BURNING OF MINOR ACTINIDES IN FUEL CYCLE OF THE FAST REACTOR: DOVITA PROGRAMME – RESULTS OF THE 10 YEAR ACTIVITIES

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Abstract

Taking into account advantages of pyrochemical technology and vibropacking method RIAR had started in 1992 the research under the DOVITA programme. It is the programme for demonstration of minor-actinide burner reactor. The reprocessing of the irradiated fuel and targets is offered by two ways: a) pyroelectrochemical and b) non-complete vacuum-thermal. The vibropacking technology is applied to fuel pins manufacturing. All products are reprocessed with the purpose of complete recycle of Pu, Np, Am and Cm. A number of experimental researches on Np recycle were carried out. The study of electrocodeposition of Np and Am with UO_2 from molten chlorides were fulfilled. The studies are continuing.

Introduction

The problem of long-lived minor-actinides (Np, Am, Cm) transmutation is one of major part of problem of nuclear power ecological safety. [1] The problem of Pu surpluses burning-out [2] adjoins to this problem. Existing and perspective reactor systems could be used for it, but task of optimum organisation of the external closed cycle for actinide burner reactor becomes the important aspect of transmutation problem.

DOVITA programme

Since 1992, SSC RIAR has proposed the demonstration programme-concept DOVITA (Dry reprocessing, Oxide fuel, Vibropac, Integral, Transmutation of Actinides), which should demonstrate opportunities of new technologies for realisation of the optimised fuel cycle for actinide burner reactor (for transmutation of Np, Am, Cm).

RIAR use BOR-60 reactor as the experimental base for carrying out of DOVITA programme investigations, because highly hardness of BOR-60 neutron spectrum is very beneficial for minor actinides burning.

The flow sheet of fuel production and reprocessing provides homogeneous option for neptunium recycle in MOX fuel. It is preferable to introduce americium and curium into special targets.

Fuel recycle is carried out by the following flow sheets:

- The basic (driven) MOX fuel, after 15-20% burn-up, is underwent to decladding, crushing, vacuum reprocessing and repeated vibropacking.
- After two cycles of irradiation, the basic fuel is reprocessed by pyroelectrochemical methods. U, Pu and Np (and portion of Am) come back to irradiation.
- The materials of targets with Am, Cm and REE are irradiated by 3-4 cycles with periodic decladding and treatment, then they are directed to pyrochemical process.
- The pyrochemical process of Am and Cm separation from REE will be carried out in system with liquid metal electrode, thus the REE portion is dumped only.

The ways for Cm utilisation: that is expedient to separate it from wastes and to destroy in systems with high-flux reactor. The Cm burning efficiency is 85% after 500 eff. days irradiation, and valuable isotope ^{252}Cf producing rate is 1 mg from 1 g of Cm.

The analysis is carried out for procedures, which would allow extracting the valuable FPs under reprocessing: the extraction of 60-70% noble metals practically would not affect on operational expenses.

The step-by-step work is carried out for the criteria choice, description and substantiation of DOVITA fuel cycle concept on the basis of the data.

The ABFR fuel cycle (see Figure 1) differs essentially from the fuel cycle of thermal neutron reactors. It is caused by it is inexpedient to release FPs from fuel containing minor actinides and plutonium up to high decontamination factors, because its activity is determined mainly by actinides radiation. As result it is necessary to realise all operations with fuel in remote conditions. ABFR closes

a fuel cycle of nuclear power as a whole, so the special requirements must be presented to amount and form of radioactive wastes formed in its external fuel cycle. Besides, taking into account a lower economic efficiency of ABFR in comparison with power reactors, the fuel manufacture should be cheap and compact.

Thus, it is necessary to take into account the following requirements:

- Minimum amount of HLW, which should be mainly in solid form, convenient for final disposal.
- Minimum quantity of technological stages for fuel reprocessing, enough for restoration of its properties before recycling into reactor; thus decontamination factor from FP can be low.
- Multi-cycle repeated irradiation of fuel and maximum possible recycle of radiotoxic components.
- Realisation of all technological operations in shielded cells in conditions of remote service.

This approach to the decision of ABFR fuel cycle problem is based on experience and knowledge obtained during development of the fuel cycle with application of pyroelectrochemical (and other "dry") methods for fuel reprocessing and production and vibropacking method for fuel pins manufacture.

$(U,Np)O_2$ and $(U,Pu,Np)O_2$ fuel

The main parameters of obtaining the $(U–5\%Np)O_2$ granulated fuel by the electrolysis in melted salts, as well as the physico-mechanical characteristics of the granulate, have been studied. Fuel pins fabricated by the vibropack technology do not differ by their qualitative values from the regular BOR-60 vibropack fuel pins. The fuel pins were irradiated in the BOR-60 reactor up to the burn-up of about 13 and 20% h.a. Figure 2. FP 20-H was irradiated up to the maximum damaging dose for the ChS-68 alloy cladding (85 dpa) determined for the BOR-60 fuel pins. After irradiation, fuel pins were investigated by the non-destructive methods (visual-optical inspection, vortex-current defectoscopy, profilometry, gamma-scanning), Figure 3, and by destructive methods (ceramography and X-ray micro-spectrum analysis), Figure 4, 5, and 6. The results of the complex post-reactor investigations show good compatibility of the ChS-68 alloy cladding and $UNpO_2$ fuel, as well as structural stability of the fuel and typical of the oxide pre-stechiometric fuel distributions of fission products in irradiated fuel.

During irradiation of the $(U–5\%Np)O_2$ fuel composition in the BOR-60 neutron spectrum, ^{237}Np mass reduction (^{237}Np transmutation) was 16.9%, and 19.0% in the FP central plane.

Essential differences in efficiency of the fuel pins with $UNpO_2$ fuel in comparison with that of the fuel pins containing UO_2 or $UPuO_2$ fuel were found as non-existent.

At present, destructive investigations of FP 20-H with burn-up about 20% have been started. The work are planned to be completed in June 2003. The batches of UO_2- 20%PuO_2- (3-6)%NpO_2 fuel have been produced and the physico-mechanical and physico-chemical properties of their components have been investigated. Fabrication of vibropack fuel pins containing this fuel is scheduled for the end of 2002.

Figure 1. **Fuel cycle of actinide burner reactor (DOVITA fuel cycle)**

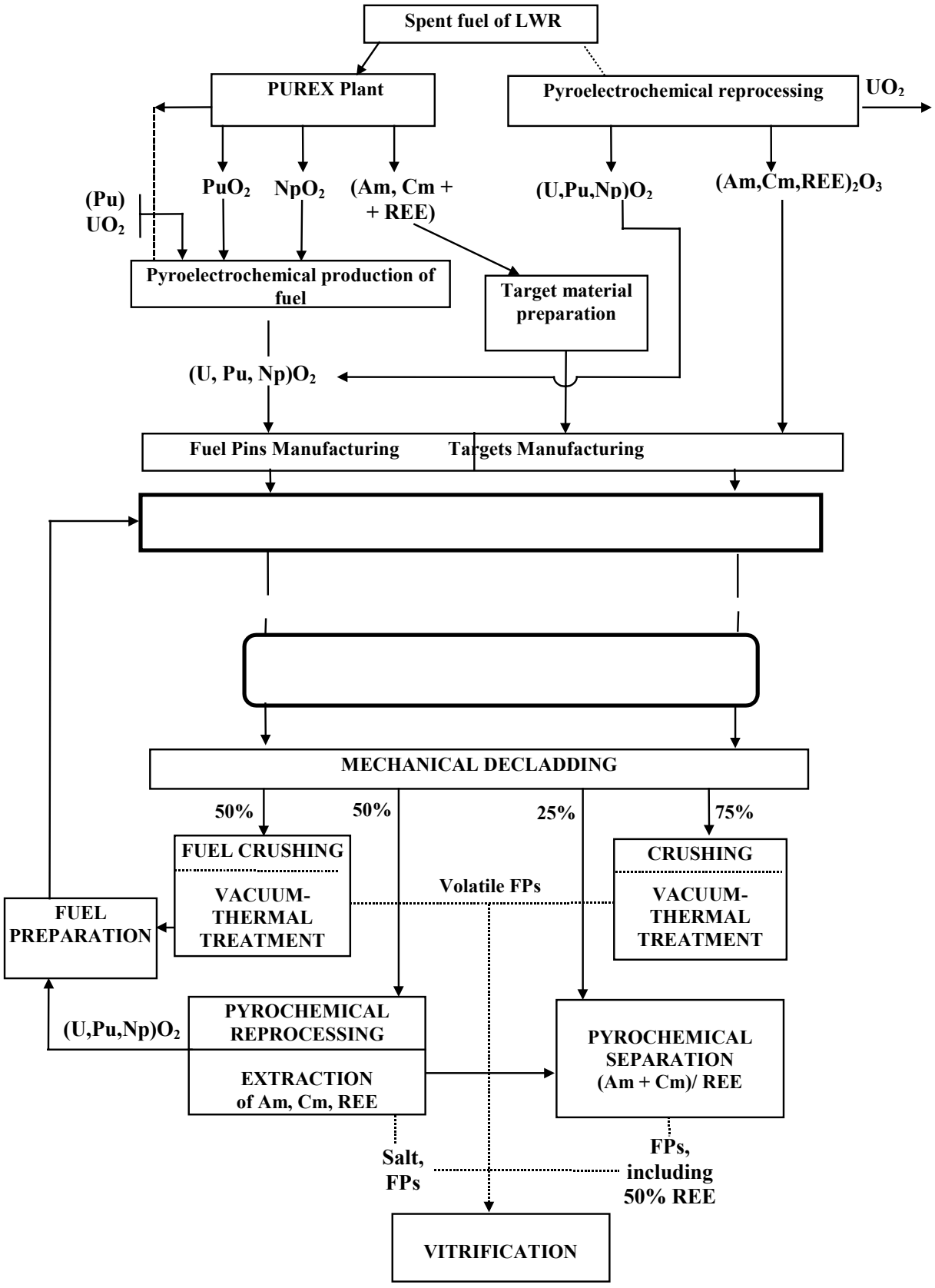


Figure 2. **Irradiation conditions of pins with (U, Np)O_2 fuel in BOR-60 reactor**

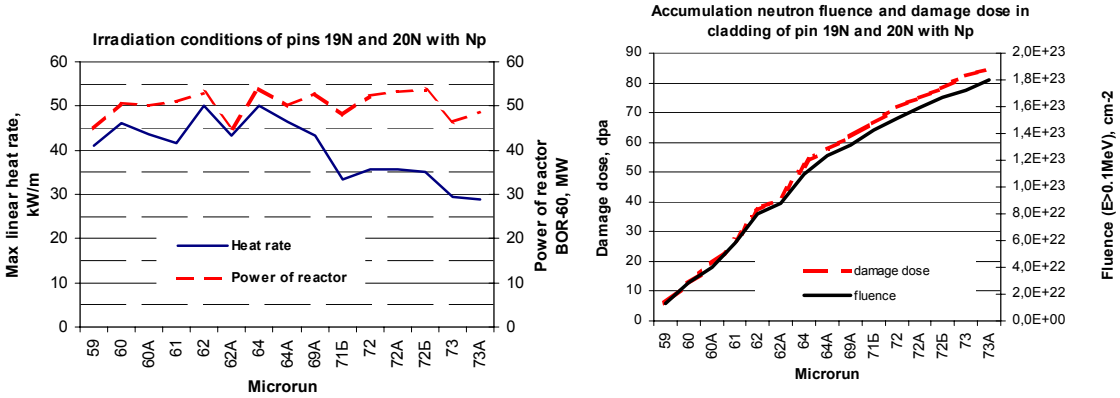
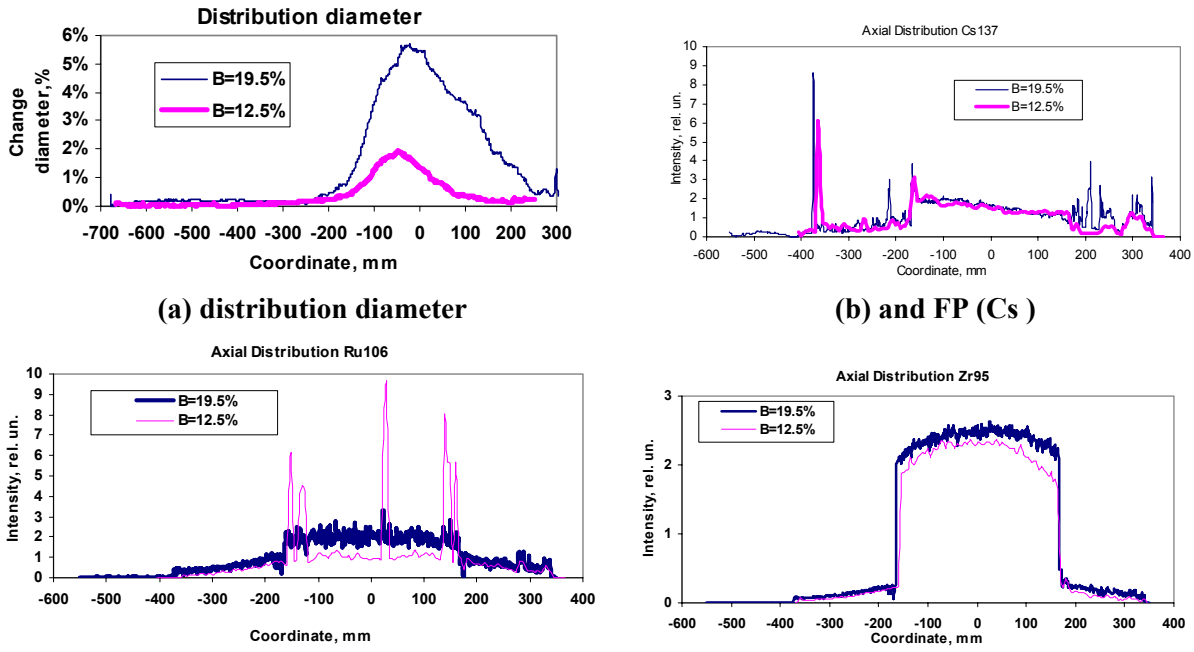


Figure 3. **Distribution diameter along the fuel pins**



(a) distribution diameter **(b) and FP (Cs)**

(c) Ru **(d) и Zr**

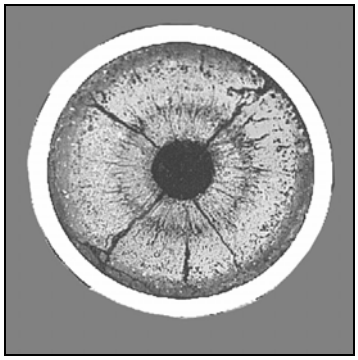
MOX fuel with addition of americium

From data on electrodeposition mixed uranium-cerium oxides (Ce was chosen as Am simulator) [4] there were carried out laboratory experiments on the obtaining (U,Pu,Am)O_2 fuel composition. There have been obtained four products with weights within the range from 50g to 70g, americium content of 2-3%wt and plutonium content over the range from 2 to 50% (Table 1).

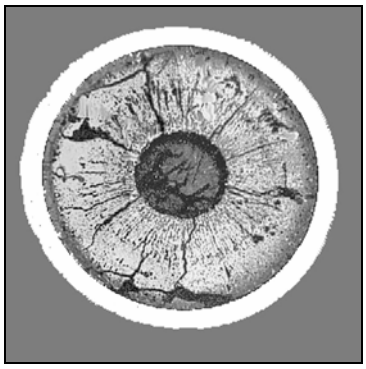
Now these products are being investigated and prepared for irradiation in BOR-60 reactor.

Americium behaviour in electrolysis of $(U,Pu,Am)O_2$ codeposition in the molten system NaCl-2CsCl at the temperature of 750°C is presented in Figure 2. Analysis of data, presented in Figure 7, allows us to conclude, that under these conditions **americium oxide electrodeposition starts at the cathodic potentials φk>1,6V.**

Figure 4. **Macrostructure of irradiated fuel $UNpO_2$** (B=12.5%)

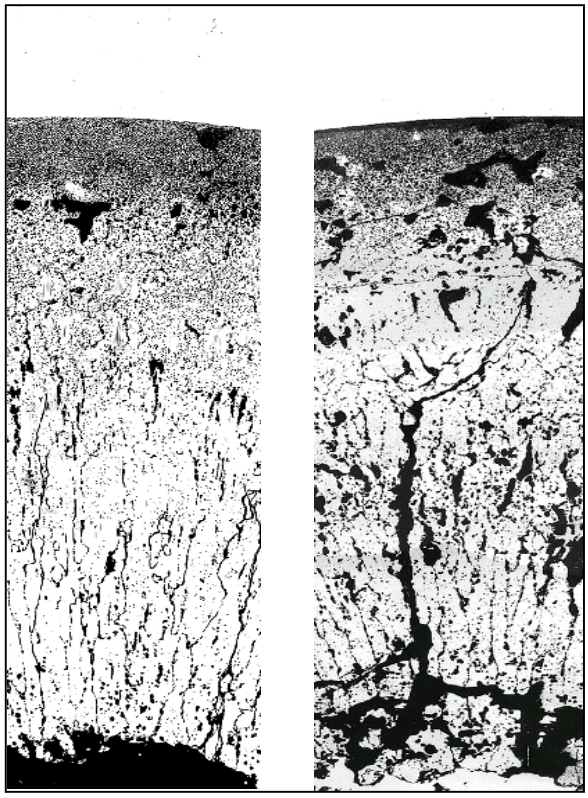


+145 mm (relative to the Midplane)



+90 mm (relative to the Midplane)

Figure 5. **Microstructure of $UNpO_2$ irradiated fuel and fuel-cladding boundary** (B=12.5%)

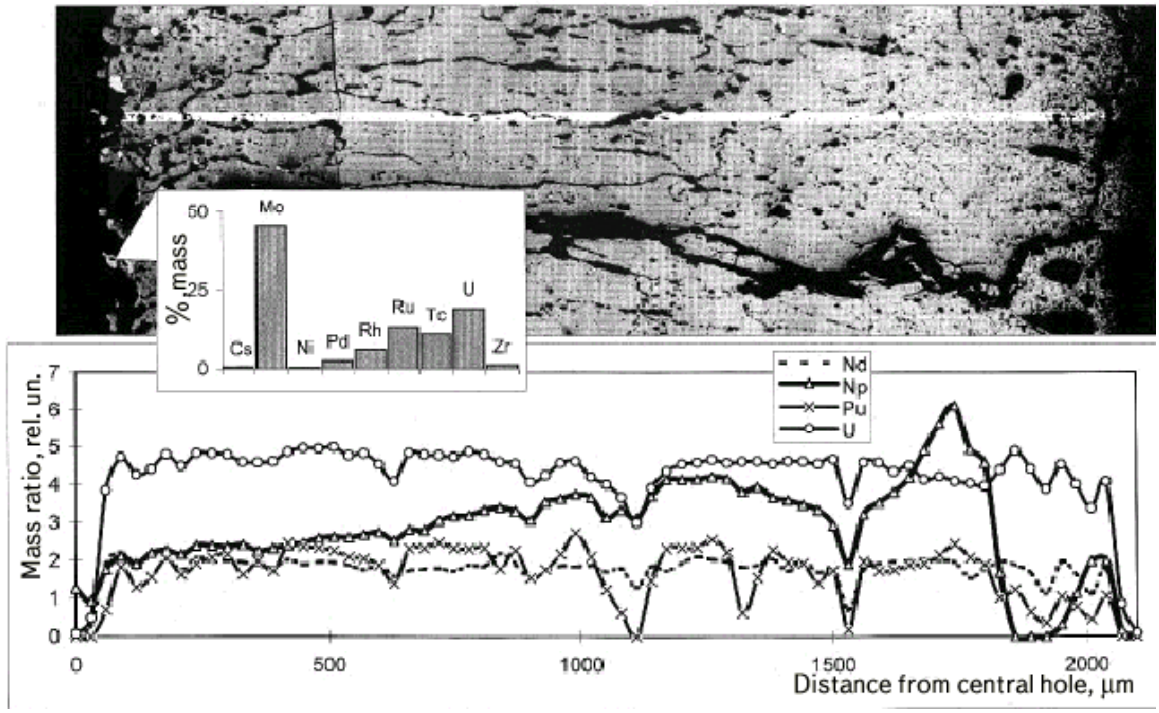


Midplane

Table 1. **Main characteristics of Am containing fuel batch**

# of batch	Weight, g	Content, %wt.			Oxygen coefficient, O/M	Pycnometer density, g/cm^3
		U	Pu	Am		
1	38.4	34.8	50.8	2.1	1.989	9.71
2	42.6	77.6	7.6	3.0	1.991	9.80
3	94.6	81.9	2.6	3.2	1.984	9.80

Figure 6. **Elements distribution along the fuel pin radius** (+28 mm relative to the Midplane) **and metal inclusion composition** (B=12.5%)



Minor actinides behaviour in SNF reprocessing

Within the framework of the DOVITA programme it was established that whole Np, as against Am and Cm, under the pyroelectrochemical reprocessing of irradiated oxide fuel can be returned to ABFR for reburning in structure as the $(U,Np)O_2$ or $(U,Pu,Np)O_2$ fuel.

Am has weak propensity to formation of oxychloride ions in melt. Therefore it can be partially entered into an oxide fuel matrix.

Cm does not take part in the pyroelectrochemical reprocessing of oxide fuel and it will collect in chloride melt together with REE and residual Am.

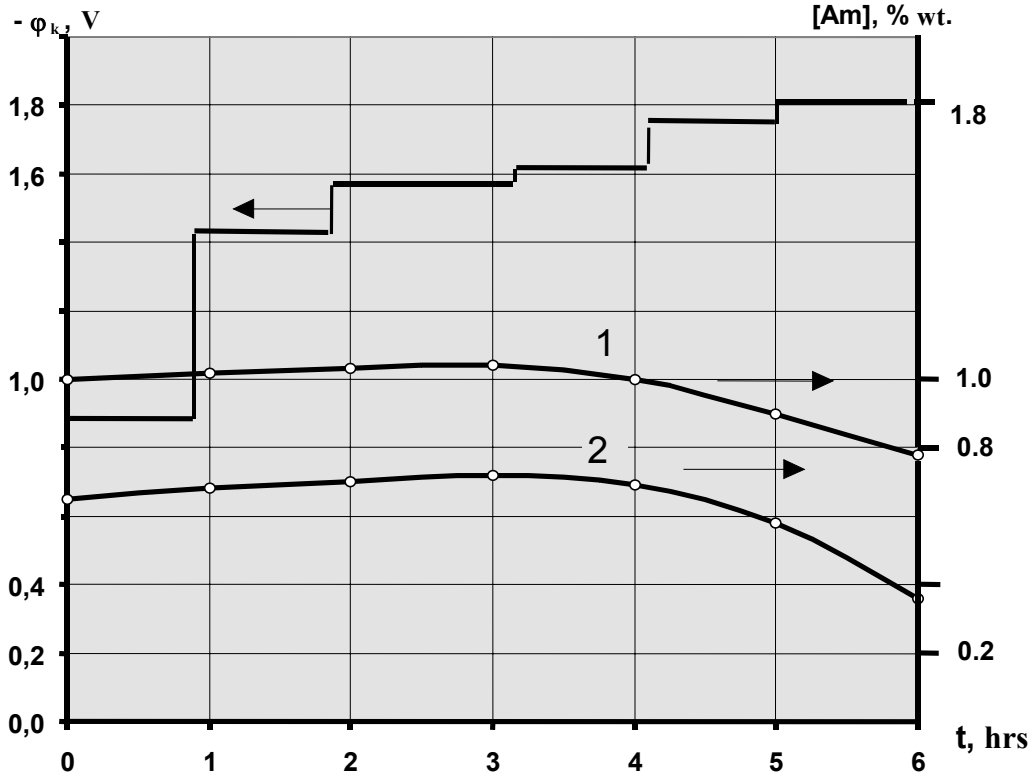
When developing technology of irradiated fuel regeneration, method of precipitating impurity phosphates is the basis for deep purification of the melt. Almost all the elements, having many charged (3+ and more) ions in the chloride melt, form difficult soluble phosphates. In this respect MAs are not exception.

This allows carrying out deep purification of the melt from considerable mass of impurities and minor-actinides as well, obtaining precipitation of phosphates mixture, which is americium and curium concentrate.

After extraction from melt the concentrate of Am and Cm can be used as the additive to pins with MOX-fuel or as a target for irradiation into ABFR.

Figure 7. Change of the Am content and cathode potential (-φk) during the (U, Pu, Am)O_2 electrodeposition in NaCl-2CsCl melt at 1 023K

(Uranium content in melt for both experiments (1 and 2): initial- 6.0%wt., final- 0.1%wt.)



Experiments on irradiated oxide fuel reprocessing

In SCC RIAR there have been performed demonstration experiments on pyroelectrochemical treatment of MOX-fuel with burn-up 4.7% h.a. for BN-350 reactor (1991-1992) and with burn-up about 21-24% h.a. for BOR-60 reactor (1995-1996). During the experiments MAs behaviour has been controlled. MAs distribution over products of reprocessing of BN-350 and BOR-60 reactors fuel is given in Table 1.

Thus, experiments results (Table 3) give agreement of Np, Am, Cm distribution over products of irradiated fuel reprocessing with being developed scheme of ABFR – DOVITA fuel cycle (Figure 1).

In addition, it is necessary to note, that the large work on development of special analytical methods was fulfilled for control of reprocessing and certification of fuel with minor-actinides, for MA control in fuel pin (Table 2).

Table 2. **MAs distribution over products of reprocessing BN-350 and BOR-60 reactors fuel, %**

Actinides	Deposits UO_2		Precipitate PuO_2		Phosphate concentrate	
	BN-350	BOR-60	BN-350	BOR-60	BN-350	BOR-60
Np	–	90.1	–	2.5	–	1.6
Am	9.7	3.6	39.4	18.1	45.8	73.1
Cm	–	3.9	–	0.0	–	91.4

The reactor irradiation of ampoules containing actinides isotopes

Two pins including eight ampoules containing pure actinides are irradiate in BOR-60 reactor since December 1998. The objectives of actinides irradiation are the adjustment of nuclear physical constants. In October 2001 the irradiation of one pin was finished. Initial and final calculated isotope compositions are presented in Table 3. Now irradiated ampoules are under investigations by radiochemical and mass-spectrometry methods.

Conceptual investigation related DOVITA programme

NPP with fast reactors by capacity up to 300 MWt (hypothetical reactor RBN-300 or group of BMN-170 reactors) is taken as demonstration model for evaluation of external fuel cycle. The choice of reactor is made only for evaluation of material balances and flows in the system as whole.

The general flow sheet of DOVITA fuel cycle is submitted on Figure 1. The flow sheet of fuel production and reprocessing provides homogeneous option for Np recycle in MOX fuel. This fuel can have the increased contents of Pu. It is preferable to introduce Am and Cm into special targets, thus their extraction from a concentrate after PUREX-process can be carried out within the DOVITA cycle framework.

Fuel recycle is carried out by the following flow sheets:

- The driven MOX fuel, after 15-20% burn-up, is underwent to decladding, crushing, vacuum reprocessing and repeated vibropacking (with additive of fresh fuel).
- After two cycles of irradiation, the basic fuel is reprocessed by pyroelectrochemical methods. U, Pu and Np (and portion of Am) come back to irradiation.
- The materials of targets with Am, Cm and REE are irradiated by 3-4 cycles with periodic decladding and treatment, then they are directed to pyrochemical process.
- The pyrochemical separation of Am and Cm from REE will be carried out in system with liquid metal electrode (in the concept, the process CRIEPI (Japan) [5] is quoted), thus the REE part is dumped only.

The ways for Cm utilisation are compared. SSC RIAR has experience of Cm burning in high-flux reactor SM and experience of its allocation from various targets /6/. It was shown, that is expedient to separate it from wastes and to destroy in systems with high-flux reactor. Thus the Cm burning efficiency is 85% after 500 eff.days irradiation, and valuable isotope ^{252}Cf producing rate is 1 mg from 1 g of Cm.

The study on fuel reprocessing for reactor burner includes two directions:

- Partial reprocessing (treatment) of the irradiated fuel.
- Pyrochemical reprocessing of the spent fuel.

Process of partial fuel reprocessing

The process of partial fuel reprocessing consists in the followings:

The irradiated fuel contained of minor-actinides, after 15-20% burn-up is underwent to mechanical decladding. Then the crushing of sintered fuel up to particles with dimensions necessary for vibropacking is carried out. This granulate is underwent to vacuum-thermal treatment for complete removal of gaseous and volatile FPs. The corrosion-active FPs (Cs, Te, Sb) are also partially removed

at the same time. The analysis and estimation of the indirect information on FPs removing from oxide fuel and experience on vacuum methods of salt removal from granulated fuel have allowed to prepare and to carry out the experiment. [10] On April-May 1997 the experiments with two kinds of fuel was carried out. The results are under analysis. It is planned further to make experimental fuel pins for the BOR-60 reactor from this fuel.

Table 3. **Isotope composition of actinides in ampoules**

Ampoule #	Isotope	Initial composition, μkg	Final composition, μkg	Burn up, % h.a.
№ 1	**^{232}Th**	419	409.2	**0.3**
	^{231}Pa	*	0.04	
	^{233}Pa	*	0.49	
	^{233}U	*	7.87	
	^{234}U	*	0.06	
№ 2	^{234}U	*	0.38	**4.9**
	^{237}Np	408	354.6	
	^{238}Pu	*	32.5	
	^{239}Pu	*	0.55	
№ 3 (monitor)	^{238}Pu	0.414	0.35	**15.2**
	^{239}Pu	109.02	88.7	
	^{240}Pu	5.35	7.6	
	^{241}Pu	0.207	0.75	
	^{242}Pu	0.0115	0.03	
	^{241}Am	*	0.07	
№ 4	^{238}Pu	0.227	0.2	**6.5**
	^{239}Pu	1.26	1.02	
	^{240}Pu	124.51	104.2	
	^{241}Pu	*	11.3	
	^{242}Pu	*	0.20	
	^{241}Am	*	0.9	
№ 5	**^{242}Pu**	126.42	115.5	**3.9**
	^{243}Am	*	5.72	
	^{244}Cm	*	0.23	
№ 6	^{234}U	*	0.08	**4.9**
	^{237}Np	*	0.6	
	^{238}Pu	*	7.39	
	^{239}Pu	*	0.12	
	^{242}Pu	*	1.94	
	^{241}Am	129	107.9	
	^{242m}Am	*	2.0	
	^{243}Am	*	0.07	
	^{242}Cm	*	2.4	
№ 7	^{238}Pu	*	0.15	**3.4**
	^{241}Am	2.678	2.27	
	^{243}Am	110.31	99.1	
	^{244}Cm	0.0113	6.9	
	^{245}Cm	*	0.13	
№ 8	**^{244}Cm**	129	103.9	**4.6**
	^{245}Cm	*	3.4	
	^{246}Cm	*	0.04	

* this isotope was absent in initial composition

The irradiation of second pin containing pure actinides is continuing now.

Data on fuel decontamination from cesium are given in Table 4. Cesium removal to the gas phase did not depend on fuel compound and burn-up. The relative low factor of cesium decontamination ($K_{decont.}$=2.0÷2.5) at 1 000°C in comparison with similar indexes at 1 550°C under thermal decladding of fuel pins evidences that cesium has the different compounds in fuel. [7]

Table 4. **Cesium removal from fuel under VTT**

Fuel compound	Fuel gamma-activity, GBq (%)				Activity output from fuel, GBq		Decontamination factor	
	starting		after VHT					
	total	^{137}Cs	total	^{137}Cs	total	^{137}Cs	total	as to ^{137}Cs
UO_2	552 (100)	518 (100)	274 (47)	241 (46)	278 (53)	277 (54)	2.1	2.2
UO_2 – 20%PuO_2 (mechanical decladding)	1070 (100)	563 (100)	735 (69)	224 (40)	335 (31)	339 (60)	1.5	2.5
UO_2 – 20%PuO_2 (electrochemical decladding)	1130 (100)	441 (100)	892 (79)	221 (50)	238 (21)	220 (50)	1.3	2.0

Behaviour of fuel components and minor-actinides. The analysis of α-spectra found a significant difference among spectra of samples for condenser and aerosol filters from those taken for fuel. Based on experimental data and calculations it was determined that plutonium, americium and curium went to the gas phase as aerosols in the ratio of ~1:4,5:7.0.

Physico-mechanical characteristics of fuel after VTT. Some characteristics of fuel after VTT are presented in Table 5.

Table 5. **Some fuel characteristics after VTT**

Parameter	Value	
	UO_2	**UO_2-20%PuO_2** mechanical decladding
Fluidity (G), kg/h	64.2	62.6
Density (ρ), g/cm^3		
bulk	4.73	4.81
pycnometrical	9.26	8.56

The fuel subjected to vacuum-thermal treatment is planned to use for production of the BOR-60 reactor experimental fuel pins. Further fulfilment of the programme will allow to carry out the scheme of short FR fuel recycling. This process is expected to be used for burning out some components in fast reactor. A partial reprocessing is considered in the frame of DOVITA programme, i.e. fuel cycle for minor-actinide burning out in fast reactor. [4]

Evaluation of pilot plant for ABFR fuel recycle (DOVITA plant)

On the basis of the above proposed flow-sheets and SSC RIAR experience on operation of Semi-Industrial Complex /15/, the layout and scheme of the Pilot Plant are developed for ensuring of the external ABFR fuel cycle. The primary evaluations of operational and capital expenses are carried out.

The basic industrial sites which are necessary in structure of the Plant should be located in the following number of the basic shielded cells and should be include the following installations:

- 1st cell: a) installation of spent FAs receiving and dismantling;
  b) installation for thermal and mechanical decladding of fuel.
- 2nd cell – installation for pyrochemical reprocessing of fuel, include:
  Site for reprocessing of the driven fuel;
  Site for separation of minor-actinides from fission products.
- 3rd cell: a)installation of partial (vacuum-thermal) reprocessing of fuel and
  b) site for reprocessed products treatment.
- 4th cell – fuel preparation to vibropacking.
- 5th cell – line on manufacture and control of fuel pins.
- 6th cell – line of manufacture and control of FA.
- 7th cell – wastes processing and preparation them to a storage and final disposal.
- 8th cell (laboratory) can be used for realisation of TRUEX-process for separation of pure Cm, and also for purification of noble metals from their concentrate received after pyrochemical reprocessing.
- 9th cell – cryogenic installation for chlorine purification and recycle.
- The analytical laboratory provides a chain of "heavy" glove-boxes.
- The systems of fissile materials monitoring and control and physical protection of building.
- Other auxiliary systems and facilities.

The Plant is designed for the following industrial capacity on manufacturing and reprocessing:

- 210-260 of FAs per year from core and 75-100 FAs per year from radial blanket of RBN-300, or 8-10 tHM/year from core and about 6-8 tHM/year from axial and radial blankets;
- up to 20 FAs per year with targets for irradiation of Am and Cm;
- the conceptual study on the Plant project is continued and the first stage is completed.

Conclusions

The DOVITA programme is not completed yet. However, the researches fulfilled during 10 years have shown, that the main part of problems connected with the ABFR fuel cycle can be solved. The complex decision of minor-actinide recycle problem is possible without creation of new exotic systems, and only on the basis of known technological methods and with application of known reactor systems.

The complex study on chemical aspects of the programme (manufacture and reprocessing of fuel) would be completed during nearest 2-3 years. It is planned to finish the demonstration reactor experiments during further 4-5 years.

The future R&D plans of SSC RIAR include a widening of researches on various fuel compositions for radiotoxical actinides transmutation.

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