EXPERIMENTAL TRANSMUTATION OF NEPTUNIUM IN THE BOR-60 REACTOR IN THE FORM OF VIBROPAC UNPO$_2$ FUEL

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Abstract

The main characteristics of granulated fuel (U–5%Np)O_2 produced by pyroelectrochemical reprocessing procedure and fuel pins fabricated by vibropacking technology were investigated. The fuel pins were under irradiation in BOR-60 up to the burn-ups of 13 and 20% h.a. Integrated postirradiation examinations showed high serviceability of development pin design, a good compatibility between ChS-68 cladding and (U,Np)O_2 fuel, structural fuel stability and distributions of the main FPs in the irradiated fuel, which were typical of the oxide hypostoichiometric fuel. The report presents the basic results of the investigations carried out.

Introduction

During development of the reactor fuel cycle for burning out minor-actinides (MA) the problems of introducing such elements as neptunium, americium and curium in fuel cycle are to be solved. The MA fraction (mainly consisting of neptunium and americium) in spent fuel is insignificant (0.07-0.7%), but these elements determine a long-term radioactivity and a potential ecological and radiation hazard during the spent fuel storage and reprocessing. That is why, for validation of fuel cycle for the MA burning out reactor it is necessary to search for the processes of Np-bearing fuel production and carry out in-pile tests.

Under DOVITA programme [1-3] the SSC RF RIAR started trials of the chosen process approaches using the previously developed technologies (electrochemical granulation, resulting in oxide fuel production, and vibropacking of fuel particles in fuel pin claddings for fast reactors). [4-6]

Fuel pin fabrication

The experimental batch of $UNpO_2$ was produced with Np content within 5%. In accordance with the typical flowsheet of vibropac fuel pins fabrication this fuel was used to produce experimental fuel pins by means of the equipment applied for the BOR fuel pins fabrication. The main characteristics of the fuel pins are provided in Table 1.

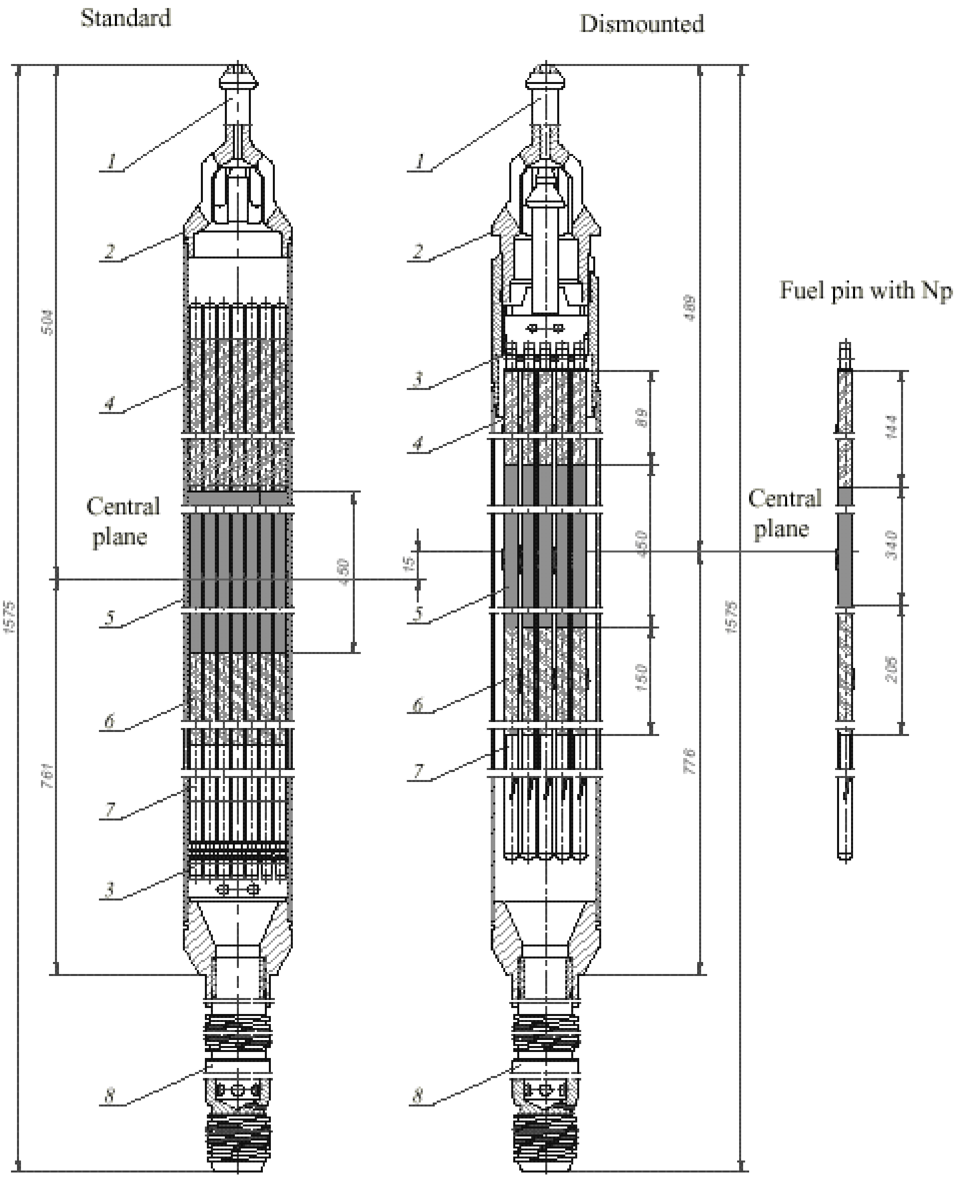
Table 1. **Characteristics of the experimental fuel pins**

1. Length, mm	
• fuel pin	1 050
• fuel column	350
• lower blanket	205
• upper blanket	144
2. Material	
• fuel column	$UO_2 + NpO_2 + U$
• blankets	UO_2
• cladding	Steel ChS-68 cold-worked (06Cr16Ni15Mo2Mn2TiVB)
3. Cladding OD, mm	
• outer	6.9
• inner	6.1
4. Weight, g	
• granulated fuel	86.5
• getter	4.5
5. NpO_2 mass fraction in granulated fuel, %	5
6. Enrichment in ^{235}U, %	
• granulated fuel	75
• blankets	0.4
7. Fuel smear density in the fuel pin, g/cm^3	9.0

In-pile tests of fuel pins

The fuel pins were tested in the BOR-60 dismantled FA developed on the basis of the BOR-60 experimental FA. This dismantled fuel assembly has the same overall dimensions, Figure 1.

Figure 1. **The BOR-60 FA and fuel pin containing $UNpO_2$ fuel**



The fuel pins were under irradiation in BOR-60 for 694 (19H) and 1 007 (20H) days at the reactor thermal power of 45-54 MW and the specific thermal loads within 29-50 kW/m, Figure 2. The BOR-60 irradiation conditions of the fuel pins with Np were computed by using code systems TRIGEX-CONSYST2-BNAB90 and SMC (system of machine computation). SMC makes it possible to carry out thorough (bearing in mind actual isotopics of fuel throughout the FA) investigations of neutron-physical characteristics (rates of fission and capture reactions, damage dose accumulation in steel as well as integral and group neutron-flux densities) of the BOR-60 reactor and to simulate different irradiation conditions of the separate fuel assemblies, fuel pins and ampoules. The fuel pins under consideration were described in detail taking into account their actual geometrical dimensions and location in the core. The fuel pin column (the core + the upper/lower blankets) was conditionally divided into separate calculation layers (in this case 14 in total). Each layer of 5 cm in thickness was characterised by its isotope composition (initial compositions for the layers of the same zone are identical), neutron field and reaction rates.

Figure 2. **Irradiation conditions of pins 19N and 20N with Np**

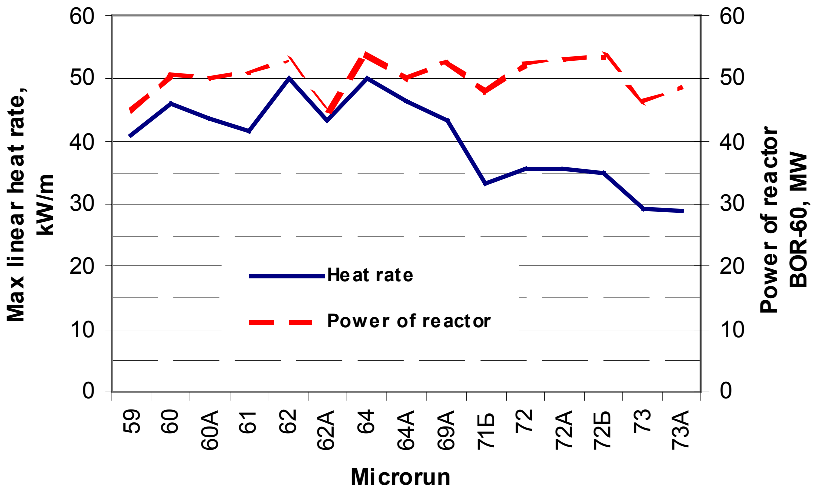
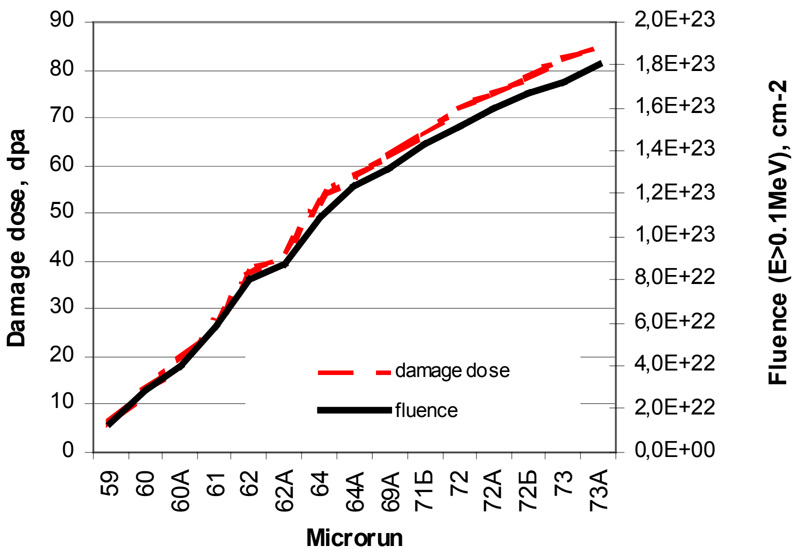


Figure 3. **Accumulation neutron fluence and damage dose in cladding of pin 19N and 20N with Np**



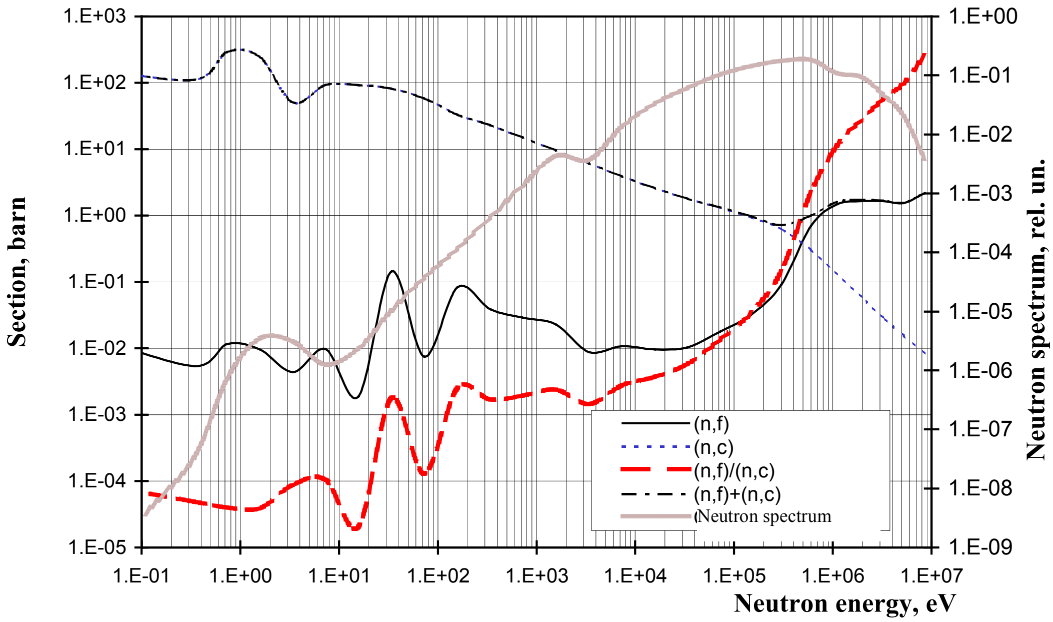
Accumulations of neutron fluence and damage dose in claddings are presented in Figure 3. During irradiation the maximum cladding temperature is 680°C.

Neptunium burning out

In-pile decrease in ^{237}Np can be caused by both ^{237}Np nucleus fission, resulting in FPs production, and neutron capture with nucleus with its subsequent transmutation into heavier isotope. In the second case there is no decrease in the number of heavy nuclei of transuranium elements. From analysis of the energy dependencies of the effective fission and capture sections, $\sigma(n,c)$ and $\sigma(n,f)$, it is inferred, that ^{237}Np fission section exceeds its capture section at the neutron energy of more than 0,4 MeV. Figure 4 presents distributions of the capture section (n,c), fission section (n,f), their ratio and sum as well as the normalised neutron spectrum during the fuel pins irradiation in BOR-60 as a function of energy. Integral sections of $\sigma(n,c)$ and $\sigma(n,f)$ reactions in the fuel columns were 0.66÷0.79 barns and 0.55÷0.65 barns, correspondingly, i.e. more than half ^{237}Np transmutes into ^{238}Pu as a result of neutron capture, and the other transmutes into fission products.

Change in $UNpO_2$ isotopics over the whole period of in-BOR-60 irradiation is provided in Table 2.

Figure 4. **^{237}Np sections**



The average fuel burnup in fuel pin 20-N at the end of irradiation achieved 17.4% h.a., and ^{237}Np mass decreased down to 16.9%. The maximum fuel burn-up and decrease in ^{237}Np mass in the midplane of fuel pin 20N were 19.5% h.a. and 19.0%, correspondingly. Uranium mass decreased by 14.2 g (by 19%). About 1 g of plutonium accumulated in fuel, with ^{238}Pu fraction exceeding 50%. In accordance with the actinides transmutation chain almost all (more than 99,9%) ^{238}Pu was obtained from ^{237}Np ($^{237}Np(n,\gamma)$ ^{238}Np--β–>^{238}Pu), and $^{239\text{-}240}Pu$ isotopes were resulted from neutron capture with ^{238}U.

Non-destructive examinations of the irradiated fuel pins

The following non-destructive examinations (NDE) of the fuel pins were performed:

- visual inspection;
- eddy-current flaw detection of the fuel pin claddings;

- OD measurements of the fuel pin cladding;
- gamma-scanning along the fuel pin length.

Visual inspection of the fuel pins bundle showed, that the fuel pins with $UNpO_2$ and those with the BOR-60 standard fuel, which were irradiated in the same FA, did not differ in appearance.

Table 2. **Fuel isotopics prior to and post irradiation**

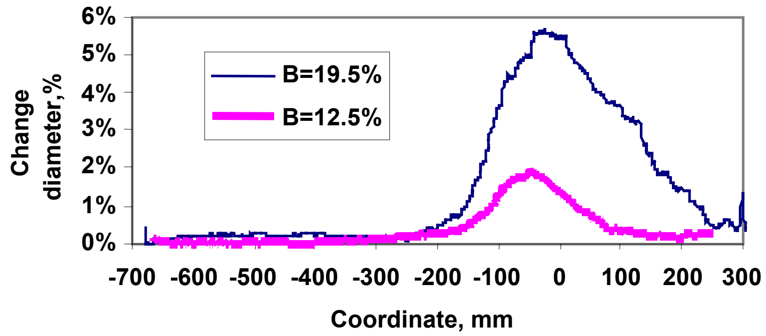
Isotope	**Initial composition** (g)	**The irradiation end**			
		19-N		**20-N**	
		Mass isotope (g)	Change (%)	Mass isotope (g)	Change (%)
^{234}U	0.64	0.60	-6.3	0.57	-11
^{235}U	53.46	43.4	-18.8	38.1	-29
^{236}U	5.48	6.8	+24.1	7.5	+37
^{238}U	16.22	15.7	-3.2	15.4	-5
^{237}Np	**4.63**	**4.12**	**-11.0**	**3.85**	**-16.9**
^{238}Pu		0.33		0.53	
^{239}Pu		0.30		0.46	
^{240}Pu		3.9E-3		0.011	
^{241}Pu		3.6E-5		2.3E-04	
^{242}Pu		3.1E-07		2.5E-06	
FP*		9.2	11.4	14.0	17.4
Total	**80.43**	**80.43**		**80.44**	

* FP – fission products isotopes

OD measurements of fuel pins 19-N and 20-N (damage dose is 55 and 85dpa, correspondingly), Figure 5, demonstrate, that in the fuel column the maximum increase in OD is 1.9% and 5.5%. Increase in OD and damage dose for the 20-N fuel pin cladding is considered limit for the cladding material ChS-68.

Gamma-scanning of both fuel pins, fig.6, indicated, that in the hypostoichiometric irradiated fuel volatile fission product ^{137}Cs trended towards migration to the colder fuel pin zones, including upper and lower blankets, where its content was quite high. Its highest content was found on the lower (from the gas plenum side) border of the lower blanket. Behaviour of fission products ^{95}Zr and ^{106}Ru is typical of the oxide fuel pins: ^{95}Zr distribution corresponds with the fluence profile, and ^{106}Ru has local peaks in fuel pin 19-N, which are not numerous, but rather intensive. In fuel pin 20-N, which was irradiated under “mild” conditions during the last stage, local peaks of ^{106}Ru intensities, Figure 2, are much lower.

Figure 5. **Distribution diameter**



Destructive examinations of the irradiated fuel pins

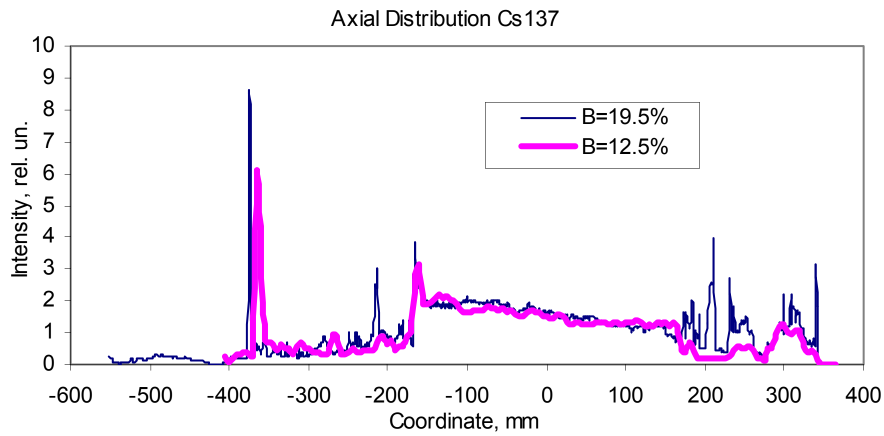
One of the fuel pins (19-N) with the maximum burn-up 12,5% h.a. was investigated by destructive methods: ceramography, EPMA, TEM, etc.

The fuel column structure is mainly similar to that typical of the irradiated oxide fuel: a central hole, columnar grains, a transition zone and a circumferential zone of the initial fuel structure, Figure 7.

Investigations of the cross sections confirmed the fuel pin cladding integrity: no cracks, damages, FCCI signs and fission products were found, Figure 8.

Radial distributions of Np, U, Pu and Nd were determined by EPMA method. Metal inclusions consisting mainly of molybdenum, uranium, technetium and noble metals were identified in the fuel structure (Ru,Rh,Pd), Figure 9.

Figure 6. **Axial distributions of Cs (a), Ru (b) and Zr (c) in fuel pin $UNpO_2$**



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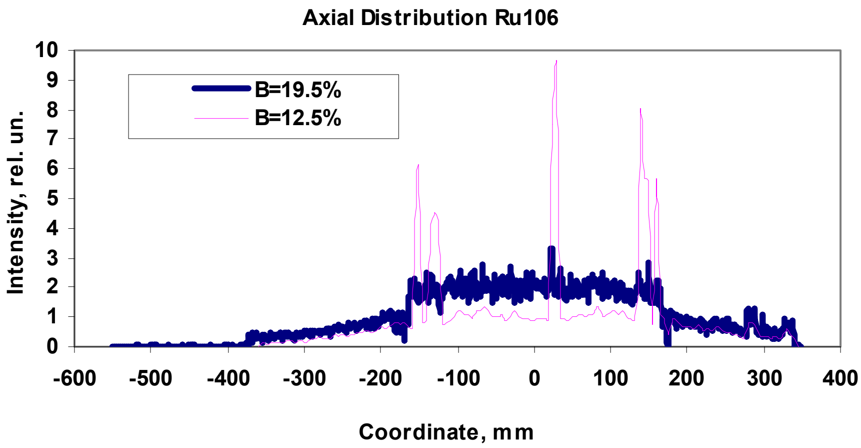
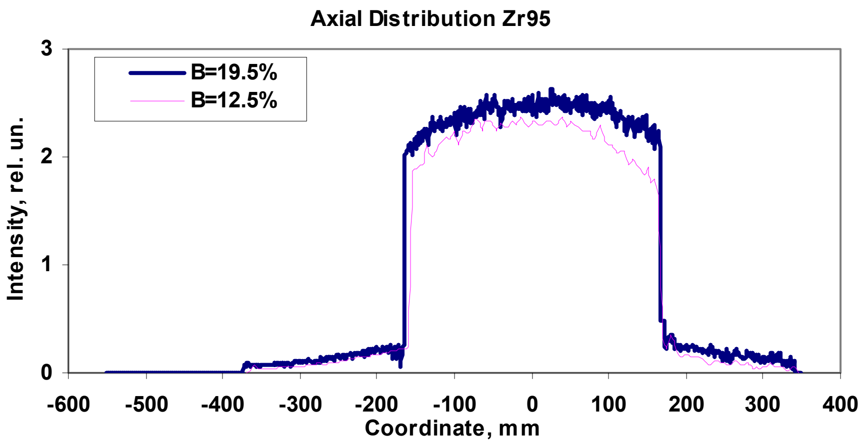
(a)

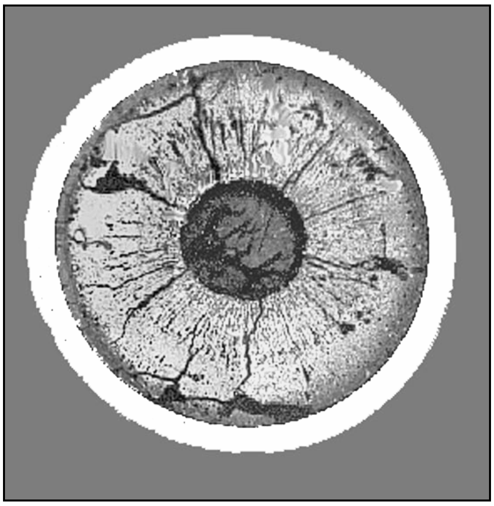


(b)



(c)

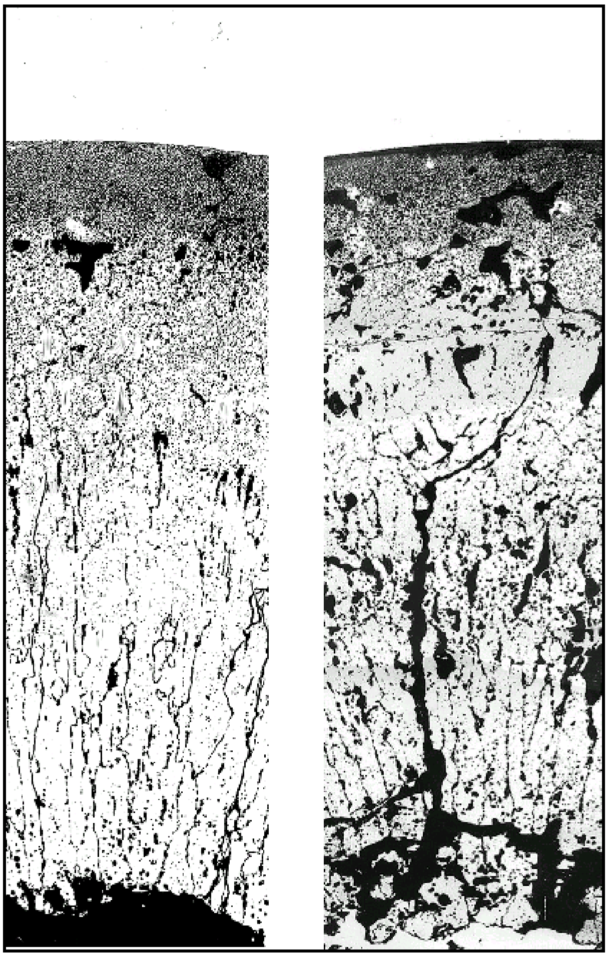
Figure 7. **Macrostructure of irradiated fuel $UNpO_2$ (B=12.5%)**



+90 mm (relative to the Midplane)

+145 mm (relative to the Midplane)

Figure 8. **Microstructure of $UNpO_2$ irradiated fuel and fuel-cladding boundary (B=12.5%)**



Conclusions

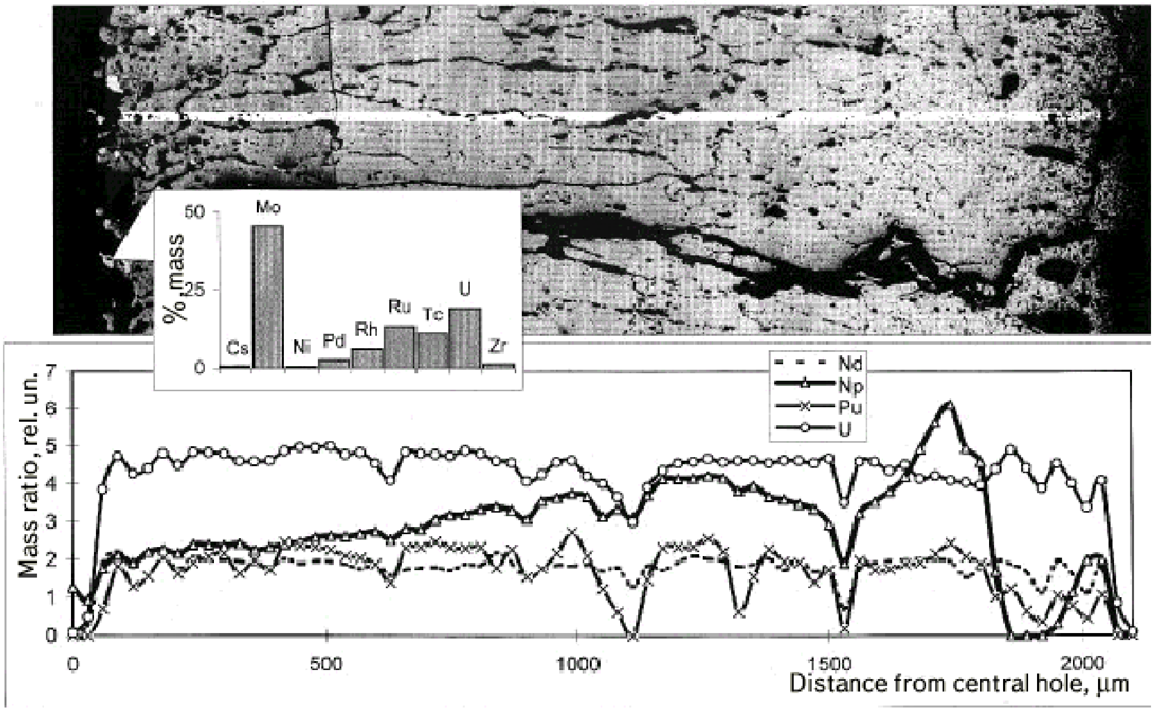
The experiment in BOR-60 confirmed the efficiency of vibropac fuel pins containing fuel composition $(U\text{-}5\%Np)O_2$ both at the burn-up of no more than ~20%h.a. and at the limit maximum damage dose for ChS-68 steel cladding (85 dpa) specified for the BOR-60 fuel pins having this type of cladding.

During irradiation of this fuel composition in the BOR-60 neutron spectrum the decrease in ^{237}Np mass (^{237}Np transmutation) is 16.9%, and in the midplane of fuel pin 20-N-19.0%.

PIE confirmed a good compatibility between ChS-68 cladding and $(U,Np)O_2$ fuel, structural fuel stability and distributions of the main FPs in the irradiated fuel, which were typical of the oxide hypostoichiometric fuel.

No significant differences in efficiencies of the fuel pins with $UNpO_2$ and fuel pins containing UO_2 or $UPuO_2$ were found.

Figure 9. **Elements distribution along the fuel pin radius (+28 mm relative to the Midplane) and metal inclusion composition (B=12.5%)**



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