ADVANCES IN REPROCESSING OF SPENT FUEL: PARTITIONING

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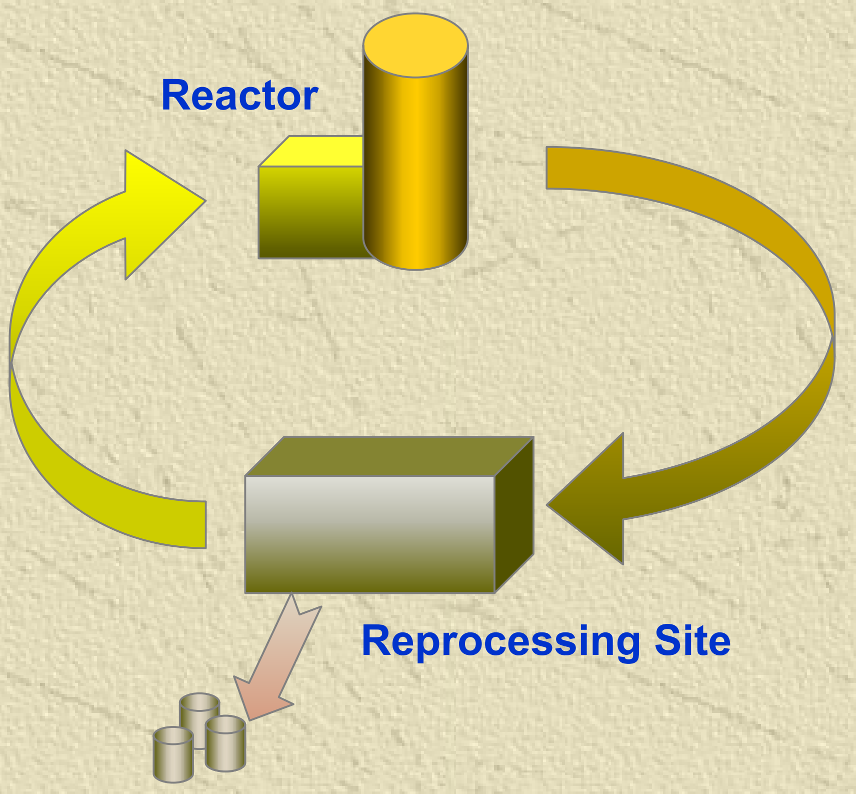
Fuel cycle in nuclear power engineering of the 21 century



Principles

- ***Closed fuel cycle***
- ***Optimization of technological system***
- ***Maximum level of inherent safety***
- ***Minimum quantity of wastes***
- ***Optimization of fuel cycle costs***

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Advanced Closed Fuel Cycle

Principles

- Optimization of technological systems
- Maximum level of “inherent” safety
- Minimization of wastes flows
- Optimization of fuel cycle cost

Requirements

- High level of safety
- Minimal impact on environment (radiation-migration equivalent for disposal of nuclear power engineering wastes)
- Maximum utilization of natural resources
- Non-proliferation

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Requirements of new technologies of the fuel cycle

The future technologies, which can significantly extend the oxide fuel potential and improve technical-economical indices of the fuel cycle with regard to the present-day requirements, are of great interest.

Non-aqueous methods are considered as possible alternative technologies for the closed fuel cycle.

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Main recent trends for application of molten salts

- Fast reactors and other advanced reactors
- Reprocessing of oxide spent fuel (MOX, Th-based, etc.)
- Reprocessing of metallic and nitride fuel (U-Pu and others)
- Partitioning of HLW (Ln/Ac separation)
- Recycling of Molten Salt Reactor (or ADS) fluid fuel

RIAR activities in the field of fuel cycle

Since 1964 RIAR has been performing large-scale investigations in the following research lines:

- ✓ Development of dry fuel production and reprocessing technologies for different nuclear reactor types
- ✓ Development and testing of the fuel rod designs for different nuclear reactor types
- ✓ Investigation of the ways for waste management generated during production processes resulting from dry technologies
- ✓ Investigation of the actinide and fission product properties in different ion liquids

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Pyrochemical processes

Basic research of the molten salt systems allowed for the development of technological processes for production of granulated uranium and plutonium oxides and mixed uranium and plutonium oxides. A distinctive feature of the pyrochemical technology is a possibility to perform all the deposit production operations in one apparatus - a chlorinator-electrolyzer

Pyrochemical reprocessing consist of the following main stages

- *Dissolution of initial products or spent nuclear fuel in molten salts*
- *Recovery of crystal plutonium dioxide or electrolytic plutonium and uranium dioxides from the melt*
- *Processing of the cathode deposit and production of granulated fuel*

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Current status of pyrochemical process in RIAR:

- Fundamental studies
- Technological development
- Industrial implementation

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Fundamental studies of pyrochemical process

- Properties of U, Pu, Th and Np in molten chlorides were studied in detail.
- The knowledge on physical chemistry and electrochemistry of basic FPs is enough for technological implementation
- Necessary direction for studies – chemistry of Am, Cm, Tc in molten chlorides
- Development of basis for new processes
    - Reprocessing of nitride fuel
    - Partitioning of HLW

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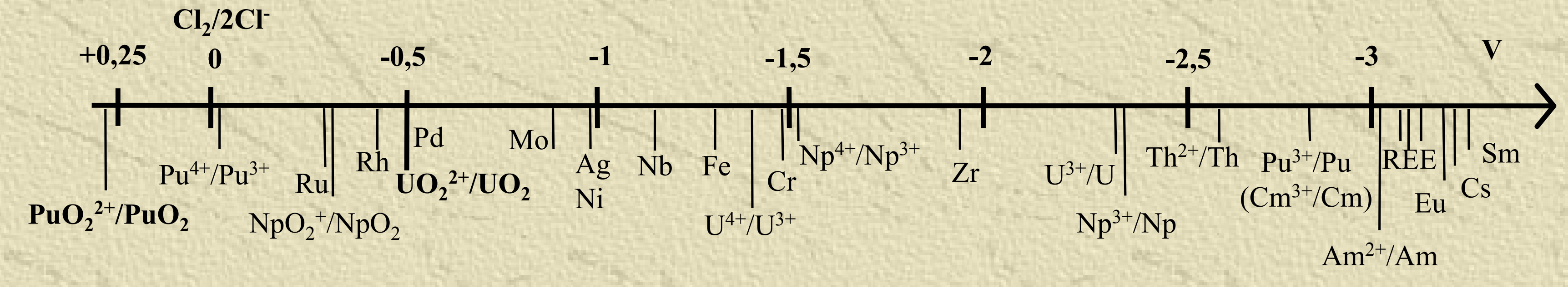
CHEMISTRY

UO_2, PuO_2, $(U, Pu)O_2$, MA, FP

Potentials row for actinides and fission products in molten 3LiCl-2KCl under 773 K



- From electrochemical point U and Pu oxides behave like metals. They are forming the complex oxygen ions MeO^{2n+}, which are reduced on cathode up to oxides.
- Under high temperatures (> 400°C) UO_2 are electrically conductive.
- In the molten alkali chlorides uranium has the stable ions U(III), U(IV), U(V), U(VI). Highest states of Pu oxidation Pu(V) and Pu(VI) are stable only in the definite field of ratios for oxidation reduction potentials of the system.

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Technological development

- For reprocessing and production of oxide fuel the main processes and equipment were developed and tested.
- Total amount of produced fuel is about 6000 kg (MOX, UO_2, special types). About 30 kg of fuel from the BOR-60 and BN-350 reactors was reprocessed. The basis of technology was created. The feasibility study was carried out for industrial plant for closed cycle of the BN-800 reactor.

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Pyroelectrochemical reprocessing of spent UO_2 and PuO_2 fuel

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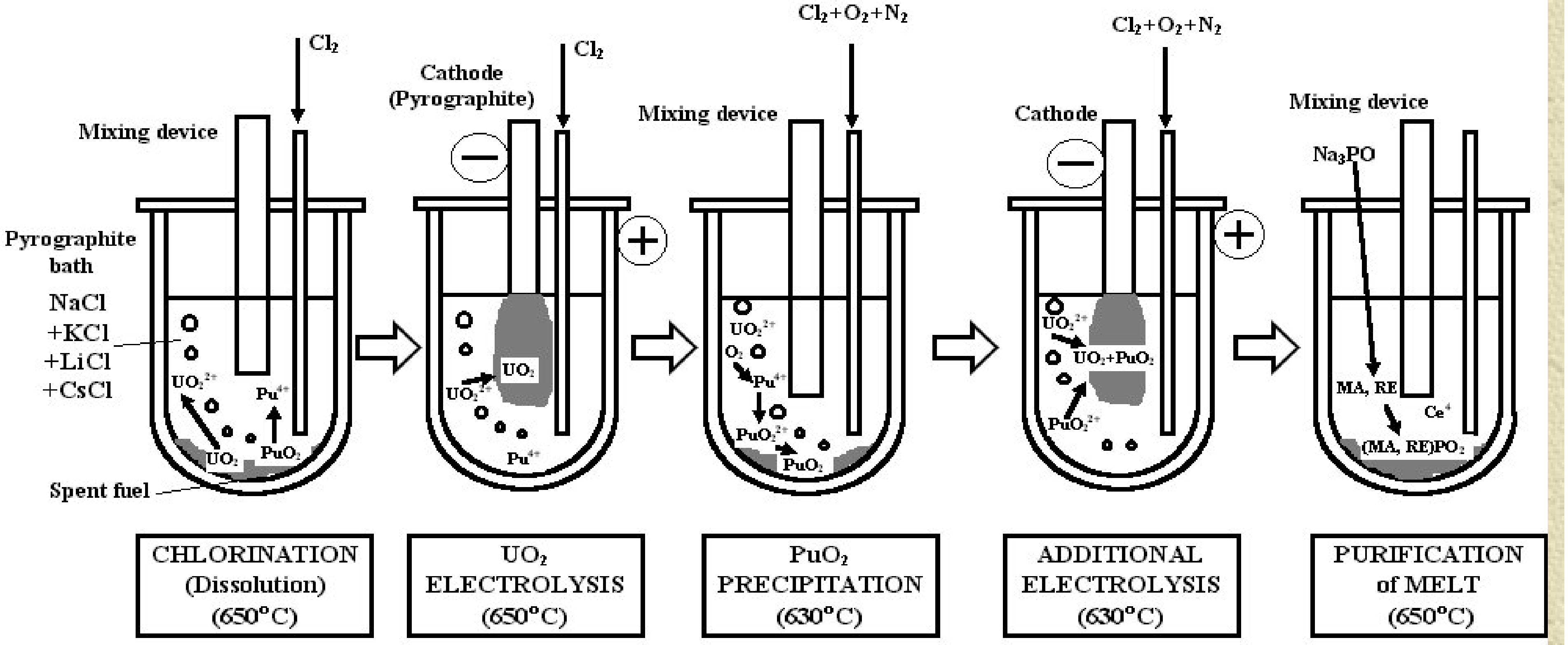
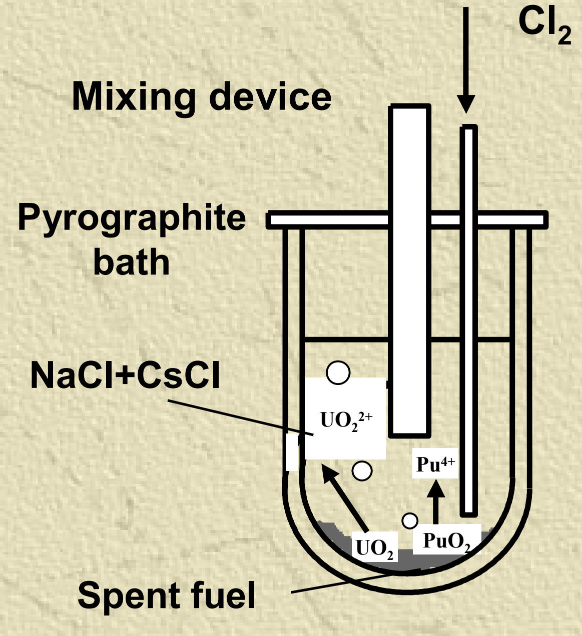


CATHODE: $UO_2^{2+}+2e^- \rightarrow UO_2$
ANODE : $2Cl^- \rightarrow Cl_2 + 2e^-$

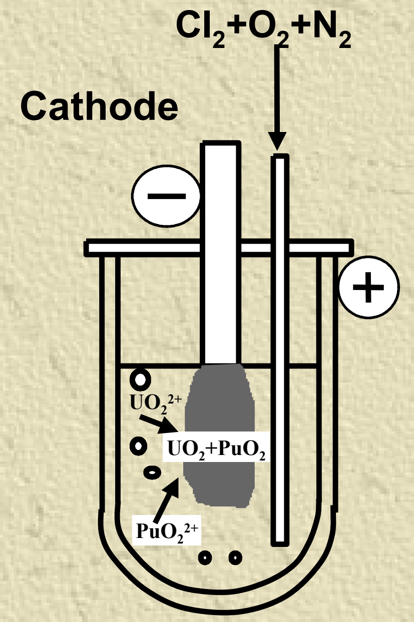
$Pu^{4+}+O_2+2Cl^- \rightarrow PuO_2^{2+} + Cl_2$
$PuO_2^{2+}+2Cl^- \rightarrow PuO_2+Cl_2$

CATHODE: $UO_2^{2+}+2e^- \rightarrow UO_2$
$PuO_2^{2+}+2e^- \rightarrow PuO_2$
ANODE : $2Cl^- \rightarrow Cl_2 + 2e^-$

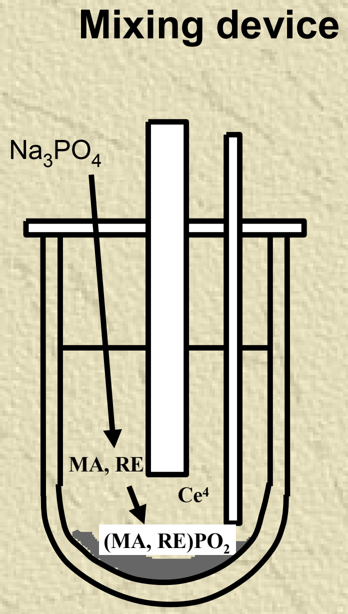
Pyroelectrochemical reprocessing of spent MOX fuel



Chlorination (dissolution)
(650°C)



Additional electrolysis
(630°C)



Purification of melt
(650°C)

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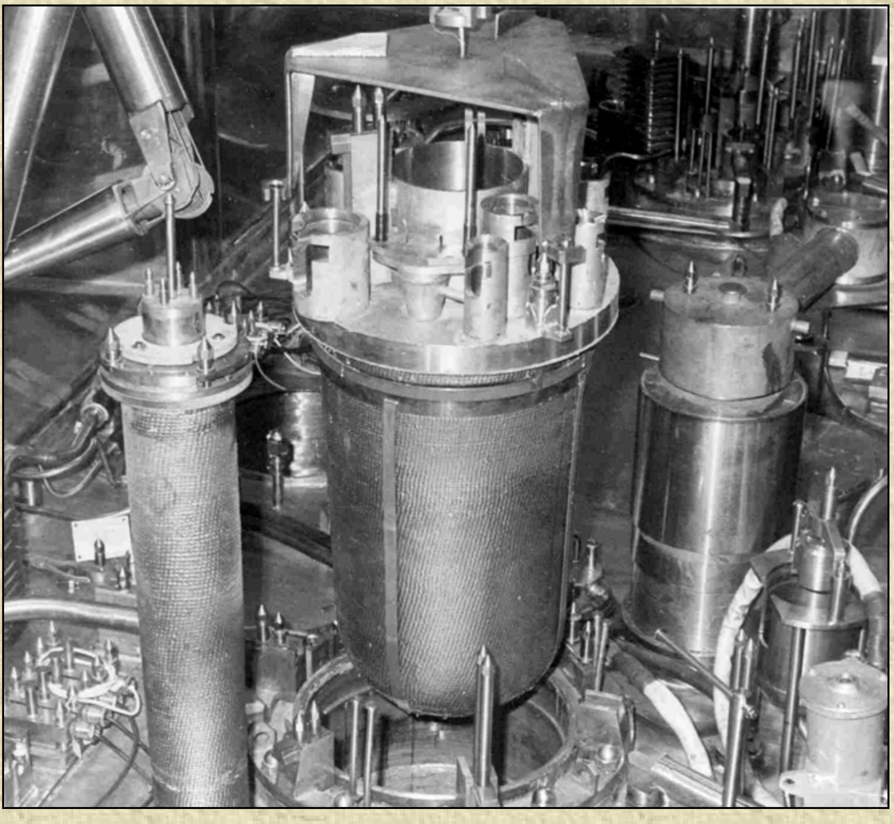
Main equipment for production MOX-fuel

6 kg MOX-fuel

Crucible diameter 250 mm

30 kg MOX-fuel



Crucible diameter 380 mm

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Granulated MOX-fuel particles obtained by pyroelectrochemical process

Particle density –	10,8 g/cm^3
O/M ratio -	2,0-2,03
Content of corrosion-active impurities, % no more:	
Carbon-	$18 \cdot 10^{-3}$
Fluorine-	$2 \cdot 10^{-3}$
Chlorine-	$7 \cdot 10^{-3}$
Total content of cationic impurities, % no more-	0,5

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Experience on pyrochemical reprocessing of the BOR-60 and BN-350 irradiated fuel

Fuel	Burn-up ,%	Mass, kg	Data of tests	Reactor
UO_2	7,7	2,5	1972..1973	BOR-60
$(U,Pu)O_2$	4,7	4,1	1991	BN-350
$(U,Pu)O_2$	21..24	3,5	1995	BOR-60
UO_2	10	5	2000	BOR-60
$(U,Pu)O_2$	10	12	2000-2003	BOR-60

PuO_2, UO_2 and MOX Decontamination factors (DF) from main FPs

Fuel type	Main FPs				
	Ru- Rh	Ce- Pr	Cs	Eu	Sb
PuO_2 for BN-350 (test, 1991)	50	220	> 3000	40	200
PuO_2 for BOR-60 (test, 1995)	33	40..50	4000	40..50	120
UO_2 for BOR-60 (test, 2000)	> 30	~	> 4000	> 200	~
$(U,Pu)O_2$ for BOR-60 (test, 2001)	20 - 30	25	~ 10000	> 100	~

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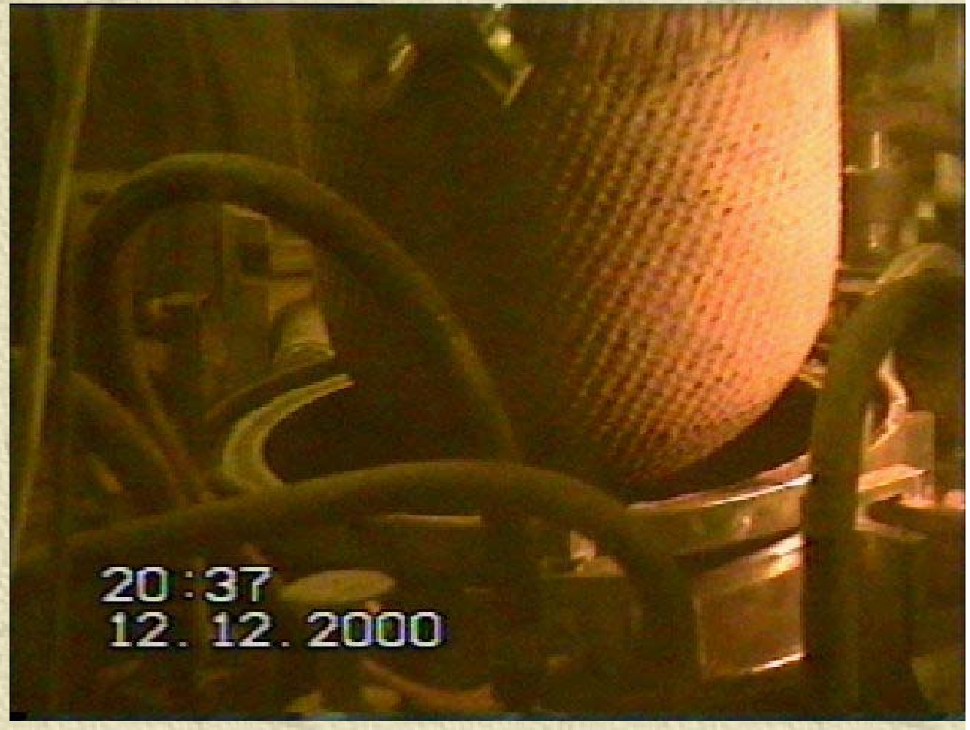
Experience on pyrochemical reprocessing of the BOR-60 irradiated fuel

MOX cathodic deposit

after pyro-process



Salt ingot after reprocessing test

with the BOR-60 fuel

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Pyrochemical processes

30 kg $(U,Pu)O_2$ deposit on cathode

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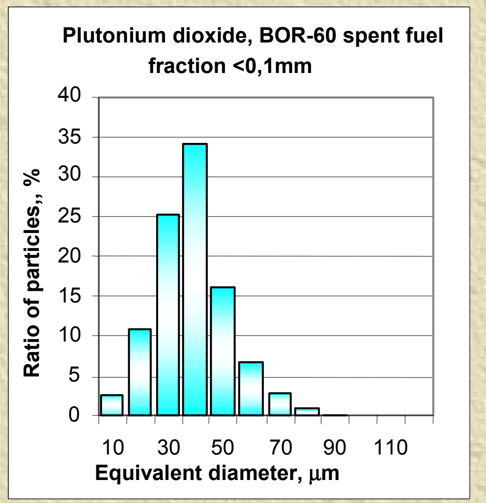
Pyrochemical reprocessing of irradiated fuel

The following experiments were carried out :

- **UO_2 SNF → granulated UO_2**
- **$UPuO_2$ SNF → granulated PuO_2**
- **$UPuO_2$ SNF → granulated $UPuO_2$**



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Demonstration experiment on UOX and MOX fuel reprocessing

Characteristics	Process $UO_2 \rightarrow UO_2$		Process $MOX \rightarrow MOX$	
	Test	For production line	Test	For production line
Yield of fuel component, %	95,91	> 99,6	94,83	>99,5
Fraction of U and Pu in recycled products, %	2,90	Will be recycled	3,90	Will be recycled
Technological losses, %	1,19	<0,4	1,27	<0,5
DF in Cs	10000		> 1000	
DF in REE	> 100		> 100	
DF in noble metals	~ 10		~ 10	

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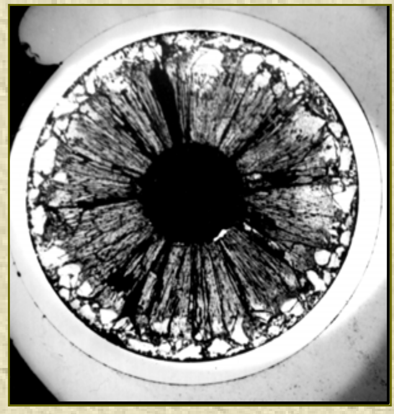
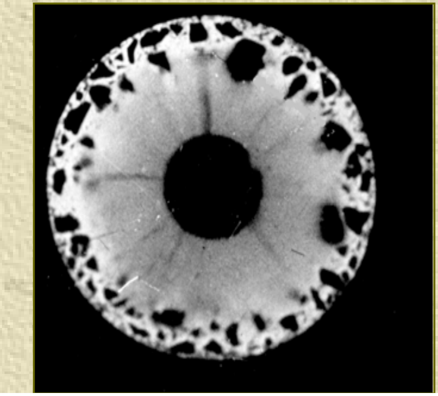
Recycle of reprocessed fuel in BOR-60

- Vipacked UO_2+PuO_2 fuel (mixture) achieved burnup ~ 15,1%,
- PIE were performed for fuel pins with a burn-up of 4,8 and 9,8 % h.a.
- Reprocessed MOX fuel was used for new fuel pins production and irradiation in BOR-60 started in June 2004





Microstructure and alpha-radiography of vibropacked fuel, the burnup of 9.8 %

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Methods for separation of MA and LLFP

During solid fuel reprocessing by molten halide salt methods:

- Cs, Sr, I remain in salt
- Np mainly precipitates together with U and Pu
- Am and Cm accompany REE
- Tc is collected as part of noble metals fraction

(Tc and I can migrate in gas phase during chlorination or fluorination)

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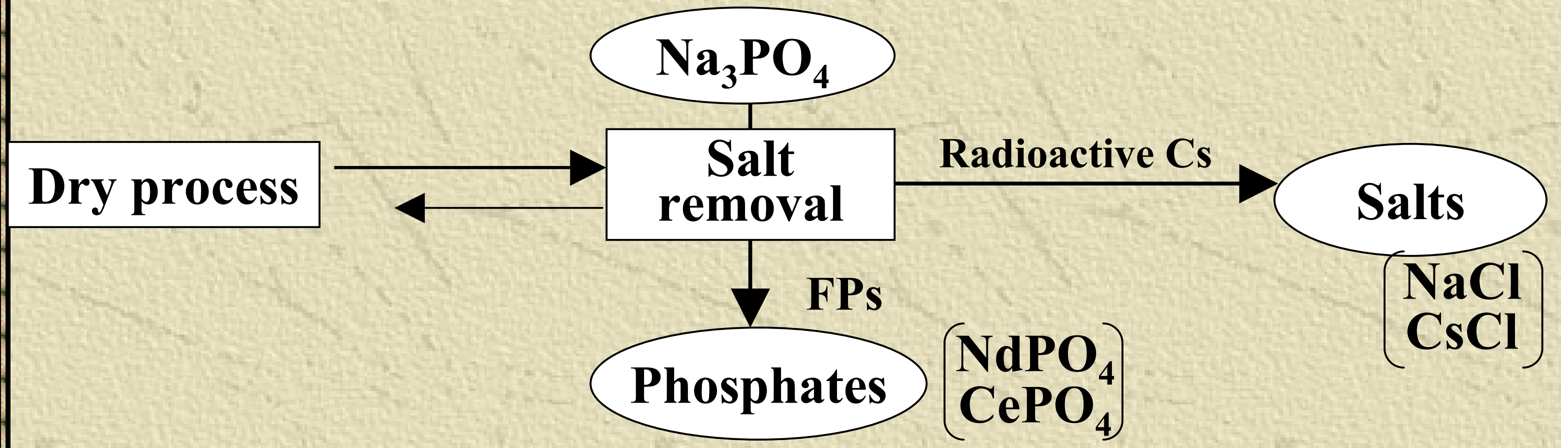
HLW flows after pyrochemical reprocessing



Na_3PO_4

Dry process

Salt removal

Radioactive Cs

Salts

$NaCl$
$CsCl$

FPs

Phosphates

$NdPO_4$
$CePO_4$

Wastes	Phosphates	Spent salt
Characteristics	Phosphates, contained FPs and other impurities	Alkali chlorides High activity, high heat release
Basic elements	11 wt. % Nd, 4,4 wt. % Ce	82 wt. % CsCl 18 wt. % NaCl
Expected amount	78 kg for 800 kg LWR fuel 43 kg for 300 kg FBR fuel	14 kg for 800 kg LWR fuel 7 kg on 300 kg FBR fuel

Experience on pyrochemical HLW treatment

Vitrification of pyrochemical HLW

Characteristics	Type of HLW		
	Phosphates	Spent salt	Phosphates + Spent salt
Glass-matrix	$Pb(PO_3)_2$ $NaPO_3$	$NaPO_3$, AlF_3 Al_2O_3	$NaPO_3$, AlF_3 Al_2O_3
Method for vitrification	vitrification, $T=950^0C$	vitrification without conversion, $T=950^0C$	vitrification without conversion, $T=950^0C$
Amount of wastes in glass, wt.%	28	20	36
Leaching rate of ^{137}Cs on 7 day, g/cm^2 * day	$7*10^{-6}$	$7*10^{-6}$	$4*10^{-6}$
Thermal stability, ^0C	400	400	400
Radiation stability	10^7 Gr (for γ & β)		10^{18} α-decay/g

Experience on pyrochemical HLW treatment

Ceramization of HLW arising from pyrochemical process

Characteristics	Type of high-level wastes	
	Phosphate deposit	Spent salt electrolyte
Type of ceramics	monazite	Cosnarite (NZP)
Method of introduction into ceramics	pressing, calcination , T=850^{0}C	Conversion to NZP from the melt or aqueous solution, pressing, calcination , T=1000^{0}C
Quantity of waste introduced into ceramics, %	100	30..40
Leaching rate of ^{137}Cs on 7-th day, g/cm2 * day	1*10^{-6}	3*10^{-6}
Thermal stability, ^{0}C	850	1000
Radiation resistance	5*10^8 Gy(for γ and β)	10^{19} α- decay/g

Other directions for Pyro-process application

Development of closed fuel cycle:

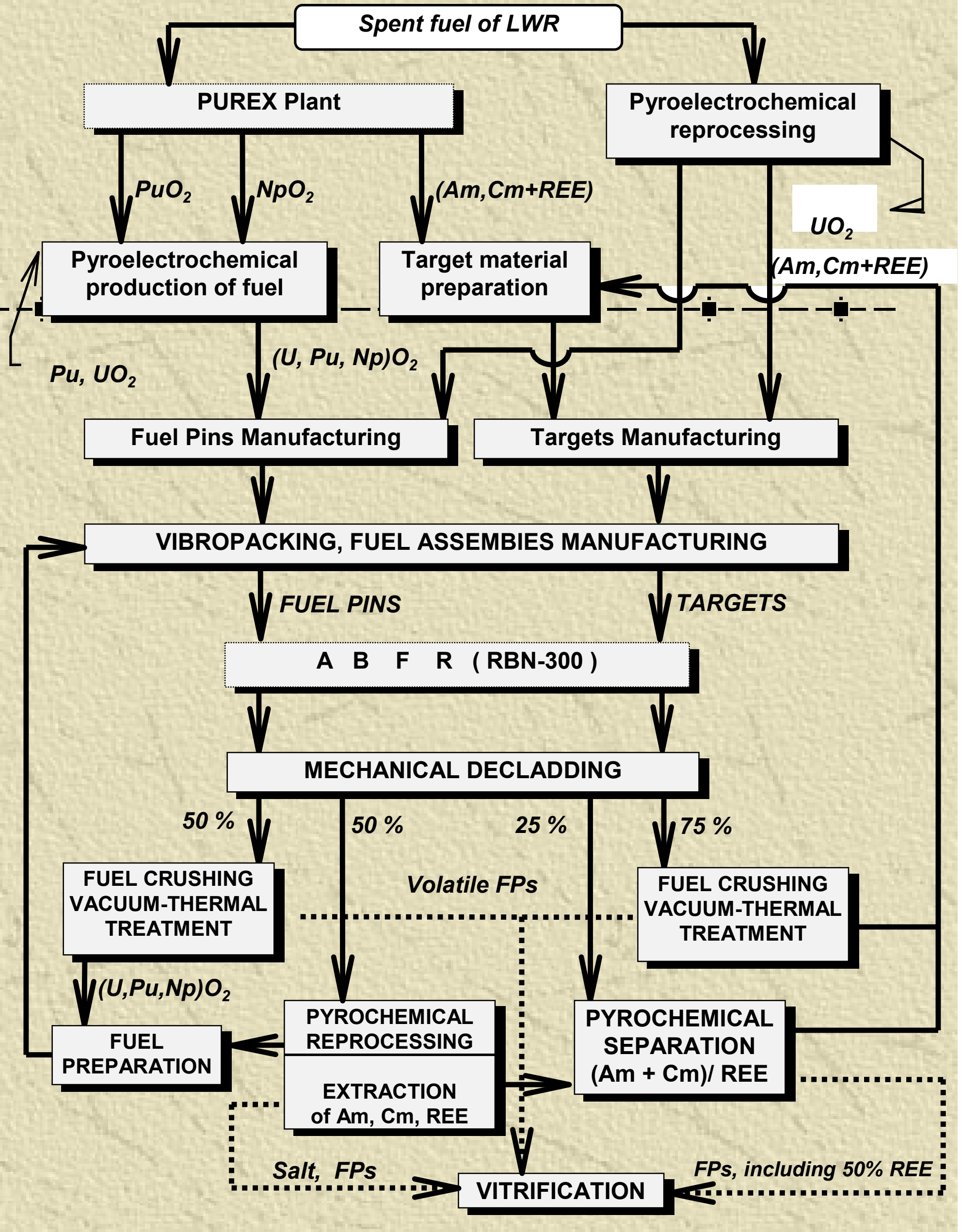
- Fuel cycle for actinide burner reactor – DOVITA
- Nitride fuel recycling for Fast reactors
- Molten salt reactor fuel recycling

Applied directions:

- Weapon plutonium conversion
- U-Al fuel reprocessing
- U-Mo fuel reprocessing

Fuel cycle of actinide burner reactor

DOVITA fuel cycle



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Activity on DOVITA Program

- (U, Np)O_2 fuel – pyrochemical production, irradiation and PIE (burn-up 12,5% and 20%).
- UO_2-20%PuO_2-(3-6)%NpO_2 fuel – pyrochemical production, irradiation.
- (U,Pu,Am)O_2 fuel - pyrochemical production, irradiation.
- Targets with Am for transmutation in the BOR-60 reactor – pyrochemical production, vibropacking, irradiation
- Pure actinide isotopes irradiation
- Behavior of Np, Am, Cm in pyrochemical processes.
- Study of Am electrochemistry in molten chlorides

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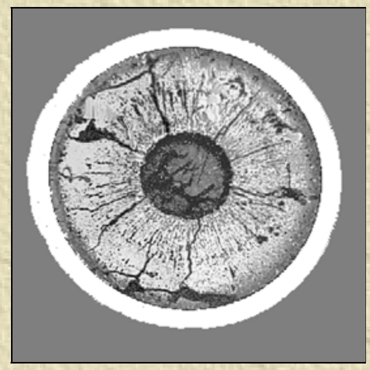
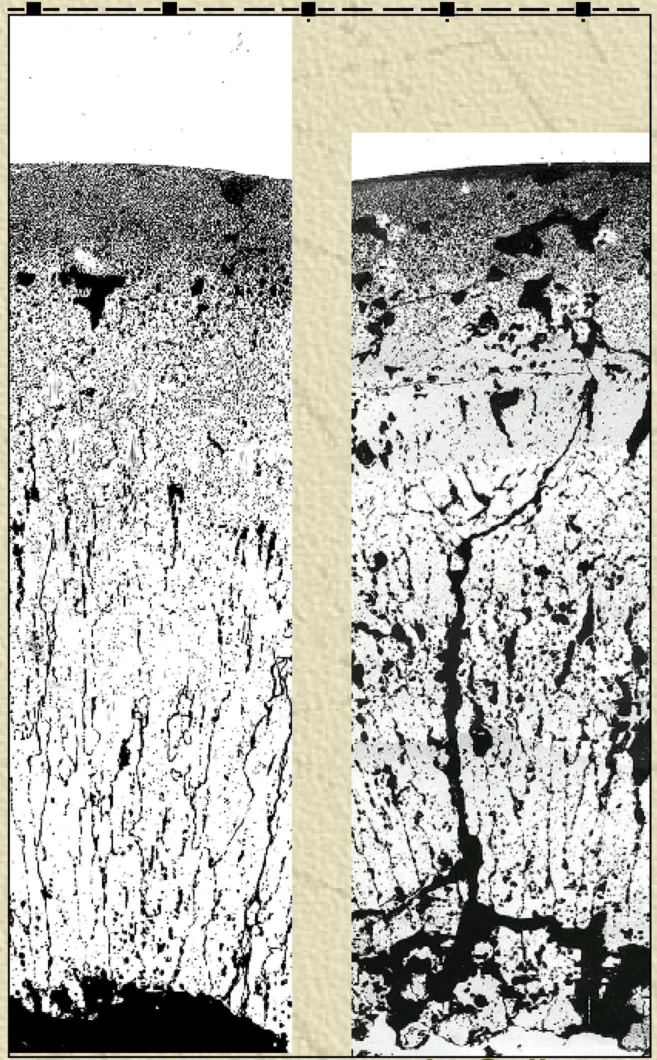
Micro- and macrostructure of irradiated fuel

$(U,Np)O_2$ (B=19,7%)

Upper plane



Middle plane



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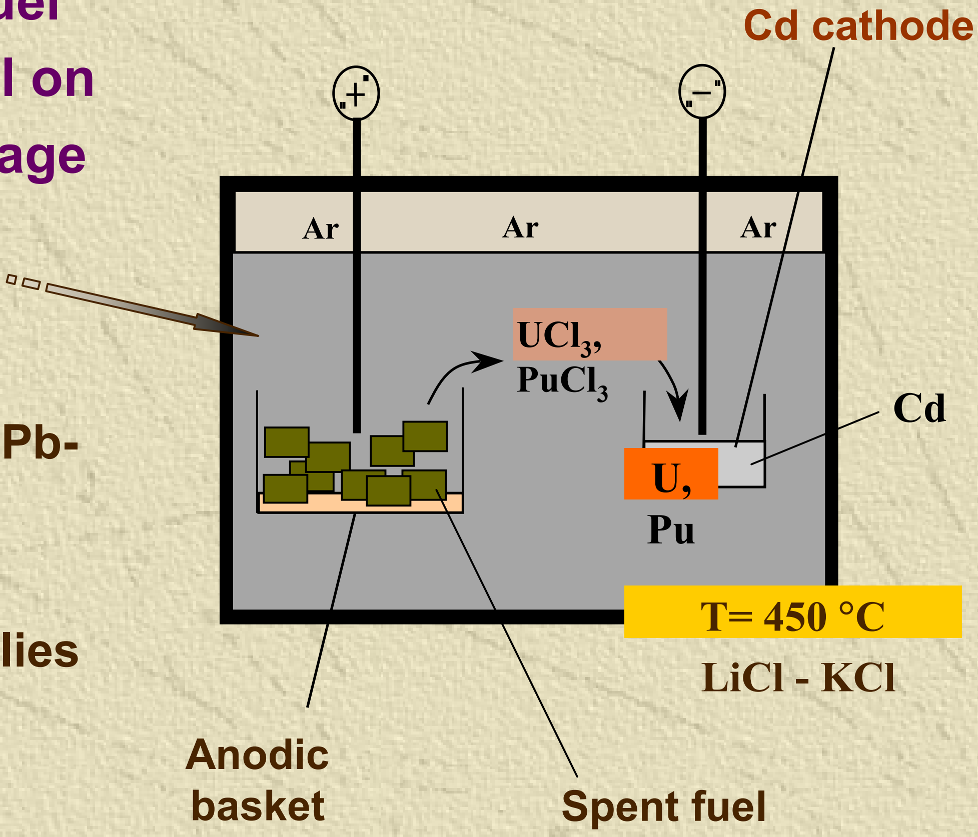
Pyrochemical reprocessing of mixed nitride fuel

- ✓Production of mononitride fuel from BREST reactor spent fuel on pyrochemical reprocessing stage
- ✓Mononitride pellets production
- ✓Manufacturing of fuel pins with Pb-bonding
- ✓Assembling of fuel sub-assemblies for BREST reactor



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Development level for Oxide Fuel Pyro-process recycling technologies

Method	Fundamental data	Laboratory testing	Pre-industrial testing	Industrial testing
Pyro-process	available	For fresh and spent fuel	Only with pure MOX	Started for BN-600
Vibro	available	For fresh and spent fuel	Only with pure MOX	Started for BN-600
Waste	Studies is continued	Studies is continued		
Remote equipment	Tested at ORYOL Facility for BOR-60 and BN-600			

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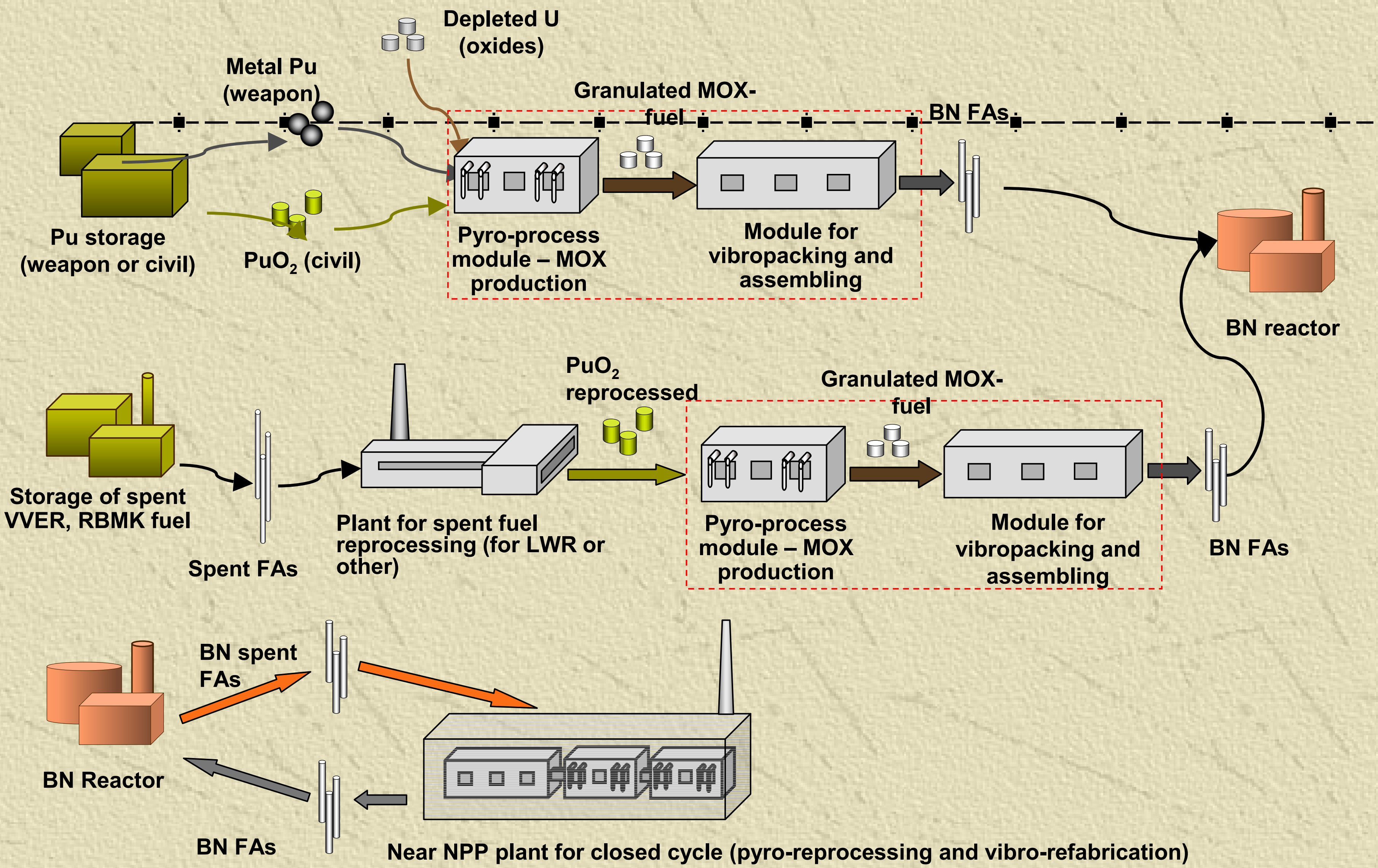
Current status of pyrochemical process in RIAR

(Industrial implementation)

- Since 1981 the BOR-60 reactor has been using vibropacked fuel produced by dry method.
- The test facility for closing of the BOR-60 fuel cycle is under design.
- BN-600 – 12 fuel assemblies with MOX fuel were irradiated, 3 LTAs are under irradiation.
- Semi-industrial facilities are under modernization for future production of 50 BN-600 MOX FAs per year. The re-start of semi-industrial facilities will be in 2005.

Implementation prospects



Depleted U (oxides)

Metal Pu (weapon)

Granulated MOX-fuel

BN FAs

Pu storage (weapon or civil)

PuO_2 (civil)

Pyro-process module – MOX production

Module for vibropacking and assembling

BN reactor

PuO_2 reprocessed

Granulated MOX-fuel

Storage of spent VVER, RBMK fuel

Spent FAs

Plant for spent fuel reprocessing (for LWR or other)

Pyro-process module – MOX production

Module for vibropacking and assembling

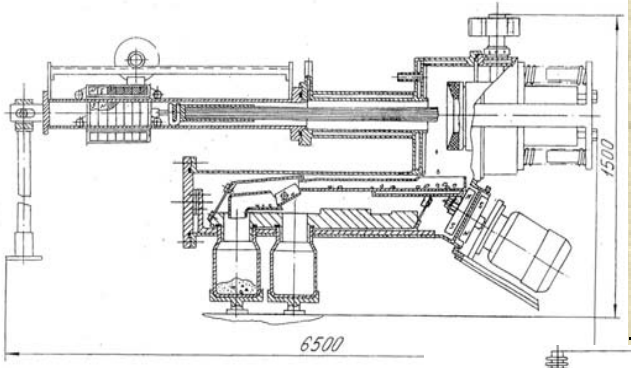
BN FAs

BN spent FAs

BN Reactor

BN FAs

Near NPP plant for closed cycle (pyro-reprocessing and vibro-refabrication)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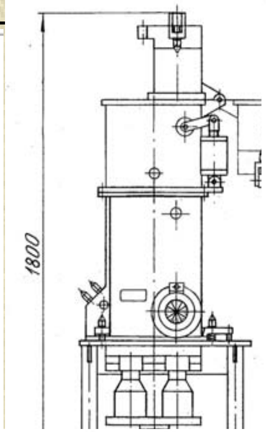
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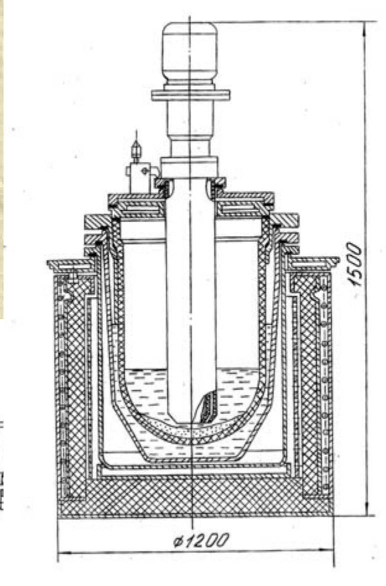
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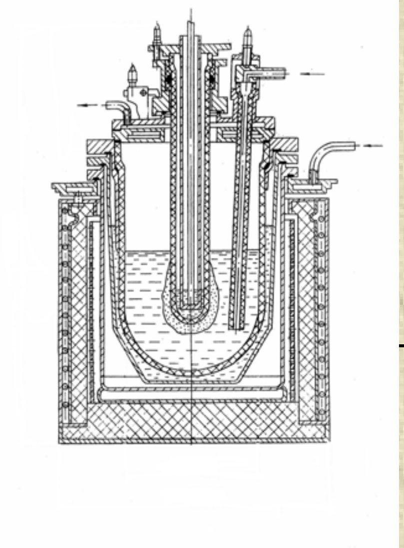

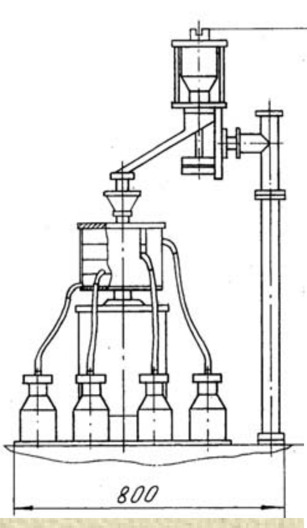
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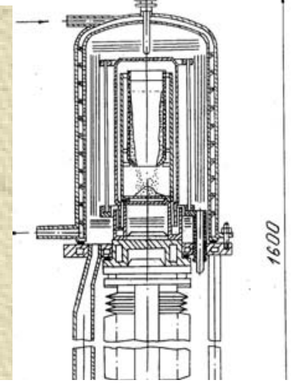
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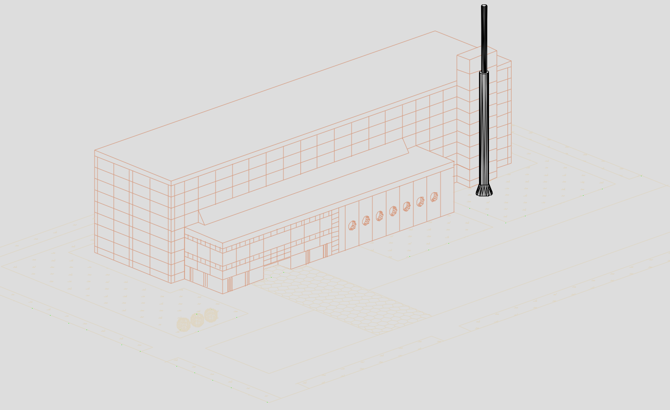

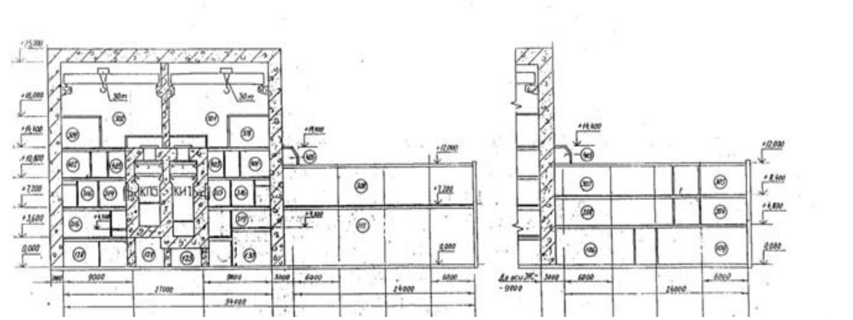
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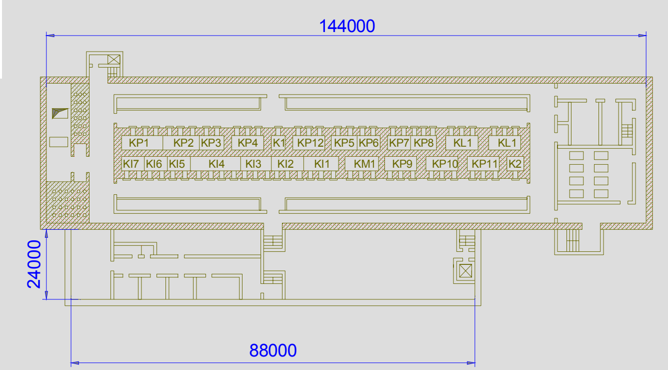
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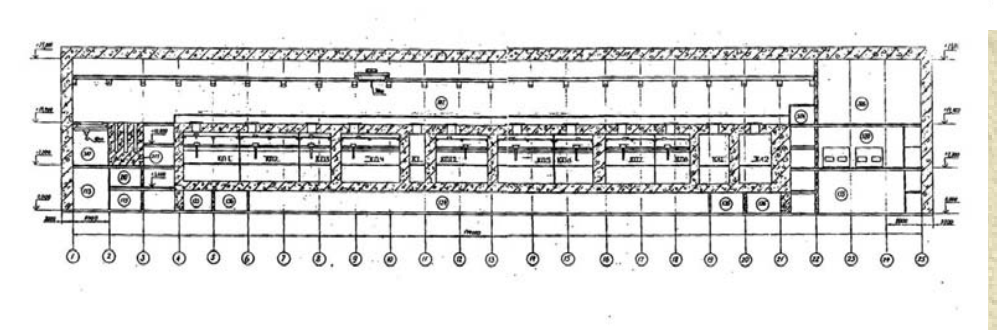

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Conclusion: ADVANCES IN REPROCESSING OF SPENT FUEL

Pyroprocess for reprocessing of spent fuel and vibropac technology can be used as basis for recycling and production of BN-type MOX fuel in different scenarios of future fuel cycles

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