Multiple Uranium Recycling with Re-enrichment in Gas Centrifuges Cascades as a Key Element of the Closed fuel cycle of light water reactors

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Technical Meeting on Strategies and Opportunities for the Management of Spent Fuel from Power Reactors in the Longer Timeframe
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Background
Background

- Slow development of Fast Nuclear Reactors (FNR)
- Inevitability of two-component structure of nuclear energy: Reactors on Thermal Neutrons (RTN) and FNR

- Necessity in new storages for spent (used) nuclear fuel
- Problems with providing fuel for RTN (mostly Light Water Reactors)

- Possible reaction – reprocessing of SNF and reuse of fissile materials from it
### Pros
- Significant reduce of SNF;
- Additional resources to provide fuel for RTNs.

### Cons
- Complexity and high cost of the SNF reprocessing procedure;
- Reprocessing technologies still under development;
- Reprocessing capacities are lower than dynamics of SNF accumulation.

#### Typical composition of SNF

<table>
<thead>
<tr>
<th>Component</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium</td>
<td>96%</td>
</tr>
<tr>
<td>Plutonium</td>
<td>3%</td>
</tr>
<tr>
<td>HAW (fission products)</td>
<td>1%</td>
</tr>
</tbody>
</table>
Russian experience in SNF reprocessing and recycling:

- Industrial technology of chemical reprocessing of the spent nuclear fuel with uranium and plutonium extraction and separation of highly active waste (RT-1);
- Monorecycling of uranium obtained from the spent nuclear fuel (LEU 2.8% for RBMK);
- Experimental technology of production mixed oxide uranium-plutonium fuel (MOX). The MOX-fuel production facility for the power unit with the BN-800 reactor was built.
- Russian plants produced fuel assemblies for reactor on thermal neutrons of European design. It was done for France and some other countries.

Now a lot of research are aimed on multi-recycling of spent nuclear fuel

The presentation is devoted to some aspects of multiple-recycling of uranium and plutonium in fuel of light-water reactors
At least 2 directions of reuse of fissile materials from spent nuclear fuel:

• Separation of U and Pu with direct usage of reprocessed U (RepU) for production of low-enriched uranium (LEU);

• Usage of U and Pu together in different types of mixed fuels (Mixed-Oxide Fuel (MOX), Regenerated Mixture of Uranium and Plutonium (REMIX)).
Recycling schemes
Multiple recycling of RepU in RUF

Cycle 1

NU → Enrichment → LEU → Nuclear Power Plant → SNF → Storage & Reprocessing → RepU

Cycle 2

RepU → Enrichment → LEU → Nuclear Power Plant → SNF → Storage & Reprocessing → RepU

Cycle 3...

Cycle N

RepU → Enrichment → LEU → Nuclear Power Plant → SNF → Storage & Reprocessing → RepU
Multiple recycling of RepU and Pu in REMIX

- Cycle 1:
  - NU → Enrichment → LEU → Nuclear Power Plant → SNF → Storage & Reprocessing → RepU & Pu

- Cycle 2:
  - RepU → Enrichment → LEU → Nuclear Power Plant → SNF → Storage & Reprocessing → RepU & Pu
  - NF

- Cycle 3...
  - Cycle N:
  - RepU → Enrichment → LEU → Nuclear Power Plant → SNF → Storage & Reprocessing → RepU & Pu
Enrichment of Reprocessed Uranium
• RepU is a multi-component mixture \((232\text{U}, 233\text{U}, 234\text{U}, 235\text{U}, 236\text{U}, 238\text{U})\);

• Re-enrichment process has many difficulties associated with minor isotopes \((232\text{U}, 234\text{U}, 236\text{U})\).

Table 1 – Isotopic compositions of RepU from different cycles

<table>
<thead>
<tr>
<th>Mass number</th>
<th>Concentration, %</th>
<th>RepU, Cycle 1</th>
<th>RepU, Cycle 5</th>
<th>NatU</th>
</tr>
</thead>
<tbody>
<tr>
<td>232</td>
<td></td>
<td>2.98 \times 10^{-7}</td>
<td>1.03 \times 10^{-6}</td>
<td>---</td>
</tr>
<tr>
<td>233</td>
<td></td>
<td>5.82 \times 10^{-7}</td>
<td>1.30 \times 10^{-6}</td>
<td>---</td>
</tr>
<tr>
<td>234</td>
<td></td>
<td>1.91 \times 10^{-2}</td>
<td>3.91 \times 10^{-2}</td>
<td>5.4 \times 10^{-3}</td>
</tr>
<tr>
<td>235</td>
<td></td>
<td>0.90</td>
<td>1.07</td>
<td>0.711</td>
</tr>
<tr>
<td>236</td>
<td></td>
<td>0.57</td>
<td>1.45</td>
<td>---</td>
</tr>
<tr>
<td>238</td>
<td>The rest</td>
<td>The rest</td>
<td>The rest</td>
<td>The rest</td>
</tr>
</tbody>
</table>
Impact on the radiation characteristics of nuclear fuel. 
**Consequence:** the content should be limited to the implementation of measures to ensure the radiation safety of personnel in the separation industry, as well as in factories for fuel fabrication. 
Typical restrictions $2-5 \cdot 10^{-7}$%

The reason of internal irradiation of personnel in separating plants and plants for production of nuclear fuel. In accordance with the international specifications of the LEU, the content should be limited.
Active absorber of thermal neutrons.

**Consequence:** the need to increase a level of $^{235}$U enrichment.

In general:

$$\Delta C_{235}^P = f \left(C_{236}^P\right)$$

$\Delta C_{235}^P$ - the value of the additional enrichment of $^{235}$U in the "fresh" fuel from the reprocessed uranium;

$C_{236}^P$ - concentration of $^{236}$U in “fresh” fuel from the reprocessed uranium.
Exposure (storage)

Recycling and Enrichment

Nuclear Power Plant

RepU or RepU + Pu: 0.90÷0.97 kg

1 kg

Natural Uranium

1 kg
• RepU enrichment is:
from the initial quantity of reprocessed uranium one should obtain low-enriched commercial uranium (LEU) of a given enrichment of $^{235}$U, for which restrictions on the concentrations of $^{232}$U, $^{234}$U and $^{236}$U isotopes are fulfilled, and the specified proportion between the RepU and fresh fuel is given.
Cascades of centrifuges for uranium enrichment

- Necessity in production of large amount of the product with necessary concentration of the target isotope;

- This task can be easily solved in consequent combination of connected in parallel centrifuges;
• This task cannot be solved with the ordinary cascade, which is used for natural uranium enrichment, because it is impossible to fulfill the $^{232,234}\text{U}$ limits while trying to enrich $^{235}\text{U}$.

• For RepU re-enrichment, it is necessary to modify the ordinary (triple-flow) cascade which is used for natural uranium enrichment.

• Several approaches were proposed so far (authored by scientists from Russia (NRNU MEPhI, UrFU, “SCP”, NRC “Kurchatov Institute’ and oth.), Japan, China).
1. Schemes on the basis of a three-flow cascade:
The RepU is diluted with products that do not contain artificial uranium isotopes at the inlet / outlet of the triple-flow cascade.

2. Schemes based on cascades with additional external flows (products, feeds):
The RepU is diluted with products that do not contain artificial uranium isotopes by supplying a diluent to one of the stages of the cascade. Diluents can be natural uranium, LEU, depleted uranium, etc.

3. Schemes based on double cascades:
Achieve concentration of $^{232}\text{U}$, $^{234}\text{U}$ and $^{235}\text{U}$ in different flows of double cascade.
1 – enrichment of natural and reprocessed uranium mixture;
2 – producing enriched product from RepU and diluting it with NatU or with other uranium mixture;
3 – producing enriched product from NatU and mixing it with RepU.
Multi-flow cascade (examples)

- $F$ – feed flow of NatU;
- $E$ – feed flow of RepU;
- $P$ – product flow;
- $W$ – waste flow.

- $F_1$ – feed flow of DepU;
- $F_2$ – feed flow of NatU;
- $F_3$ – feed flow of RepU;
- $P$ – product flow (LEU);
- $W$ – waste flow.
Double-cascade

\[ E_1 \rightarrow 232-236\text{U} \rightarrow 235,236\text{U} \rightarrow P_1 \rightarrow W_1 \rightarrow P_2 \rightarrow W_2 \rightarrow \text{LEU} \]

- \( E_1 \) – feed flow of initial RepU;
- \( P_1 \) – product flow of the 1\text{st} cascade;
- \( W_1 \) – waste flow of the 1\text{st} cascade;
- \( P_2 \) – product flow of the 2\text{nd} cascade;
- \( W_2 \) – waste flow of the 2\text{nd} cascade.

\( C_{235} \): up to 90%
Modified double cascade

\begin{itemize}
  \item $E_1$ – feed flow of initial RepU;
  \item $P_1$ – product flow of the 1st cascade;
  \item $W_1$ – waste flow of the 1st cascade;
  \item $P_2$ – product flow of the 2nd cascade;
  \item $W_2$ – waste flow of the 2nd cascade;
  \item $F_p$ – LEU-diluent;
  \item $P_0$ – final product (LEU).
\end{itemize}

- $C_{235} = 5$ - 15% for $\leq 20%$ in LEU
- $C_{235} \geq 5\%$ for $\geq 5\%$ in LEU
- $C_{235} = 3$ - 5% in LEU
EXAMPLE 1:
Simulation of multiple recycling of U with single cascades
Scenario of Multiple Recycling of RepU with single cascades

GENERAL ASSUMPTIONS:
1) Average enrichment of 4.6%
2) Irradiation in the core up to burnup 46.9 MWt-days/kg U
3) Reactor type – WWER-1000
4) Cooling duration is 5 years.
5) “One to one” scenario
   (from 1 kg of fresh fuel →
   → ~0.92-0.94 kg RepU)
6) Complex approach based coordinated simulation of isotope-selective mass transfer and neutron-physical processes was used

CASCADE CALCULATION:
1) The calculation model is a quasi-ideal cascade;
2) Constrained parameters:
   • Concentration of $^{235}$U in product - 4.6%;
   • Concentration of $^{235}$U in ‘waste’ flow - 0.1%;
   • Concentration of $^{232}$U in product is limited;
   • (not more than $5 \cdot 10^{-7}$%).
3) Reactivity compensation coefficient – 0.29;
4) 2 schemes were chosen:
Main goals

Simulation of multiple recycle of U and Pu in LWR (WWER-1000) to:

- assess possible savings of natural uranium and separating work on different recycles
- study evolution of the isotopic composition of RepU during multiple recycling
- assess possibility of using multiple-recycled fuel in LWR
Evolution of the isotopic composition (cascade with two feed flows)

<table>
<thead>
<tr>
<th># Cycle</th>
<th>Before cascade</th>
<th>After enrichment</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{232}$U, %</td>
<td>$^{234}$U, %</td>
</tr>
<tr>
<td>1</td>
<td>2.44e-7</td>
<td>2.26e-2</td>
</tr>
<tr>
<td>2</td>
<td>5.25e-7</td>
<td>3.17e-2</td>
</tr>
<tr>
<td>3</td>
<td>7.64e-7</td>
<td>3.75e-2</td>
</tr>
<tr>
<td>4</td>
<td>7.40e-7</td>
<td>3.58e-2</td>
</tr>
</tbody>
</table>
Evolution of the isotopic composition (cascade with three feed flows)

<table>
<thead>
<tr>
<th># Cycle</th>
<th>Before cascade</th>
<th>After enrichment</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{232}$U, %</td>
<td>$^{234}$U, %</td>
</tr>
<tr>
<td></td>
<td>$^{235}$U, %</td>
<td>$^{236}$U, %</td>
</tr>
<tr>
<td></td>
<td>$^{232}$U, %</td>
<td>$^{234}$U, %</td>
</tr>
<tr>
<td></td>
<td>$^{235}$U, %</td>
<td>$^{236}$U, %</td>
</tr>
<tr>
<td>1</td>
<td>$2.44 \times 10^{-7}$</td>
<td>$2.26 \times 10^{-2}$</td>
</tr>
<tr>
<td>2</td>
<td>$5.27 \times 10^{-7}$</td>
<td>$2.89 \times 10^{-2}$</td>
</tr>
<tr>
<td>3</td>
<td>$7.54 \times 10^{-7}$</td>
<td>$3.31 \times 10^{-2}$</td>
</tr>
<tr>
<td>4</td>
<td>$7.45 \times 10^{-7}$</td>
<td>$3.08 \times 10^{-2}$</td>
</tr>
</tbody>
</table>

Concentration of $^{235}$U in depU-diluent - 0.2%
Assessments of Dependence of Natural U and Separation Work Consumption on Natural U Cascade Parameters
(“+” – saving; “-” – overspending)

<table>
<thead>
<tr>
<th># Cycle</th>
<th>Two feed flows</th>
<th>Three feed flows</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Natural U, %</td>
<td>Separation Work, %</td>
</tr>
<tr>
<td>1</td>
<td>19.15</td>
<td>4.47</td>
</tr>
<tr>
<td>2</td>
<td>19.02</td>
<td>3.70</td>
</tr>
<tr>
<td>3</td>
<td>13.11</td>
<td>2.10</td>
</tr>
<tr>
<td>4</td>
<td>13.58</td>
<td>2.28</td>
</tr>
</tbody>
</table>
• The use of modified single separation cascades to enrich RepU allows to cope with the problem of even isotopes in multiple recycling and save natural uranium;

• Single cascade fail to fulfill complete-return condition during multiple recycling;

• The choice of separation cascade scheme may significantly influence on the important parameters of the fuel cycle such as natural uranium consumption and separative work.
EXAMPLE 2:
Simulation of multiple recycling of U and Pu in fuel of LWR with double cascades
Case I: Uranium recycling without Pu.
**Reactor type:** russian type of LWR - WWER-1200.
**Load of active core:** 48 fuel assemblies (FA) with enrichment of 4.95% and 24 FA with enrichment of 4.4%.
**Cooling duration of SNF:** 5 years.
**Fuel material:** low-enriched uranium made with full return of RepU from SNF.

Case II: Uranium recycling with Pu as REMIX-fuel.
**Reactor type:** russian type of LWR - WWER-1200.
**Load of active core:** 48 fuel assemblies (FA) with enrichment of 4.95% and 24 FA with enrichment of 4.4%.
**Cooling duration of SNF:** 5 years.
**Fuel material:** Plutonium and low-enriched uranium made with full return from SNF.
Plutonium is distributed equally along all FA
Main assumptions for cascade calculations

• External parameters:
  Concentration of $^{235}\text{U}$ in product – a) 4.95% ; b) 4.4% ;
  Concentration of $^{235}\text{U}$ in ‘waste’ flow - 0.1% ;
  Concentration of $^{232}\text{U}$ in product is limited;
  (not more than $5 \cdot 10^{-7}$ %);
  Relative concentration of $^{234}\text{U}$ and $^{235}\text{U}$ in LEU
  – lower than 0.02;
  Compensation of $^{236}\text{U}$ was taken into account.

• The calculation model is a quasi-ideal cascade.
Cascade schemes

1\textsuperscript{st} Cycle:

Other cycles:
Residual Concentration of $^{235}\text{U}$ in Dependence of Recycle Number, %
Saving of Natural Uranium in Dependence of Recycle Number, %
Residual Concentration of $^{232}$U in Dependence of Recycle Number, %
Residual Concentration of $^{236}$U in Dependence of Recycle Number, %
Separation Work in Dependence of Recycle Number
(“+” – savings, “-” – losses), %

![Graph showing separation work in dependence of recycle number. The x-axis represents recycle numbers 1 to 6, and the y-axis represents percentages from -5 to 25. The graph compares Rep U and REMIX-C.]
Residual Concentration of $^{236}$Pu in Dependence of Recycle Number, %
Residual Concentration of $^{238}\text{Pu}$ in Dependence of Recycle Number, %
Proliferation Resistance Analysis of LWR Fuel Produced from RepU
Purpose of the Study

- In this study we analyze the proliferation resistance of enriched reprocessed uranium in the sense of IAEA Safeguards implementation.
- We will consider scenario of the nuclear material diversion and outline the conditions when this material could be turned into a direct use material in terms of IAEA.
- We also assess the scale of enrichment facility needed to produce a significant quantity (SQ) of highly enriched uranium (HEU) throughout the duration of three months to two years, having the predetermined amount of diverted material (one SQ of LEU stolen from the fresh fuel).
- The main goal is to compare proliferation risks of NatU and RepU.
The problem of detecting the uranium diversion from fuel of a VVER-type reactor:

- Enrichment 4.6%
- Fuel operation cycle 1.5 years

691 fresh fuel rods

1167,9 kg of UO₂

1029,4 kg of U

75 kg of fissile $^{235}\text{U}$
Dependence of the number of "significant quantities (SQ)" of highly enriched uranium (>90%, HEU), derived from a significant amount of LEU, varying the ratio of the $^{235}\text{U}$ content in waste flow
The necessity to meet Timeliness Criterion

Dependence of the number of centrifuges* required for one significant quantity of HEU production on required time (for the different selected concentrations $^{235}$U in waste flow). Timeliness requirements: 1SQ of HEU – 3 months, LEU -1 year

The way of diverted material detection (1)

- Currently, the IAEA uses more than 100 different Non Destructive Assay (NDA) systems to check and monitor nuclear materials. Such verification occurs without changing the physical or chemical properties of controlled materials. NDA instruments range in size and complexity from small portable units used by safeguards inspectors during on-site verification activities to large in situ NDA systems designed for continuous unattended in-plant use.

- Most nuclear materials under IAEA safeguards, such as enriched uranium, emit $\gamma$ rays that makes possible its NDA. Determination of the $\gamma$ ray energies and their relative intensities serves to identify the isotopic composition of the materials. When combined with a measurement of absolute intensities, the $\gamma$ ray energies can provide quantitative information on the amount of material that is present.

- Enriched uranium fuel, made from natural uranium for example, has a strong 186 keV $\gamma$ ray associated with the $\alpha$ decay of $^{235}\text{U}$, and the $^{235}\text{U}$ enrichment can be verified by measuring this $\gamma$ ray. But to enhance the safeguards, it is necessary to check the amount of nuclear material in each fuel assembly based on the express analysis of fresh fuel assemblies. This is rather difficult for fuel made of natural uranium, since its indicative gamma lines are affected by the presence of radioactive background and shielding of internal fuel rods by external ones.
The way of diverted material detection (2)

• The distinguishing characteristic of fuel made of reprocessed uranium is that this option makes it possible to detect the replacement of even one fuel rod. Since the $^{208}$Tl line of 2.6 MeV has good penetrating power (the higher the gamma line energy, the less it is weakened by a shield), the substitution of 1 fuel rod leads to decrease in thallium concentration to $311/312$. The identification process for such fuel type includes the following steps:
  1. There is an expected value of $^{208}$Tl concentration in the fresh fuel assembly depending on the storage time that (as special gamma signature of this fuel assembly that can be calculated knowing the time the material has been stored).
  2. The energy of $^{208}$Tl in fresh fuel assembly is measured by the corresponding gamma line, then the concentration of this nuclide is calculated, and the error is assessed.
  3. Based on these data, the fact of diversion of nuclear material is concluded.

• What does the identification mean? Let us set the probability belief equal 0.95. Within this confidence factor we want to determine the fact of diversion, knowing that the difference between the calculated and measured $^{208}$Tl concentration value will be greater than its doubled error.
• The identification function:

$$\ln(T_{\text{expos}}) = \frac{2 \cdot \sigma(N_{\text{Tl-208}})}{N_{\text{Tl-208}}(T_{\text{expos}}) - N_{\text{Tl-208}}} < 1$$

where $T_{\text{expos}}$ – storage time after reprocessing, $N_{\text{Tl-208}}$ – nuclear concentration of thallium obtained from measurements, – nuclear concentration of thallium obtained from calculations depending on the storage time.
Relative spectrum set-up time for different volumes of diverted nuclear material

Time necessary for spectral analysis, where 1 – time necessary for express test (1000 sec)

Hold-up time after uranium reprocessing, days
Reuse of recovered from SNF uranium and plutonium brings some advantages for nuclear energy as it allows to:

1. significantly reduce amount of SNF;
2. save from 15 to 30% of natural uranium with moderate losses of separation work (up to 5%);
3. stimulate development of corresponding technologies which is important for future sustainable development of nuclear energy with FNR.

Undoubtedly that the choice of separation technique (separation scheme) is one of the important things which define the effectiveness of closing fuel cycle of LWR.
• Despite the special properties of the reprocessed uranium, when enriching its significant quantity (SQ) in form of low-enriched uranium (LEU), it is possible to obtain one significant quantity of highly enriched uranium (HEU), which indicates the importance of detecting the nuclear material diversion.

• Compared LEU fuel made from natural uranium, due to unique physical properties of reprocessed uranium, to provide physical inventory verification under IAEA safeguards implementation is possible even with a quick NDA test even in case of a single fuel rod diversion.
Thank you for your attention!