Joint ICTP/IAEA School on Physics and Technology of Fast Reactors Systems

9 - 20 November 2009

Current Status of Development in Dry Pyroelectrochemical Technology of Spent Nuclear Fuel Reprocessing

Alexander Bychkov
State Scientific Centre Research Institute of Atomic Reactors
Dimitrovgrad
Russia

niiar@niiar.ru
CURRENT STATUS OF DEVELOPMENT
IN DRY PYROELECTROCHEMICAL TECHNOLOGY
OF SPENT NUCLEAR FUEL REPROCESSING

Alexander Bychkov
State Scientific Centre Research Institute of Atomic Reactors

Dimitrovgrad, Russia
E-mail: niiar@niiar.ru, Web site: http://www.niiar.ru
Russian Nuclear Power and Industry development is currently based on internal investment resources and on the two Federal Target-oriented Programs (FTP):

- FTP “Development of the Atomic Energy and Industrial Complex for 2007-2015 years” - **NPP construction**
- FTP “Assurance of Nuclear and Radiological Safety for year 2008 and till 2015” - **RAW Heritage**

FTP “New Generation Nuclear Energy Technologies” (2010-2020) – on final preparation Stage – and will support **Nuclear Innovations – New Technolgical Platform**, including:

- **Nuclear Closed Cycle on the basis of different fast reactors (Na, Pb, Pb-Bi)**
- **Dry technologies for recycling and new fuel**
- **New Experimental Facilities**
- **Fundamental studies, fusion developments etc.**
Forecast for Capacities of Russian Nuclear Power Industry till 2050

- Generation II extended for 15 years
- Generation I extended
- Generation III VVER
- BN-800
- BN-K

- Generation IV (BN) – growth according to the demand after 2020

- 14,4 GW for 10 years (1+7 units)

- 19 GW for 10 years

- 34,2 GW

- 41 GW

- 50 GW

- 13 GW for 5 years

- 19 GW for 10 years

- 13 GW for 5 years

- 41 GW

- 50 GW
New Technological Platform

Beloyarsk 3, BN-600

Na-related topic: BN-350, BN-600 (all facilities - 140 reactor-years)
Pb-related topic: floating reactors (Pb-Bi) on nuclear submarines
Mixed uranium-plutonium fuel
SNF reprocessing

Basic paths to arrange the architecture of CFC with fast neutron power units

Place of “fast reactor” in energy production/FC

Integral economics and ecology of electric energy production and FC

Selection of reprocessing technology

Complementary to VVER

Selection of coolant

Base for new construction

Light-metal (Na)

Heavy-metal (Pb)
New Technological Platform

Development of prototypes of CFC infrastructure key elements in Russia

- Dense fuel
- Fast reactor power unit
- SNF reprocessing unit
- VVER upgrade, power range

- Consolidation of competence and responsibilities for NTP
- Model of new power engineering architecture in economic logics
- Confirmation of technological readiness for industrial scaling

- 2014*: BN-800 and MOX fuel production startup
- 2014: Selection of base fast reactor technology and decision on VVER upgrade
- 2017: Establishment of “dense” fuel production
- 2018: Startup of SNF industrial reprocessing and RW disposal prototype
- 2019: Start of research reactor MBIR (RIAR) operation
- 2020: Construction of power unit prototypes
- 2026: Scaling of new technological platform

Development of prototypes of CFC infrastructure key elements in Russia

- Consolidation of competence and responsibilities for NTP
- Model of new power engineering architecture in economic logics
- Confirmation of technological readiness for industrial scaling

- 2014*: BN-800 and MOX fuel production startup
- 2014: Selection of base fast reactor technology and decision on VVER upgrade
- 2017: Establishment of “dense” fuel production
- 2018: Startup of SNF industrial reprocessing and RW disposal prototype
- 2019: Start of research reactor MBIR (RIAR) operation
- 2020: Construction of power unit prototypes
- 2026: Scaling of new technological platform
Development of New Technologies
- key projects

New Type of NPP Power Units

- Na Coolant Fast Reactors
  - BN-800
  - MOX fuel
  - BN-K

- Heavy Metal Coolant Fast Reactors
  - BREST
  - SVBR

- Closed Nuclear Fuel Cycle Technologies
  - Dense Fuel
  - SNF Reprocessing
  - RW Final Disposal

International Cooperation in the field of Energy Material Research

Fundamental Science

Multifunctional Fast Research Reactor MBIR

Research Base Development

Controlled Fusion Synthesis and Energy Conversion

Providing R&D
Conception of Federal Tasks Program
“New Generation Nuclear Energy Technologies”

RIAR – main experimental site

- Pilot Industrial Line for BN-800 (and BN-600) MOX Fuel Assemblies Manufacture
- Multi-functional Fast Test Reactor (MBIR) – 2018 → 2020 (loops)
- Large Multi-Purpose Pyrochemical Reprocessing Complex -2015
  - Molten salt Reprocessing Facility for different type of fuel
  - Fluoride volatility Reprocessing Facility
- New Lab for Experimental and Innovative Fuel Production – 2010-1012 (incl. Fuel and Targets with MA)
- Demonstration of Closing Fuel Cycle based on Pyrochemical technologies - 2016-2020 - ... on a levels:
  - Up to 50 spent FAs of BN-600/800
  - Full scale closed fuel cycle for MBIR started initial fuel loading
  - Other experimental implementations
- Open for International Programs
New Russian Sodium Fast Research Reactor – Multi-functional Fast Test Reactor (MFTR)

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum flux $\Phi_{\text{max}}$, n/cm²·sec</td>
<td>$\sim 6.0 \cdot 10^{15}$</td>
</tr>
<tr>
<td>Thermal power, MWth</td>
<td>$\sim 150$</td>
</tr>
<tr>
<td>Electric power, MWe</td>
<td>$\sim 50$</td>
</tr>
<tr>
<td>Number of independent experimental loops (≈1 MWth, sodium, heavy metal and gas coolant + salt coolants)</td>
<td>3 (+1 behind reactor vessel)</td>
</tr>
<tr>
<td>Driven Fuel</td>
<td>Vi-pack MOX, (PuN+UN)</td>
</tr>
<tr>
<td>Core height, mm</td>
<td>400-500</td>
</tr>
<tr>
<td>Maximum heat rate, kW/l</td>
<td>1100</td>
</tr>
<tr>
<td>Fuel Cycle</td>
<td>Full Scale Closed FC based on Pyro Processes</td>
</tr>
<tr>
<td>Test Fuel</td>
<td>Innovative Fuels, MA Fuels and targets</td>
</tr>
<tr>
<td>Maximum fluence in one year, n/cm²</td>
<td>$\sim 1,2 \cdot 10^{23}$ (up to 55dpa)</td>
</tr>
<tr>
<td>Design lifetime</td>
<td>50 year</td>
</tr>
<tr>
<td>RR creation time (no more than, years)</td>
<td>2018</td>
</tr>
</tbody>
</table>
BN-800 and demonstration of CFC

- Support and recovery of technologies providing the design of fast sodium reactors (engineering tasks, fabrication of special-purpose equipment for Na-related work, Na-handling procedures, etc.)

- Mastering of industrial technology for MOX fuel production

- Demonstration of operation using reprocessed MOX-fuel

OBJECTIVES

Demonstration of competitiveness of the BN-800 operation to provide the development of the competitive “fast reactor” topic

Development of effective FC economics within the future Nuclear Power and Industry Complex and NTP
Long-term Objectives

Complete implementation of long-term objectives is efficient in case of large-scale operation of fast reactors

Design and operation of fast neutron reactors with CFC as a basis for the overall scenario of nuclear power engineering development using recycled and bred fuel

Application of advanced Russian technologies of world-level priority and ahead-of-time development of national processing and technical base

Basic principles of CFC:

- To minimize costs on fuel reprocessing, fuel elements fabrication and waste management
- To minimize RW volume and, if possible, complete recycle of MAs for transmutation in this reactor system
- To eliminate usage of pure fissile material, accommodation of productions in hot cells and remote-operated boxes

Nowadays Russia has an absolute technological priority in this field proved by the active Russian/Japanese cooperation related to the new FR CFC technologies.

Russia has developed processes to be a basis for CFC of fast neutron reactors, i.e. inter-related and inter-compatible pyrochemical reprocessing and vibropacking of fuel pins.
Implementation Pyroprocess for BN-800 Fuel Cycle

Combination of pyroprocess and vibropacking technology is the basis for BN-type MOX fuel production and recycling in different scenarios.

Pu storage (weapon or civil)

Depleted U (oxides)

Metal Pu (weapon)

PuO₂ (civil)

Pyro-process module – MOX production

Granulated MOX-fuel

Module for vibropacking and assembling

BN FAs

MCC

Krasnoyarsk

RI AR

Dimitrovgrad

BN Reactor

BN spent FAs

In future:
For BN and BREST
For MOX and (U,Pu)N

BN-800 reactor

On site NPP pyro-reprocessing facility
Key technologies of the Fast Reactor closed fuel cycle with MOX fuel

- Pyroelectrochemical reprocessing (recycling through molten salt)
- Vibropacking technology (crystalline particles with getter)
- Remote controlled automated technologies for fuel pins and fuel assembly manufacturing
Spent Fuel Management Options

- **Direct Disposal**
  - SNF store
  - REACTOR

- **Classical Option**
  - SNF store
  - REACTOR

- **The ‘3rd Way’ Option**
  - SNF store
  - REACTOR

- **Reprocessing**
  - MOX or re-enriched U
  - Fabrication
  - Reprocessing

- **Transmutation**
  - MA
  - FP
  - Partial treatment
  - Partitioning
  - Conditioning

- **Recent Focus**: Optimize treatment system

- **Wastes**
REPROCESSING AS AN OPTION FOR SFM

EVOLUTION OF PURPOSE

- **PAST**: The classical option for spent fuel management (to recover pure fissile materials for recycle as MOX, especially in FBR)
- **PRESENT**: A fraction of spent fuel inventory recycled to thermal reactors as MOX and enriched UOX (mainly in LWR)
- **FUTURE**: Anticipation for innovative nuclear systems, P&T, simple recycling, etc

INDUSTRIAL MATURITY

- Currently, the only industrially available option (~1/3 of global inventory of spent fuel being reprocessed)
- Technical and/or infrastructural base for future applications to other futuristic options (i.e., P&T)
Advantages of the Pyroprocessing Option

- Proliferation Resistant (No Pu Separation)
- Sustainability (TRU Fuel for Gen IV)
- Economics (U,Pu Reuses)
- Environment friendly (U Separation & Low-Level Waste)
- TRU Transmutation (Reduce Toxicity)
- Volume Reduction Of High-Level Wastes
- Extension Of a HLW Disposal Facility life
Non-aqueous methods in nuclear power engineering

Non-traditional SNF reprocessing methods

- Pyroelectrochemical processes in molten salts
  - Oxide type Fuel
  - Non-oxide
- Fluoride-gas technology
- Supercritical fluid extraction in CO₂
- Ionic room-temperature liquids

Molten salts in nuclear power engineering

- SNF reprocessing
- Reprocessing and partitioning of waste
- Use as liquid fuel (MSR)
- Use as a coolant
R&D on SNF Pyro Reprocessing in a World

- Oxide SNF reprocessing into Oxide – RIAR (Russia), JAEA (Japan)
- Oxide SNF reprocessing into Metallic – CRIEPI (Japan)
- Nitride SNF reprocessing – JAEA (Japan), RIAR, Bochvar Institute (Russia)
- Metallic SNF reprocessing – ANL (US), CRIEPI (Japan)
- SNF metallization – KAERI (Korea), CRIEPI, RIAR
- HLW partitioning in molten salts – CRIEPI, RIAR, KAERI, CEA (France), ITU (EU)
- Fluoride volatility processes – CRIEPI, Hitachi/TEPCO (Japan), Kurchatov Institute (Russia), RIAR, INR (Rez., Cech. Rep.)
- Others applications (RIAR, ANL, CRIEPI, CEA)
Main stages of RIAR activity in the Field of FR Fuel Cycle


- Development of the pyrochemical technology for granulated fuel production and fuel rod fabrication by vibropacking
- Construction and operation of the remotely controlled facility ORYOL (fabrication of the BOR-60 fuel rods and FAs)
- **Pyrochemical reprocessing of spent nuclear reactor fuel**
- Loading of vibropack MOX fuel into the BOR-60 reactor
- Construction and operation of semi-industrial complex for the BN-600 fuel fabrication
- Pyrochemical production of granulated MOX fuel based on the weapon-grade plutonium
- Fabrication and irradiation of the experimental BOR-60 and BN-600 MOX FAs using weapon-grade plutonium
- Fabrication and irradiation of vibropack fuel rods based on the fuel compositions with minor-actinides additives
- Fabrication and irradiation of the experimental vibropack VVER fuel rods
Experience in Closed Fuel Cycle including pyrochemical processes

Research Institute on Atomic reactors – RIAR (Dimitrovgrad) is center of non-aqueous methods development:

- Pyrochemical investigation - from early 1960-s
- Demonstration of fluoride volatility reprocessing technology – 1970s
- Pilot facility for pyrochemical MOX-fuel production for fast reactor – from late 1970-s up to 1990-s
- Pyroelectrochemical reprocessing experience – 1990-2005
- Study on transmutation cycle, nitride fuel and others – from 1990s
- Expected creation of “Test Demonstration Center – Dry technologies” 2009-2013
Current status of pyrochemical development for oxide fuel

**Fundamental research**
Properties of U, Pu, Th, Np, Am have been studied. Knowledge of physical chemistry and electrochemistry of basic FP is sufficient for processes understanding and modeling. The needed research lines – study of Cm and Tc chemistry. Development of nitride fuel recycle methods is carried out.

**Development work**
All technological steps and equipment have been developed for the oxide fuel reprocessing and fabrication processes. The process was tested more than to **7200 kg of fresh fuel** for different reactors and up to **40 kg** of BN-350 and BOR-60 **irradiated fuel**. The essentials of technology have been elaborated and feasibility study has been completed for the BN-800 large-scale CFC plant. **More than 45 000 fuel pins and more than 1000 FAs**

**Industrial implementation**
As the readiness of technology is high, work is underway on industrial implementation of U-Pu fuel. The BOR-60 operates on vi-pack fuel. The design of the CFC facility is in progress. 30 FAs have been tested and irradiated in BN-600. **These technologies are under implementation as basic for BN-800 industrial MOX fuel production.**
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 consists 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**
CHEMISTRY

UO₂  PuO₂  (U, Pu)O₂  MA  FP

Potentials row for actinides and fission products in molten 3LiCl-2KCl under 773 K
Basis of “oxide electrolysis” in molten chlorides

- From electrochemical point U and Pu oxides behave like metals. They are forming the complex oxygen ions $\text{MeO}^{2n+}$, which are reduced on cathode up to oxides.
- Under high temperatures (> 400°C) $\text{UO}_2$ are electrically conductive.
- In the molten alkali chlorides uranium has the stable ions $\text{U(III)}$, $\text{U(IV)}$, $\text{U(V)}$, $\text{U(VI)}$. Highest states of Pu oxidation $\text{Pu(V)}$ and $\text{Pu(VI)}$ are stable only in the definite field of ratios for oxidation reduction potentials of the system.
- From any oxidation state plutonium can be converted into oxide by changing of the oxidation-reduction potential of the system.
- $\text{UO}_2$ and $\text{PuO}_2$ are reduced in the electropositive area. The majority of fission products are reduced at more negative potentials.
The potential/pO$_2^-$ (or potential of oxygen electrode) diagram of plutonium in NaCl-2CsCl melt at 863K

From any oxidation state plutonium can be converted into oxide by changing of the oxidation-reduction potential of the system.
Chlorination (dissolution) (650°C)

Cathode: \[ \text{UO}_2^{2+} + 2e^- \rightarrow \text{UO}_2 \]
\[ \text{PuO}_2^{2+} + 2e^- \rightarrow \text{PuO}_2 \]

Anode: \[ 2\text{Cl}^- \rightarrow \text{Cl}_2 + 2e^- \]

Sequence of operations for pyroelectrochemical reprocessing of spent fuel in MOX-fuel.
Pyrochemical processes

Oxide fuel deposit on cathode
а) Microstructure of (U,Pu)O₂ cathodic deposit (×10)

б) Microstructure of (U,Pu)O₂ and distribution of U, Pu, and Cl on scanning line
Technological process - periodical
Final products – granulated oxide fuel in the form of oxides of U and Pu or codeposited oxides of Pu and U
Equipment for pyroprocess

- Chlorator-electrolyzer
- Chlorator-settler with a device for removing bottom sludge PuO₂
- Crusher-grinder
- High-temperature vacuum furnace
- Cleaner
- Classifier

*Translation from Russian to English*

- Аппаратотмывкиотсолей
- Дробилка-измельчитель
- Высокотемпературнаявакуумнаяпечь
- Классификатор
TECHNOLOGY

MOX-fuel

6 kg MOX-fuel

Crucible diameter 250 mm

30 kg MOX-fuel

Crucible diameter 380 mm
**Mass balance on products**
(mass. %) for UO$_2$-PuO$_2$ granulation (on batch of 30 kg)

<table>
<thead>
<tr>
<th></th>
<th>Direct yield after basic</th>
<th>Recycled products</th>
<th>Losses</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>for direct return to process head</td>
<td>for return after treatment</td>
</tr>
<tr>
<td>U+Pu</td>
<td>97.90±0.30</td>
<td>1.31±0.08</td>
<td>0.57±0.03</td>
</tr>
<tr>
<td>U</td>
<td>97.87±0.34</td>
<td>1.36±0.07</td>
<td>0.56±0.03</td>
</tr>
<tr>
<td>Pu</td>
<td>98.04±0.15</td>
<td>1.08±0.11</td>
<td>0.59±0.06</td>
</tr>
</tbody>
</table>
DDP MOX → PuO₂ flow sheet

Fuel chlorination 700 °C

Preliminary electrolysis 680 °C

Precipitation crystallization 680 °C

Electrolysis-additional 700 °C

Melt purification 700 °C

DDP MOX → MOX flow sheet

Fuel chlorination 650 °C

Preliminary electrolysis 630 °C

Main MOX electrolysis 630 °C

Electrolysis-additional 630 °C

Melt purification 6500 °C
DDP Double Salt flow sheet

1st Salt:
- Fuel chlorination at 700 °C
- Preliminary electrolysis at 680 °C
- Fast Precipitation crystallization at 680 °C
- Fast Electrolysis at 700 °C
- Melt purification at 700 °C

2nd Salt:
- Fuel chlorination at 650 °C
- Main MOX electrolysis at 630 °C
- Melt purification at 6500 °C

Draft purification from captured salt

After accumulation of impurities during some cycles
RIAR experience in reprocessing of spent fuel of the BOR-60 and BN-350 reactors

### Fuel type

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>Burn up, %</th>
<th>Mass, kg</th>
<th>Period</th>
<th>Reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td>UO2</td>
<td>7,7</td>
<td>2,5</td>
<td>1972..1973</td>
<td>BOR-60</td>
</tr>
<tr>
<td>(U,Pu)O₂</td>
<td>4,7</td>
<td>4,1</td>
<td>1991</td>
<td>BN-350</td>
</tr>
<tr>
<td>(U,Pu)O₂</td>
<td>21..24</td>
<td>3,5</td>
<td>1995</td>
<td>BOR-60</td>
</tr>
<tr>
<td>UO2</td>
<td>10</td>
<td>5</td>
<td>2000</td>
<td>BOR-60</td>
</tr>
<tr>
<td>(U,Pu)O₂</td>
<td>10</td>
<td>12</td>
<td>2000...2001</td>
<td>BOR-60</td>
</tr>
<tr>
<td>(U,Pu)O₂</td>
<td>16</td>
<td>5</td>
<td>2004</td>
<td>BOR-60</td>
</tr>
</tbody>
</table>

### Decontamination factors (DF) from main FPs

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>Main FPs</th>
</tr>
</thead>
<tbody>
<tr>
<td>PuO₂ for BN-350 (test, 1991)</td>
<td>Ru-Rh</td>
</tr>
<tr>
<td>PuO₂ for BOR-60 (test, 1995)</td>
<td>50</td>
</tr>
<tr>
<td>UO₂ for BOR-60 (test, 2000)</td>
<td>33</td>
</tr>
<tr>
<td>(U,Pu)O₂ for BOR-60 (test, 2001)</td>
<td>&gt; 30</td>
</tr>
<tr>
<td></td>
<td>20 - 30</td>
</tr>
</tbody>
</table>

---
Mass-balance of Actinides and FP after the BOR-60 Fuel Reprocessing (1995)

Element's Distribution among Products

Element Fraction of total, %

Element: U, Pu, Np, Am, Cm, Ru, Sb, Cs, Ce, Eu

- UO2-1
- PuO2
- UO2-2
- Phosph.
- Spent El.
- Av.Salts
- Samples
- Sublimate
MOX-MOX reprocessing experiments

2004
Main MOX - 3 400 g,
Pu content - 30 %wt.
Current efficiency – 35 %

2000
1st Main MOX - 3 200 g,
Pu content - 10 %wt.
Current efficiency – 15 %
MOX cathodic deposit just after pyro-process

4:07
Salt ingot after reprocessing test with the BOR-60 fuel
Granulated MOX-Fuel

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metal content, %wt</td>
<td>87.75</td>
</tr>
<tr>
<td>Pyknometric density of granules, g/cm³</td>
<td>10.7</td>
</tr>
<tr>
<td>Bulk density of polydispersed granulate, g/cm³</td>
<td>6.0</td>
</tr>
<tr>
<td>O/M ratio (oxygen ratio)</td>
<td>2.00±0.01</td>
</tr>
<tr>
<td>Mass fraction of process impurities, %:</td>
<td></td>
</tr>
<tr>
<td>chlorine – ion</td>
<td>0.006</td>
</tr>
<tr>
<td>carbon</td>
<td>0.006</td>
</tr>
</tbody>
</table>
### Parameters of demonstration experiment on reprocessing of spent UO₂ and MOX fuel

<table>
<thead>
<tr>
<th>Parameter</th>
<th>UO₂ → UO₂ process</th>
<th>MOX → MOX process</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Experiment</td>
<td>Continuous-running process</td>
</tr>
<tr>
<td>Nuclear materials in target products, %</td>
<td>95,91</td>
<td>&gt;99,6</td>
</tr>
<tr>
<td>Nuclear materials in recyclable products, %</td>
<td>2,9</td>
<td>In process cycle</td>
</tr>
<tr>
<td>Loss, %</td>
<td>1,19</td>
<td>&lt; 0,4</td>
</tr>
<tr>
<td>Decontamination factor</td>
<td></td>
<td></td>
</tr>
<tr>
<td>cesium</td>
<td>10000</td>
<td></td>
</tr>
<tr>
<td>REE*</td>
<td></td>
<td>&gt;100</td>
</tr>
<tr>
<td>noble metals*</td>
<td></td>
<td>~10</td>
</tr>
</tbody>
</table>

* estimation based on the previous laboratory studies and demonstration experiments
Methods for separation of MA and LLFP

During solid fuel reprocessing by molten halide salt methods:
- Cs, Sr, I move to component of salt
- Np, mainly, move with U and Pu
- Am and Cm accompany REE
- Tc collected as part of noble metals fraction
  - (Tc and I can moved in gas phase during chlorination or fluorination)
### Waste treatment site

**Solid waste characteristics formed on MOX-facilities**

<table>
<thead>
<tr>
<th>Type of wastes</th>
<th>Specific amount, kg on 1 kg of fuel</th>
<th>Maximum specific activity, Bq/kg wastes</th>
</tr>
</thead>
<tbody>
<tr>
<td>On MOX-fuel facilities</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Phosphate concentrate</td>
<td>0.013</td>
<td>$1.5 \times 10^9$</td>
</tr>
<tr>
<td>Pyrographyte units</td>
<td>0.2</td>
<td>$2.3 \times 10^8$</td>
</tr>
<tr>
<td>All filters (cell and boxes)</td>
<td>0.13</td>
<td>$1.5 \times 10^8$</td>
</tr>
<tr>
<td>Other wastes (polyethelen, cloths etc.)</td>
<td>0.02</td>
<td>$1.0 \times 10^8$</td>
</tr>
<tr>
<td>On fuel pins facility</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cladding of defective fuel</td>
<td>0.04</td>
<td>$9.3 \times 10^7$</td>
</tr>
<tr>
<td>Other wastes</td>
<td>0.02</td>
<td>$1.0 \times 10^8$</td>
</tr>
</tbody>
</table>
**Pyrochemical Wastes treatment**

**Salt residue**

- Radioactive Cs

**Salt purification**

- Fission products
  - Phosphates
    - \( Na_3PO_4 \)
    - \( \text{NdPO}_4 \)
    - \( \text{CePO}_4 \)

**Basic elements**

- 11 wt.% Nd
- 4,4 wt.% Ce

**Quantity**

- <0,15 kg/kg of fast reactor SNF
- <0,03 kg/kg of fast reactor SNF

**Waste**

<table>
<thead>
<tr>
<th>Waste</th>
<th>Phosphates</th>
<th>Salt residue</th>
</tr>
</thead>
<tbody>
<tr>
<td>Special features</td>
<td>contain fission products</td>
<td>Alkaline metal chlorides, high activity, significant heat release</td>
</tr>
<tr>
<td>Basic elements</td>
<td>11 wt.% Nd 4,4 wt. % Ce</td>
<td>81,96 wt.% CsCl 18,04 wt.% NaCl</td>
</tr>
<tr>
<td>Quantity</td>
<td>&lt;0,15 kg/kg of fast reactor SNF</td>
<td>&lt;0,03 kg/kg of fast reactor SNF</td>
</tr>
</tbody>
</table>

Evaluations by Toshiba
# Vitrification of HLW from pyrochemical process

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>HLW type</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Phosphate precipitate</td>
</tr>
<tr>
<td>Glass matrix type</td>
<td>Pb(PO$_3$)$_2$ NaPO$_3$</td>
</tr>
<tr>
<td>Introduction method</td>
<td>vitrification, $T=950^\circ$C</td>
</tr>
<tr>
<td>Introduced waste amount, %</td>
<td>28</td>
</tr>
<tr>
<td>$^{137}$Cs leaching rate as of the 7th day, g/cm$^2$ * day</td>
<td>$7\times10^{-6}$</td>
</tr>
<tr>
<td>Thermal resistance, $^\circ$C</td>
<td>400</td>
</tr>
<tr>
<td>Radiation resistance</td>
<td>$10^7$ Gr (for $\gamma$ and $\beta$)</td>
</tr>
</tbody>
</table>
## Ceramization of pyroprocess HLW

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Type of high-level wastes</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Phosphate concentrate</td>
</tr>
<tr>
<td>Type of ceramics</td>
<td>monazite</td>
</tr>
<tr>
<td>Method of introduction into ceramics</td>
<td>pressing, calcination, $T=850^\circ\text{C}$</td>
</tr>
<tr>
<td>Quantity of waste introduced into ceramics, %</td>
<td>100</td>
</tr>
<tr>
<td>Leaching rate of $^{137}\text{Cs}$ on 7-th day, $\text{g/cm}^2 \times \text{day}$</td>
<td>$1\times10^{-6}$</td>
</tr>
<tr>
<td>Thermal stability, $^\circ\text{C}$</td>
<td>850</td>
</tr>
<tr>
<td>Radiation resistance</td>
<td>$5\times10^8 \text{ Gy (for } \gamma \text{ and } \beta)$</td>
</tr>
</tbody>
</table>

- **Phosphorus concentrate**: Monazite
- **Spent salt electrolyte**: Cosnarite (NZP)
Flow sheet of low-temperature chlorine recycle facility

1- combined condensers; 2- liquid nitrogen supply unit;
3- Drier – adsorption zeolite column; 4, 5- liquid nitrogen and chlorine evaporators;
off-gas (■), chlorine: solid (■), liquid (■) and gaseous (■); nitrogen: liquid (■) and gaseous (■);
spent coolant (■)
Concept scheme of OCRFM apparatus

(CL – Condenser of liquid-phase deposition; CS – condenser of solid-phase deposition)
Excluding of pure plutonium

For utilization of Pu in Fast reactors:
- High DF no necessary
- Additional treatment in closed cycle of Fast reactor
- Possibility of recycle of other TRU (Np, Am, Cm)
What we have now on Dimitrovgrad Dry Process (DDP) development?

- Three processes combined from the same operations (dissolution and recovery)
- Equipment and ways for improvement
- New understanding of MA behavior and methods for their partitioning
- Initial data on wastes (glass form and ceramic as additional option)
- Understanding of decladding procedures (but only old demo-tests)
- Methods for refabrication (some technical arrangements)
- Tested methods and new ideas for analytical control
Vibropacking technique

Fuel rods with granulated fuel are fabricated by vi-pack technique according to the standard procedures (in glove boxes or hot cells) that has been used at RIAR for 30 years.

The main advantages of the vi-pack technique and vibropacked fuel rods are the following:

- Simplicity of the process due to the reduced number of process and control operations, that makes the automation and remote control of the process easier.
- Possibility of usage of the granulate in any form; both in the form of a homogeneous composition and mechanical mixture.
- Reduced thermo mechanical impact of vi-pack fuel on the cladding (as compared with a pelletized core).
- More flexible requirements for the inner diameter of the fuel rod claddings.

To correct the oxygen potential in the fuel and eliminate the process impurities effect, *getter* based on U metal powder is introduced into the granulated fuel.
Production and testing of vibropacked fuel rods on the basis of MOX-fuel

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>Number of fuel assemblies</th>
<th>Burnur, max.%</th>
<th>Load, kW/m</th>
<th>Temperature, °C</th>
<th>Reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td>(U, Pu)O₂</td>
<td>Weapon grade, power grade</td>
<td>330</td>
<td>30,3</td>
<td>51,5</td>
<td>BOR-60</td>
</tr>
<tr>
<td>UO₂ + PuO₂</td>
<td>Weapon grade, power grade</td>
<td>132+20</td>
<td>14,8</td>
<td>45</td>
<td>BOR-60</td>
</tr>
<tr>
<td>(U, Pu)O₂</td>
<td>Weapon grade</td>
<td>30</td>
<td>10,5</td>
<td>46</td>
<td>BN-600</td>
</tr>
<tr>
<td>(U, Pu)O₂</td>
<td>power grade</td>
<td>4</td>
<td>development of the production technique</td>
<td>BN-600</td>
<td></td>
</tr>
</tbody>
</table>
Results of the material science studies of vibropacked fuel pins

**Micro- and macrostructure** of the cross section of the **BOR-60** fuel rod with **UPuO₂** fuel (the burnup of 32% h.a.) and **BN-600** fuel rod (the burnup of 10.5 %)
Experience in application of new technologies for the fuel cycle of the BOR-60 and BN-600 reactors

Semi-Industrial realization of remote controlled methods for manufacturing of fuel pins and assemblies of the BOR-60, BN-350 and BN-600 reactors:

- ORYOL facility (1977 - 1986),

25 year experience allow to formulate main principles for design and services of fuel and fuel pins production in remote controlled conditions.

The 3rd generation facility is under construction from 2001.
Experience in operation of automatic facility for the BOR-60 fuel rods and FAs fabrication

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>Mass, kg</th>
<th>Period</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\text{UO}_2) 90% enriched in (235\text{U})</td>
<td>98.85</td>
<td>1977...1987</td>
</tr>
<tr>
<td>((\text{U, Pu})\text{O}_2)</td>
<td>816.71</td>
<td></td>
</tr>
</tbody>
</table>

Irretrievable loss of the fuel composition during the whole period of the facility operation is 0.12 kg, that makes 0.013 per cent of the total amount of the fuel reprocessed at this facility.

Oxide fuel

<table>
<thead>
<tr>
<th>FAs production</th>
<th>Control of fuel rods with vi-pack fuel</th>
<th>Fabrication of fuel rods with vi-pack fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td>(K_1 = 0.98)</td>
<td>(K_1 = 0.85)  (K_2 = 0.97)</td>
<td>(K_1 = 0.93)  (K_2 = 0.95)</td>
</tr>
</tbody>
</table>

\(K_1\) – reliability factor (ORYEL 1)

\(K_2\) – reliability factor (ORYEL 2)
Equipment for the pyrochemical process and vibropacking

- Chlorator-electrolyzer
- Automatic fuel pins line
- Hot shielded box
- Assembling line
Recycle of reprocessed fuel in the BOR-60 reactor in vibrocompacted mode

- Fuel $\text{UO}_2+\text{PuO}_2$ (mechanical mixture) has burn-up about 17%,
  - Some fuel pins were under PIE (b.u.4,8 - 9,8 %)
- MOX reprocessed fuel used for new fuel pins production in 2002 and under irradiation in the BOR-60 from 2004 (burnup 15% )
Other processes and fuels which are under R&D in RIAR

- Nitride fuel – recycling by pyro-process and simplified pelletizing
- Metallic fuel (U-Pu-Zr, U-Al, U-Be) reprocessing
- RBMK Spent Fuel conditioning (metallization by Li/or electrochemical)

- Different fuels/targets with Np, Am, Cm
- Treatment of non-traditional fuel (coated particles (UN covered by W, U-Mo alloy, UC, Pu alloy, PuO₂ etc.)
- Molten salt fuel - (study of reprocessing and MA behavior)
“Investigation and evaluation of radiation and environmental safety of engineering processes of the nuclear fuel cycle at SSC RF RIAR” integrated task-oriented Program has been pursued since the 90-s.
Estimation of radiation safety of fuel cycle technological processes (RIAR)

- Direct recovery rate of fuel: 99.3...99.7%
- Losses of fuel: 0.1..0.2%
- Radiowastes volume: 0.2 kg/kg
- Safety (explosion, fire): high
- Possibility of accident per year: < 0.01 в год

Radiation doses

- Effective annual dose for Pu operation personnel: 0.60 mSv
- Effective collective dose: 0.06 man*Sv/year

Radiation effect on environment and population

- Effective total dose for population from realises of radionuclides through exhaust system: - 2.5 mkSv/year (2.5% of permitted level)
- Pu content in air, soil, river bottom precipitates, plants in 10..1000 times less of permitted level
Implementation Pyroprocess for BN-800 Fuel Cycle

Combination of pyroprocess and vibropacking technology is the basis for BN-type MOX fuel production and recycling in different scenarios.

In future:
For BN and BREST
For MOX and (U,Pu)N
Reprocessing Plant for Two BN-800
Reprocessing Plant for Two BN-800
Introduction into plutonium power energy and CFC.

1st Step – Weapon Pu disposition

- Conversion and production of granulated MOX fuel
- Controlled storage

- Reactor BN-800
- NIAR
- BAES
- OTVS BN-600
- OTVS BN-800
- BN-800 reactor
- BN-600 reactor

- Uranium (enriched)
- Weapon Pu
- Granulated (U, Pu)O₂
Introduction into plutonium power energy and CFC.

2nd step – semi-industrial testing of CFC elements with power grade Pu
Introduction into plutonium power energy and CFC.

3rd Step – Closed fuel cycle plant for recycling of BN-800 fuel with remote technologies

1 этап до 2020 года

In parallel: Krasnoyarsk EDRC is under operation:
- Testing of VVER and RBMK SNF reprocessing
- Utilization of new Pu (U-Pu-Np) in BN fuel
Introduction into plutonium power energy and CFC.

4th Step – Closing of BN-800 fuel cycle

2 этап после 2020года

Завод ЗТЦ

Переработка топлива, изготовление твэлов, ТВС

Decision on construction of VVER and RBMK SNF reprocessing plant
Concept of the closed fuel cycle Plant for reprocessing and production of (U,Pu)N fuel (for the BREST reactor)

Production of mononitride fuel from the BREST spent fuel at the stage of pyrochemical reprocessing
Production of mononitride fuel pellets
Fabrication of fuel rods with sublayer on the basis of pelletized fuel
Manufacturing of the BREST fuel assemblies

Hot cell design and infrastructure are similar as MOX recycling Plant
Since 1992 RIAR has been performing own R&D DOVITA Program

- Dry technologies for MA fuel reprocessing and preparation
- Oxide fuel application as the most widely studied one
- Vibropacking automated technology of the fuel pin production
- Integrated disposition of fuel reprocessing and fuel element refabrication facilities on the same site with the reactor
- TA The whole complex of approaches will permit a creation of the compact plant for Transmutation of Actinides
DOVITA fuel cycle

Fuel and fuel pins production

Targets production

FAs production

AFBR reactor

Partially treatment without reprocessing

Pyrochemical reprocessing

Homogeneous recycling

Inert matrix or UO₂

NpO₂

PuO₂

UO₂

\{Am, Cm\}O₂

[Am, Cm]O₂ + residual REE

[U, Pu, Np]O₂

Phosphate concentrate of FP's to preparing for storage

Used chloride salts containing of residual Cs-137 and Sr

Heterogeneous recycling
Recent RIAR activities in frame of DOVITA-2 Program

- R&D for production of vi-pack oxide Fuel with MA
  - Under continuation
- R&D for production of vi-pack nitride Fuel with MA
  - Under development
- R&D for production oxide pellet fuel with MA
  - Under development
- R&D for production metallic fuel with MA
  - Feasibility Stage
- Molten Salt chemistry of Am and Cm for fluorides
  - Under preparation
- Partitioning of MA/REE in molten chlorides/fluorides
  - liquid cathodes
  - reductive extraction
  - Oxide precipitation
Conclusions

- Basic studies on pyrochemical processes in molten chlorides are mainly completed.
- Different technological methods developed and tested for oxide and nitride fuel reprocessing and refabrication.
- Pyroelectrochemical technology for production of MOX vibropacked fuel for the BN-800 fast reactor is under implementation in Russia. Both types of plutonium – military and power grade civil - will be used for MOX fuel production.
- Dry technologies were chosen as basic advanced processes for the closed fuel cycle with the fast reactors.
- New facilities are under design and construction for investigation and demonstration of industrial closed fuel cycle with the fast reactors in Russia.
Thank you for your attention!

Alexander Bychkov
State Scientific Centre Research Institute of Atomic Reactors