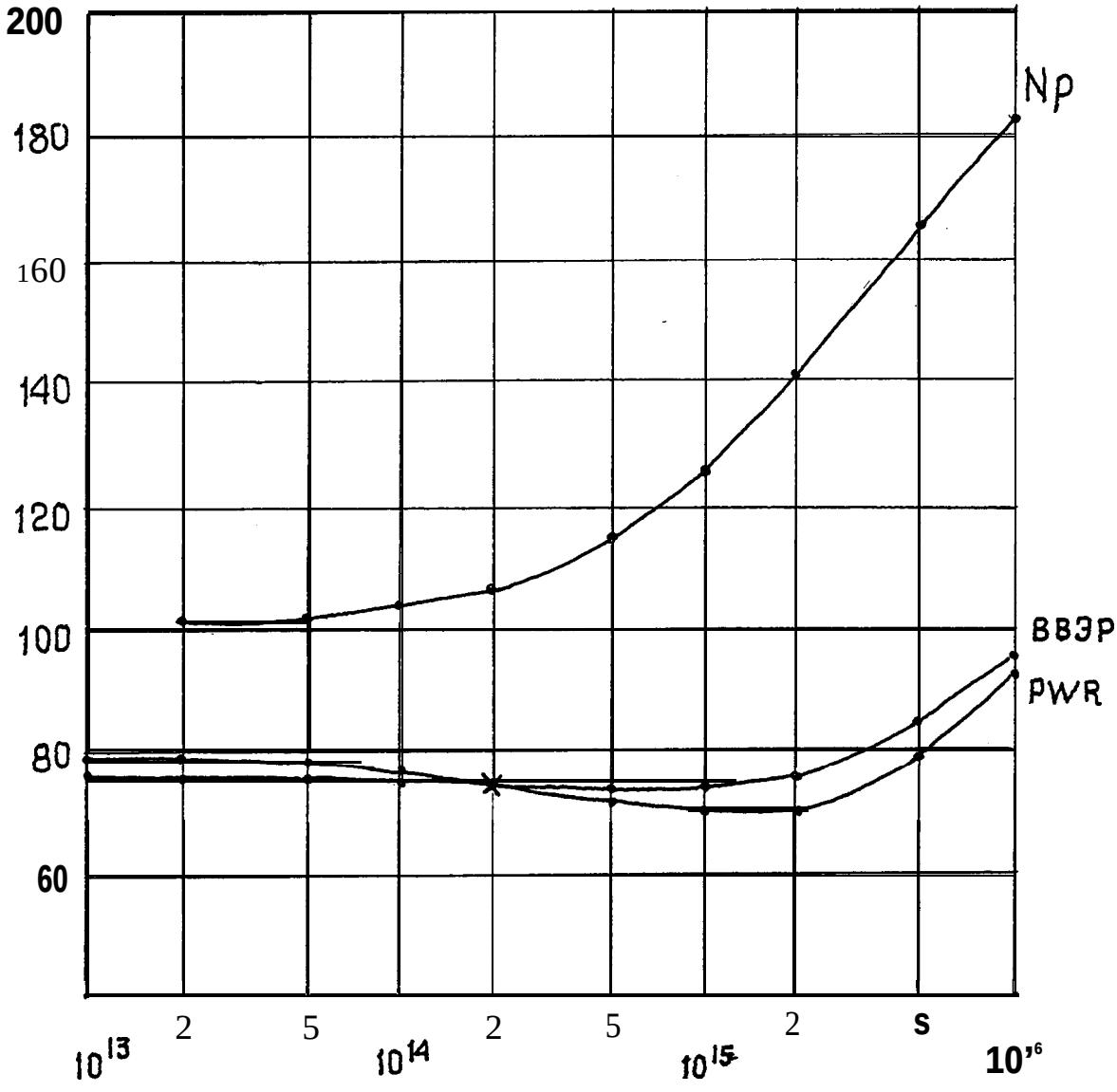
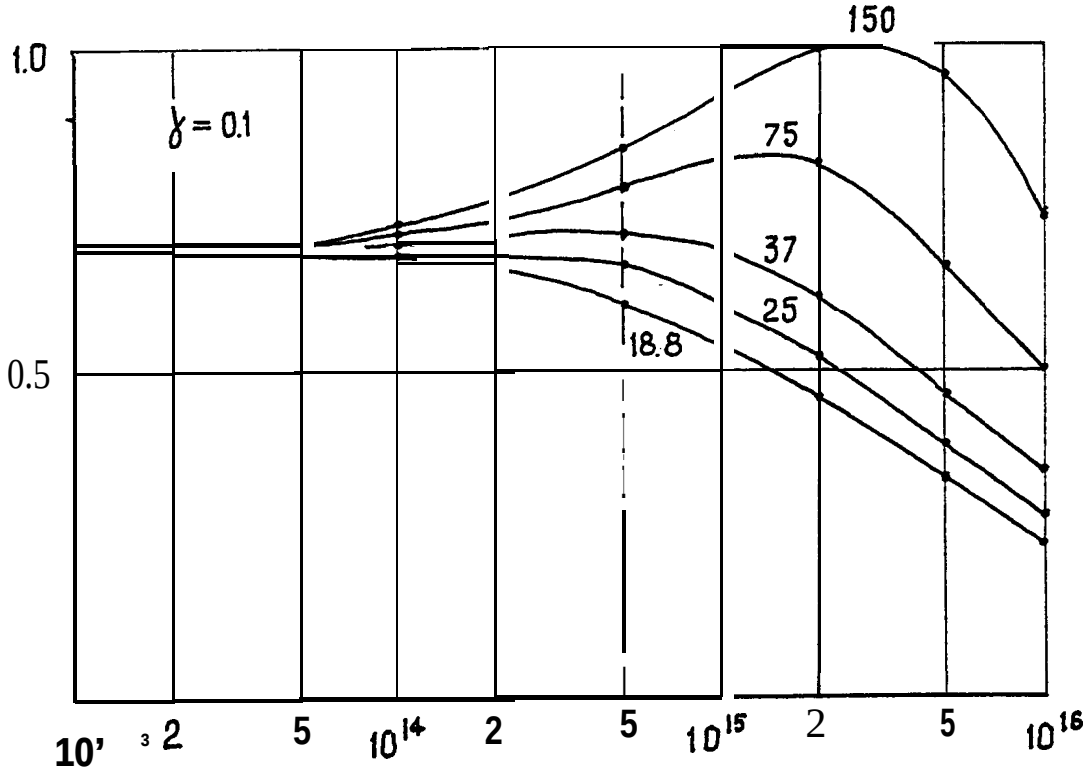


Fig. 5. Effective fission cross sections as functions of thermal neutron flux and waste loading type.



Fig, 6. Neutron flux dependence of transmutation efficiency [fissions per spallation neutron) with different power densities: epithermal index is 0.1; VVER-1OOO waste with 3-year cooling; numerical values show power density in W/cm^{-3}.

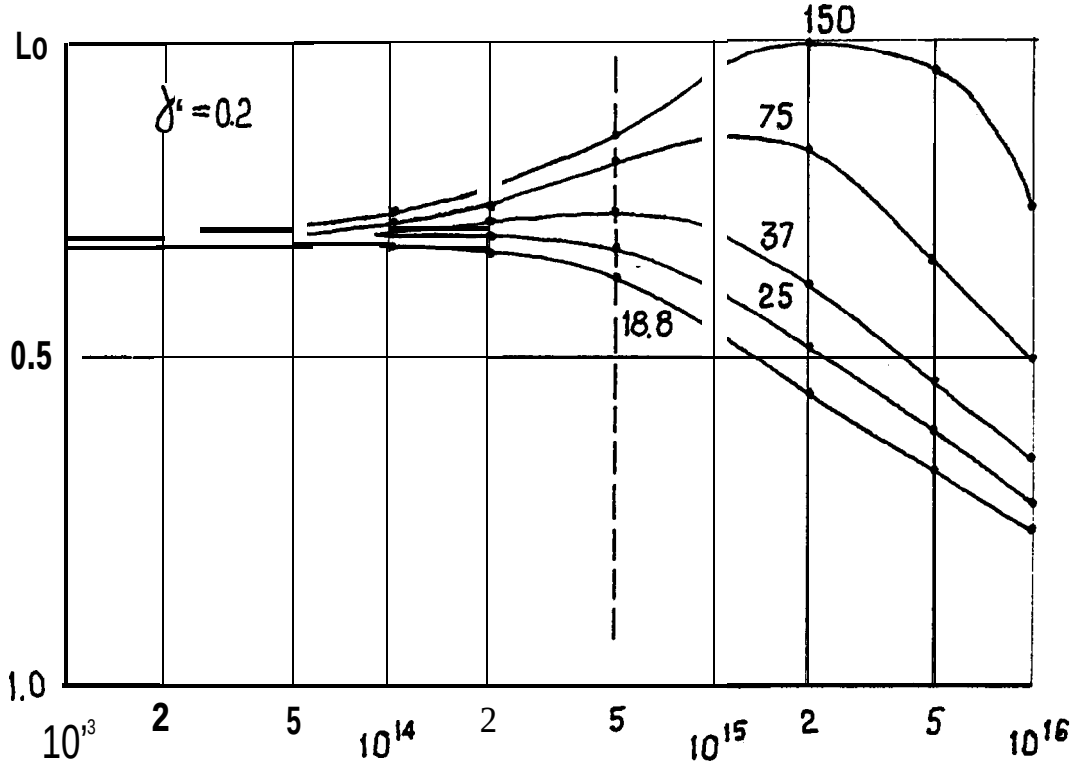


Fig. 7. Neutron flux dependence of transmutation efficiency {fissions per spallation neutron) with different power densities: epithermal index is 0.2; VVER-1OOO wade with 3-year cooling; numerical values show power density in W/cm^3.

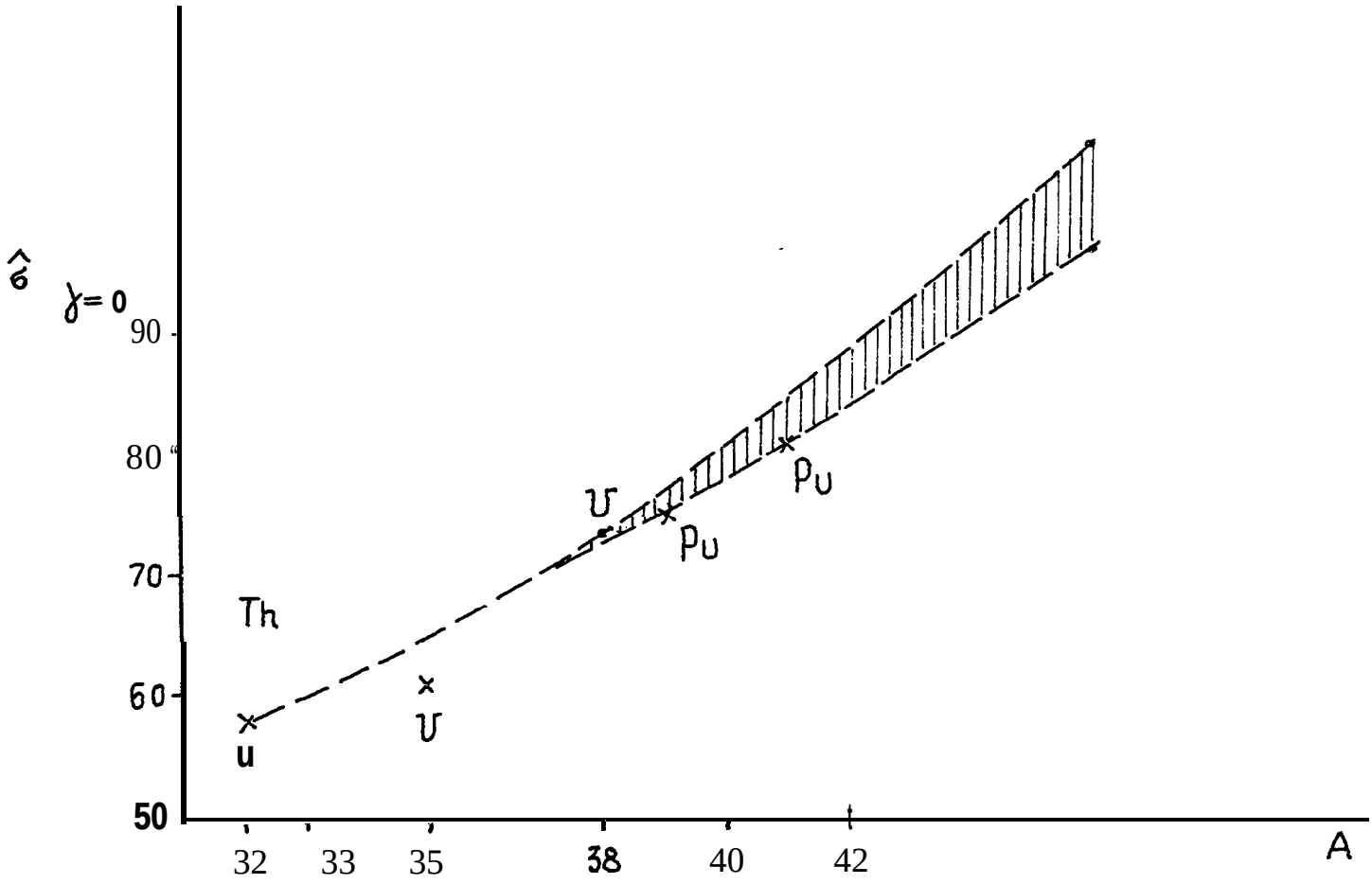


Fig. 8. Effective one-group capture cross section per a fission product pair in barns as a function of actinide atomic number[5].

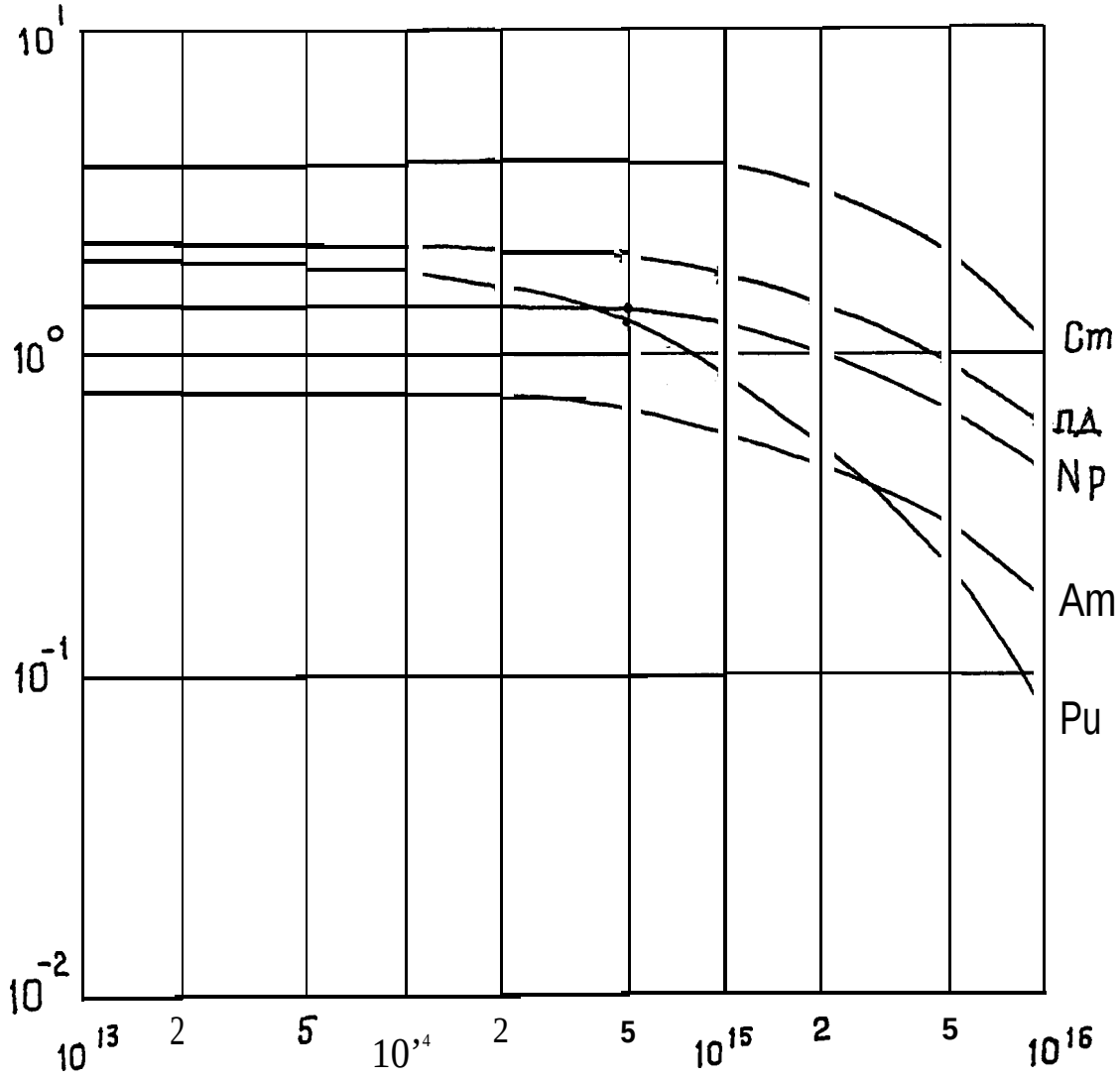


Fig. 9. Streams of transuranium elements, drawn for reprocessing (kg/day): initial loading is waste of WER-1OOO with 3-year cooling; power density is 18.8 W/cm^3.

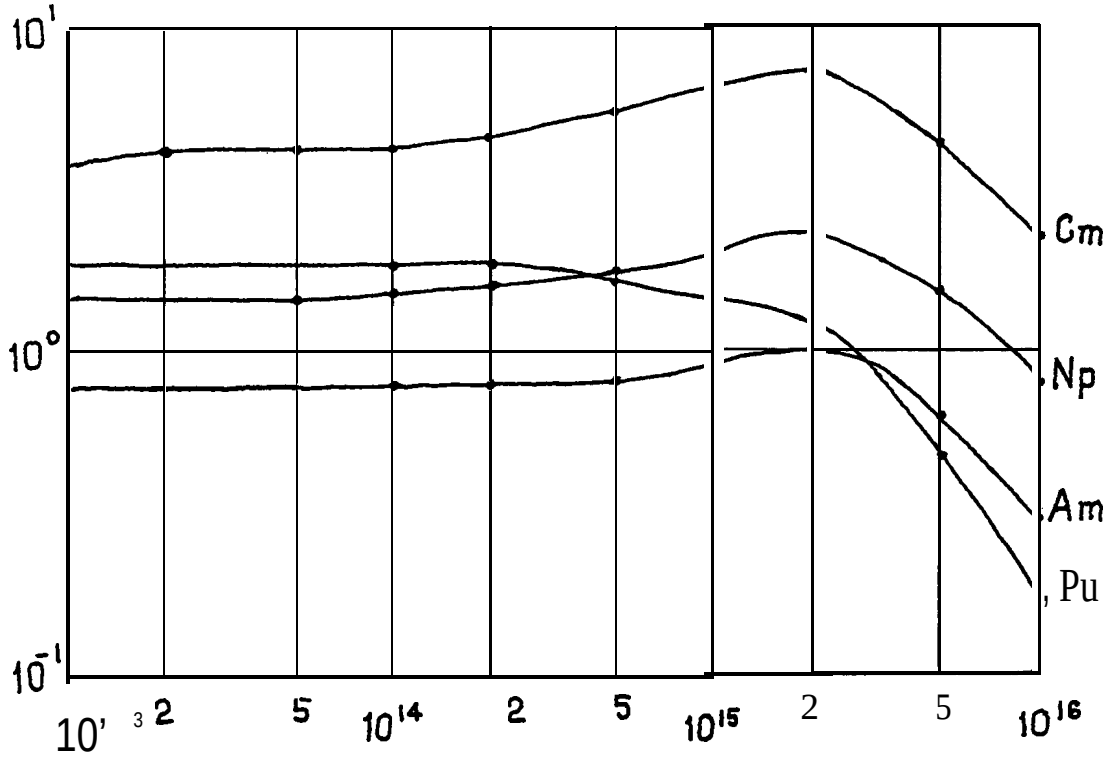


Fig. 10. Streams of transuranium elements, drawn for reprocessing (kg/day): initial loading is waste of WER-1OOO with 3-year cooling; power density is 150 W/cm^3.

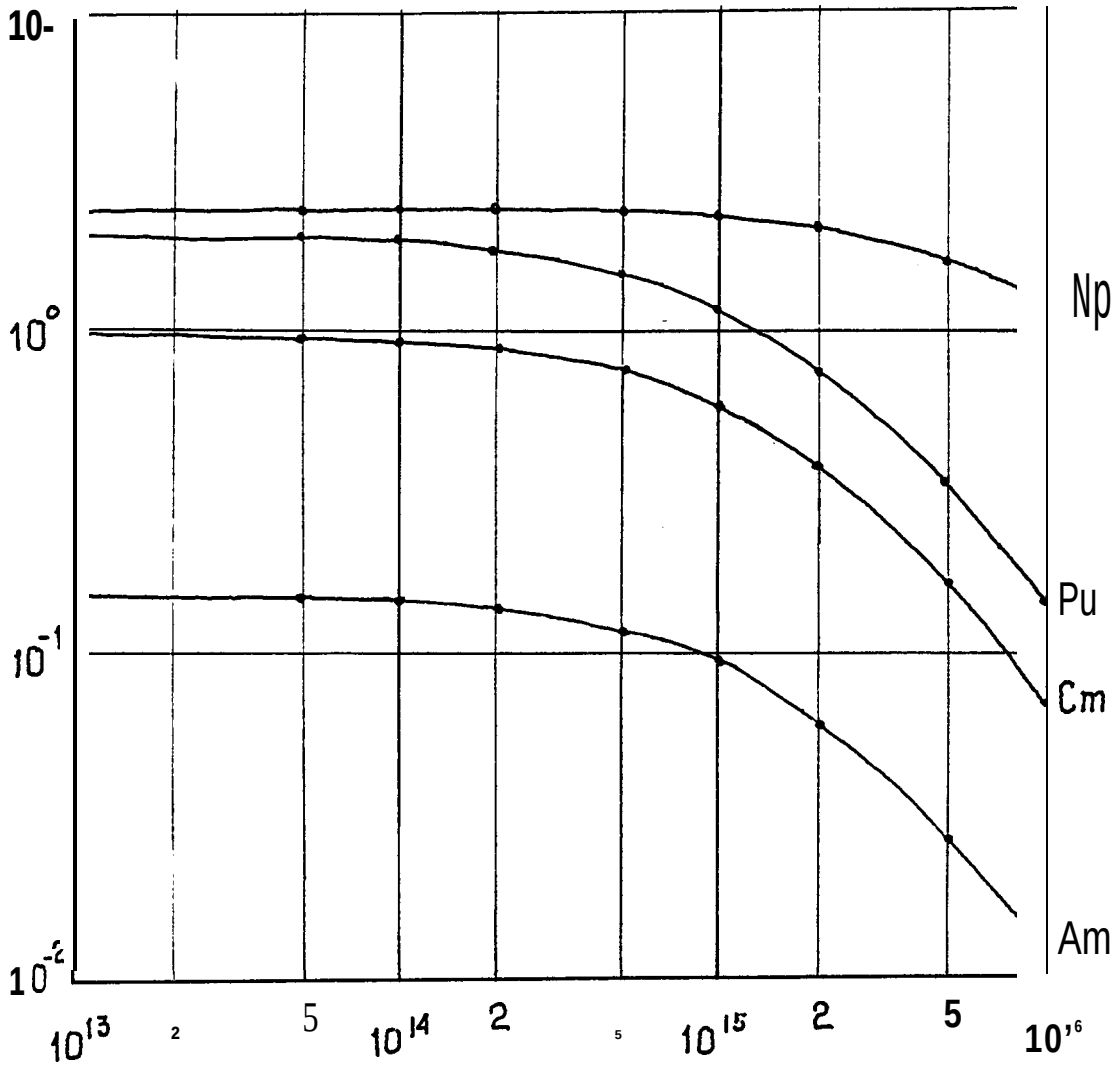


Fig. 11. Streams of transuranium elements drawn for reprocessing (kg/day) in case of ^{237}Np incineration: power density is 18.8 W/cm^3.

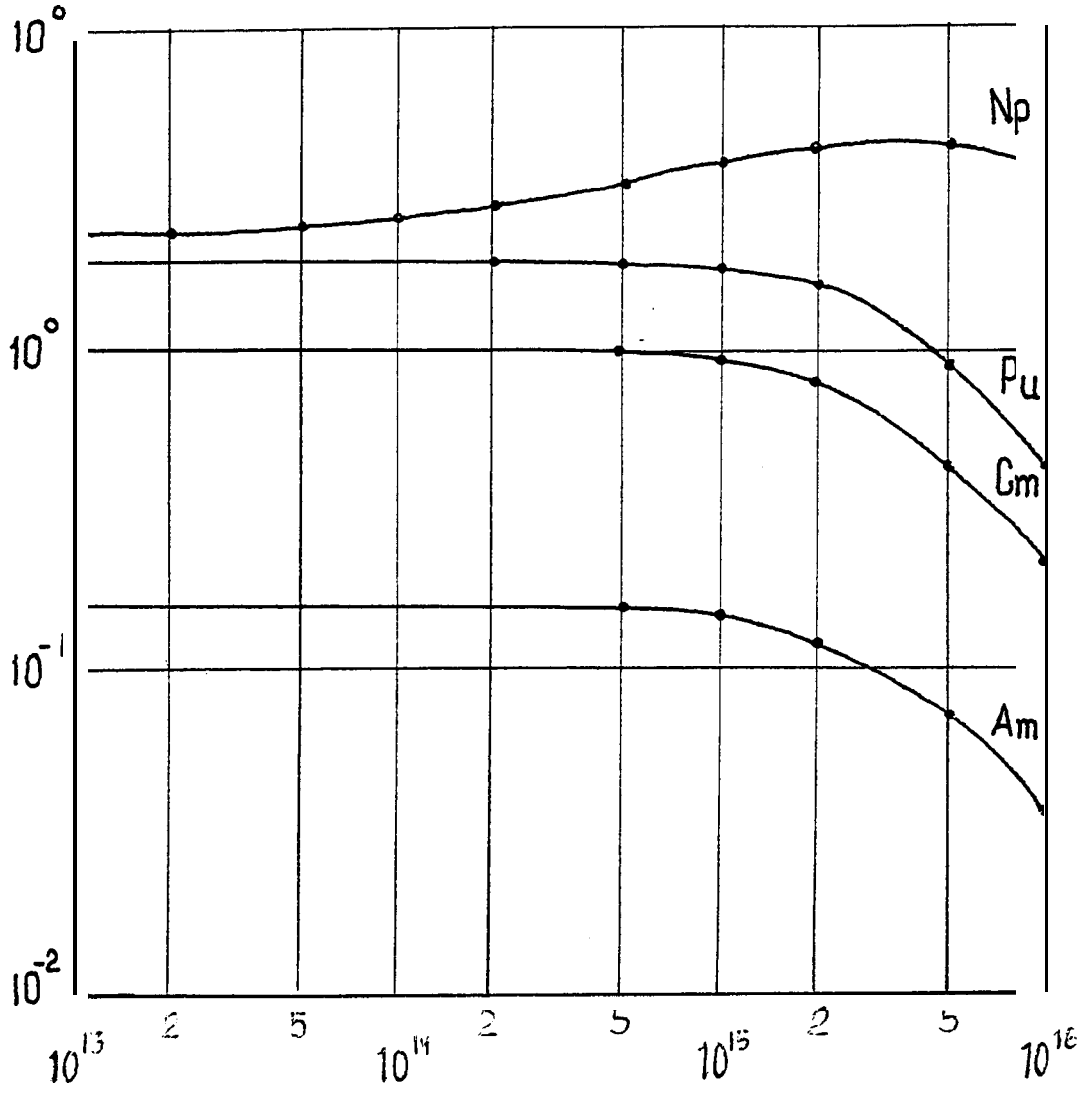


Fig. 12. Streams of transuranium elements drawn for reprocessing (kg/day) in case of ^{237}Np incineration: power density is 150 W/cm^3.

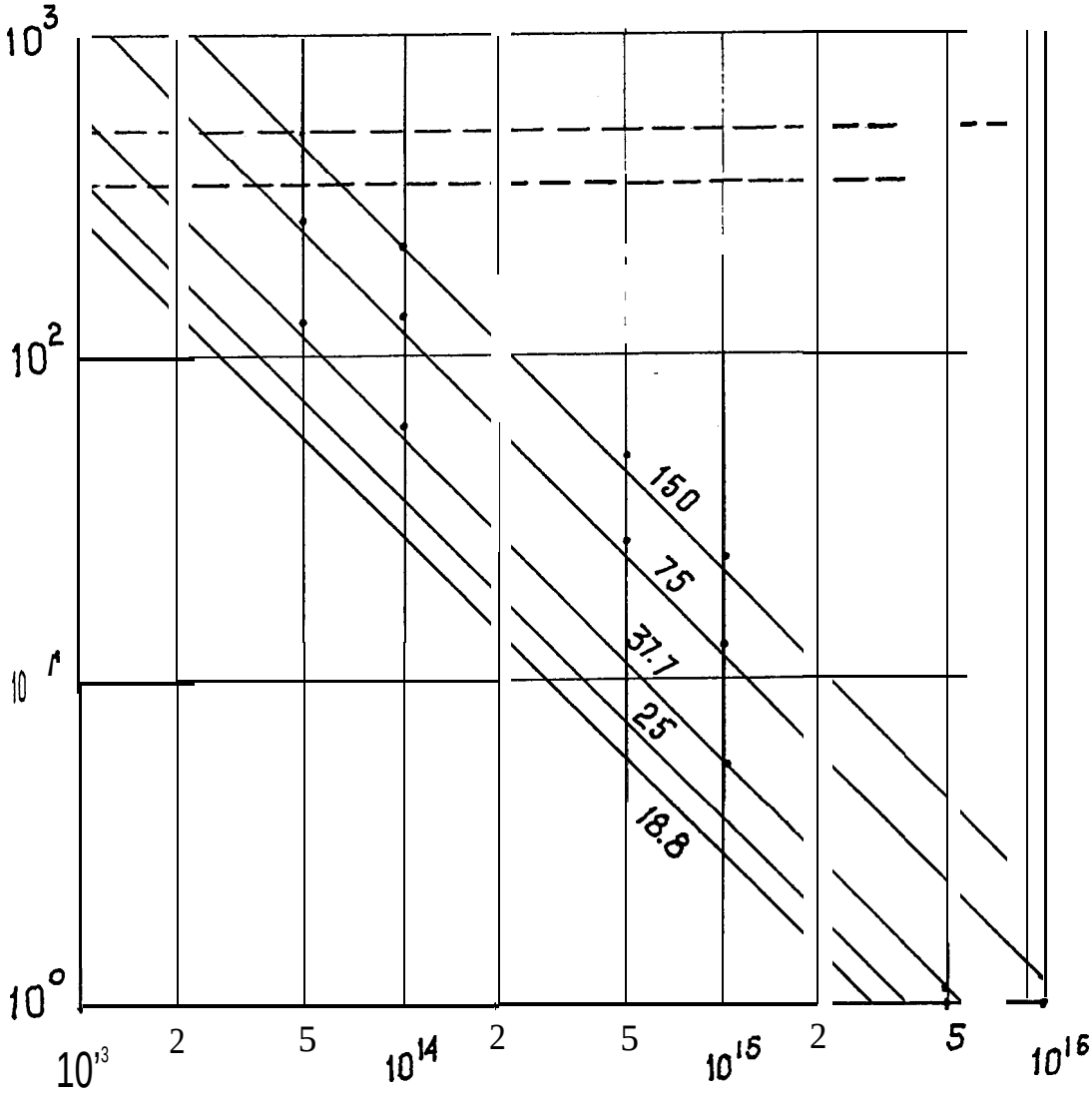


Fig. 13. Actinide concentrations in heavy water solution (g/l) under constant power density as functions of thermal neutron flux: the WER-1OOO waste with 3-year cooling; numerical values show power density in W/cm^3; dotted lines mark roughly the volubility limit.