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TECHNOLOGICAL APPROACHES TO NON-STANDARD SPENT NUCLEAR FUELS REPROCESSING

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MAIN POINTS

1. Technological approaches to spent ATF reprocessing

2. Technological approaches to spent HTGR fuel reprocessing

3. Molten-salt reactor and spent salt reprocessing

1.1 ACCIDENT TOLERANT NUCLEAR FUEL IN RUSSIAN FEDERATION

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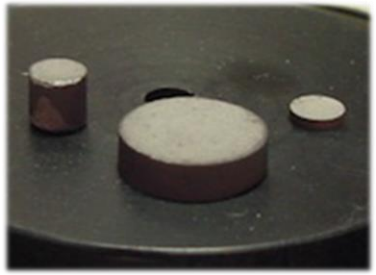
- ATF options
  - Modification of claddings
    - Chrome coated zirconium
    - New alloys/materials
  - Modification of fuel composition
    - U_3Si_2
    - UMo
    - UO_2 with Cr

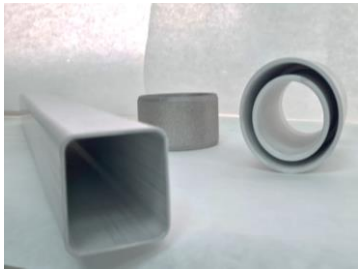


tablet U_3Si_2

Zr-alloy+Cr cladding

42CrNiMo cladding

SiC cladding



Dispersed U_3Si_2 fuel

1.2 ISSUES DURING ATF REPROCESSING IN PUREX-PROCESS

1. Mo and Si sediments formation during hydrometallurgical processing to obtain the fed flow to the solvent extraction.

2. New construction materials behavior during SNF reprocessing.

3. Due to the fuel burnup increasing, a greater amount of fission products is formed – volatile (T, I), sediment-forming (Mo, Zr), intermetallics of noble metals.

4. High requirements to U, Pu and minor actinides recovery.

5. New types of RW for handling.

1.3 VOLOXIDATION (SNF VOLUME OXIDATION)

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$$U_3Si_2 + 6O_2 \rightarrow U_3O_8 + 2SiO_2$$

BEFORE

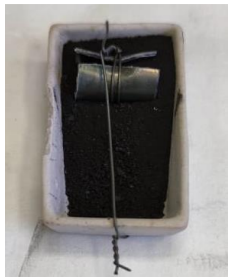
AFTER

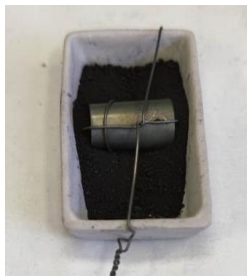
600°C, O_2


a) Cr + Zr


b) 42XHM


c) SiC/Zr/SiC


a) Cr + Zr


b) 42XHM


c) SiC/Zr/SiC

700°C, O_2


d) Cr + Zr


e) 42XHM


f) SiC/Zr/SiC

- At 600 ℃ and above the complete oxidation of U_3Si_2 to U_3O_8 after 4 hours.
- 4-5 times increasing of the sample volume.
- According to the X-ray diffraction analysis the powders contain fractions of U_3O_8 and SiO_2.

1.4 CLADDING WASHING AFTER VOLOXIDATION

The same conditions (as for dissolution):
HNO_3 8-9 mole/l, t=95°C

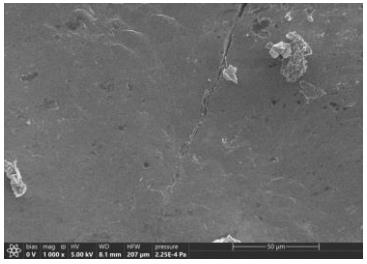
42CrNiMo alloy
In washing solution: 18 times more Ni,
4 times more Cr.

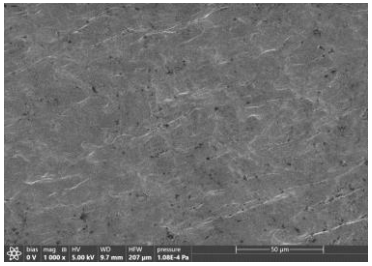
Zr+Cr
Washing time 1 hour, the presence of cladding components was not detected

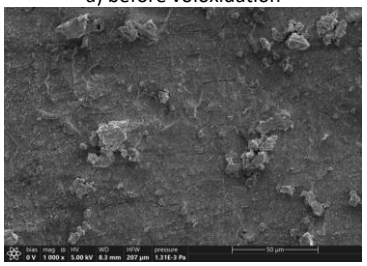
SiC
Washing time less than 15 minutes, the presence of cladding components was not detected

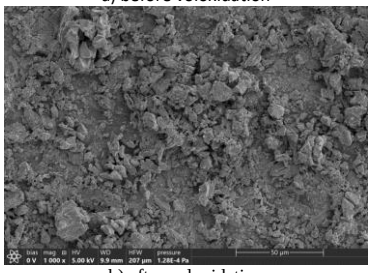
~ 0.5% of U remains on the cladding, irreversible losses of U less than 0.01% after washing

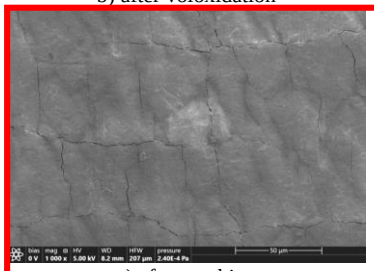
Zr + Cr | **42CrNiMo alloy**

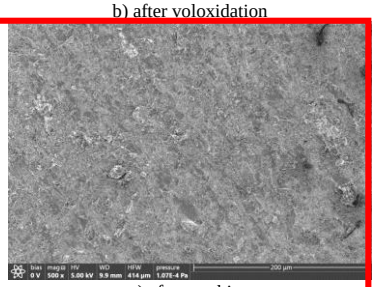

a) before voloxidation


a) before voloxidation

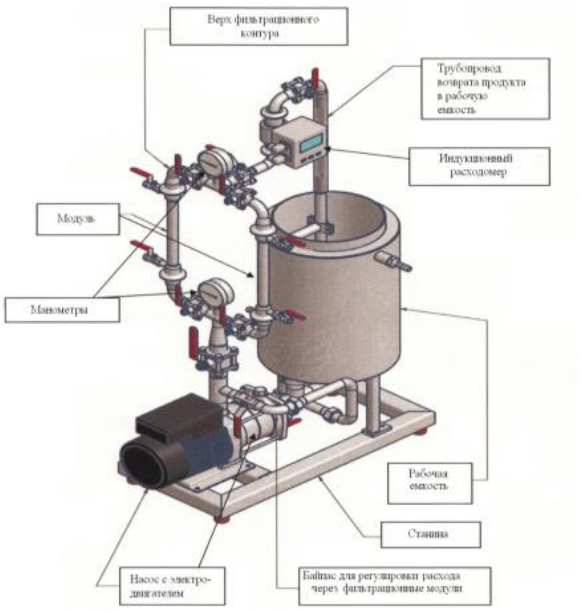

b) after voloxidation


b) after voloxidation


c) after washing

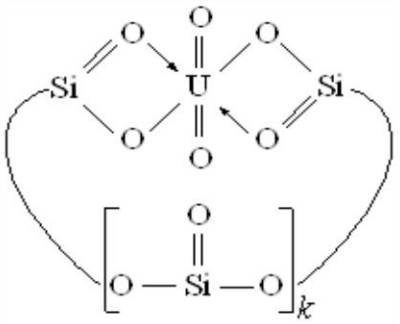

c) after washing

1.5 TWO-STEP CLARIFICATION OF THE SNF SOLUTION



Laboratory scale tangential filtration unite

- First step – centrifugation to reduce the amount of solid particles to 700-1000 mg/l.
- Second step – tangential ultrafiltration to get the total clarification factor **10^4** and to provide less than 5 mg/l of solid remaining in the SNF solution.



Si could form complex extractable oligomeric compounds with U, a special flocculant was selected and tested to prevent Si coextraction in the 1 cycle of the Purex-process.

1.6 PYROCHEMICAL PROCESSING OF SPENT ATF

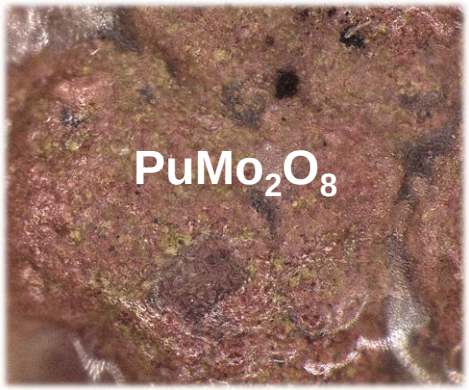
R&D results approved the complete direct dissolution of U_3Si_2 in MoO_3 with decontamination of crystallized uranium and plutonium compounds from fission products .

$$U_3Si_2 + 6O_2 \rightarrow U_3O_8 + 2SiO_2$$

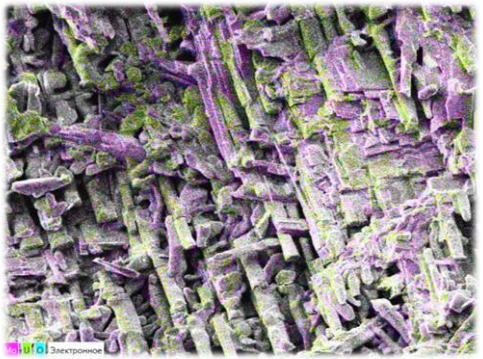
$$U_3O_8 + 3MoO_3 + ½O_2 \rightarrow 3UO_2MoO_4$$

Si could be removed from the melt with purification factor 10^2 (currently in study)

After dissolution of U_3Si_2 in MoO_3



Product of Pu crystallization in MoO_3



Crystals of UO_2MoO_4 In the matrix of MoO_3

The search of stable constructive materials is the important task.

1.7 VITRIFICATION OF RW FROM HYDROMETALLURGICAL PROCESSING

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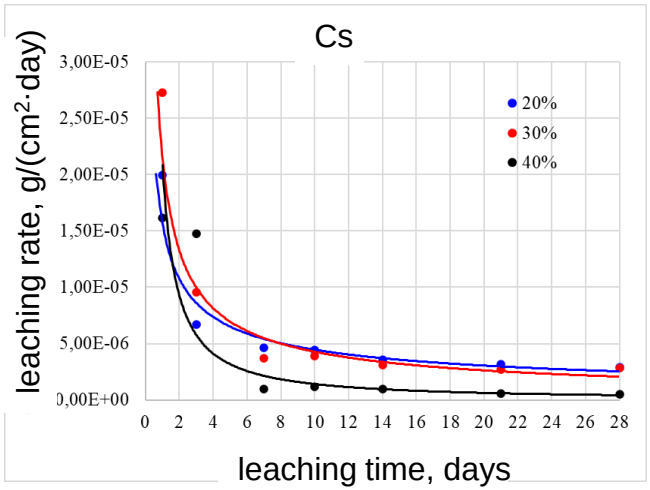

40% of RW inclusion in **borosilicate glass**

It was found and tested the method of 10 mass % of Mo inclusion in the form of molibdates.

- Mechanical strength meets to accepted values for solidified radioactive waste.
- Distribution of components is uniform.
- Leaching rate of Cs, Sr & Na from glass with RW meets the requirements for matrices.

2.1 HTGR development in Russian Federation

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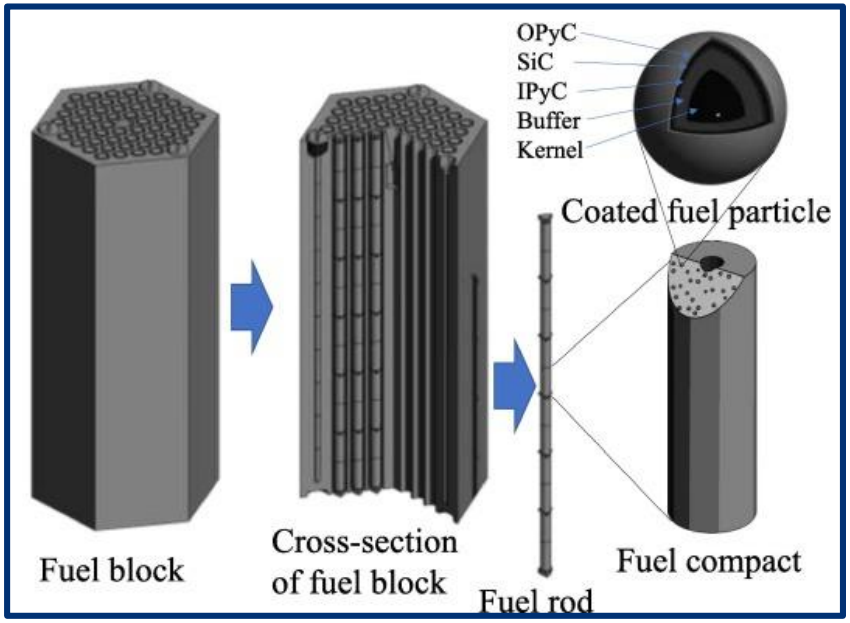
- Hydrogen Energetics Project
- Prismatic type assembly
- UO_2 kernel, 18 % U-235 enrichment
- Burnup 120 MWd/tHM



Composition of a prismatic type HTGR assembly

Fukaya Y., Goto M., Ohashi H. Feasibility study on reprocessing of HTGR spent fuel by existing PUREX plant and technology //Annals of Nuclear Energy. – 2023. – T. 181. – C. 109534.

Carbon ~ *94 %* of assembly`s total mass

2.2 HEAD-END PROCESSES PRINCIPAL FLOW-SHEET

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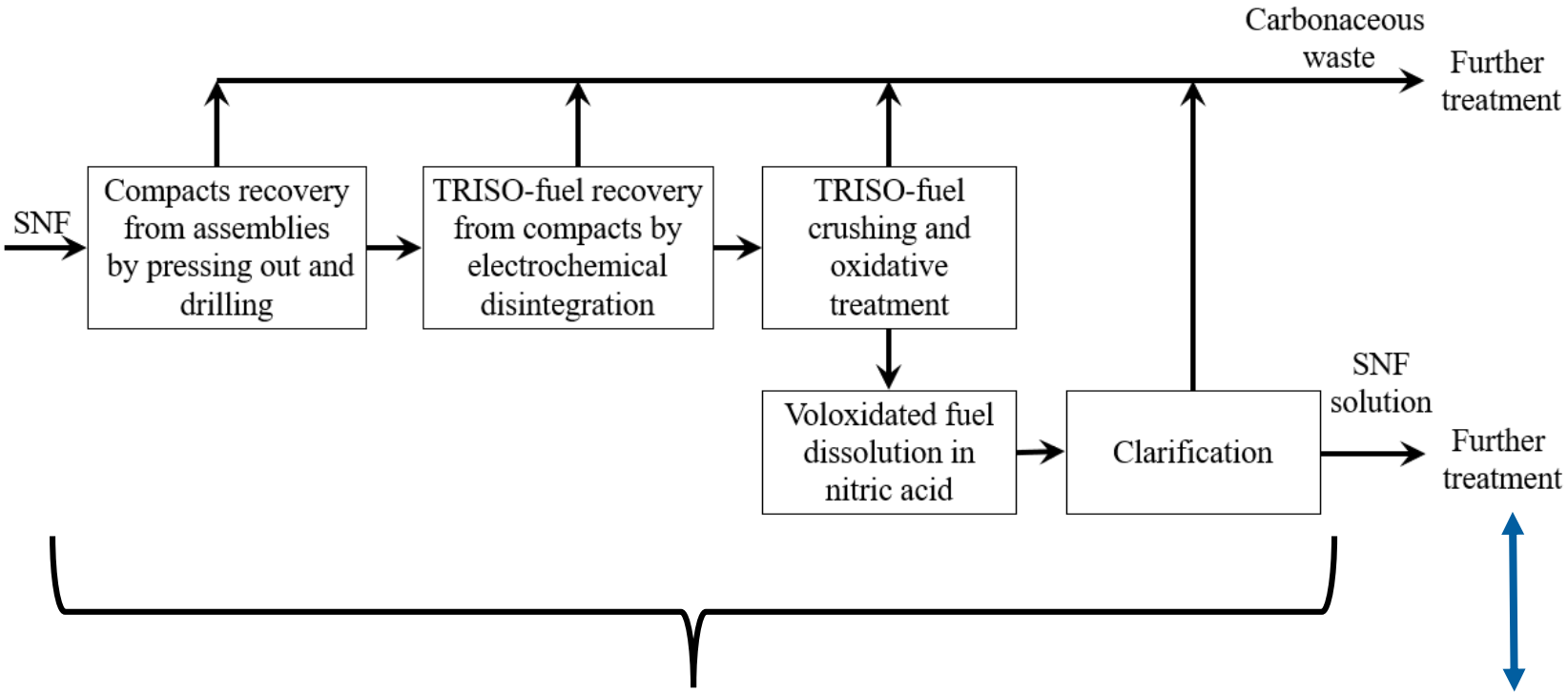

2.3 R&D steps

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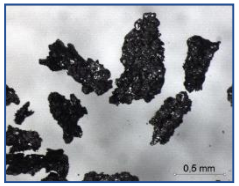
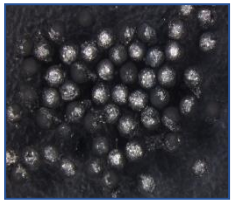
COMPACTS RECOVERY
drilling testing

TRISO-FUEL RECOVERY
electrochemical disintegration

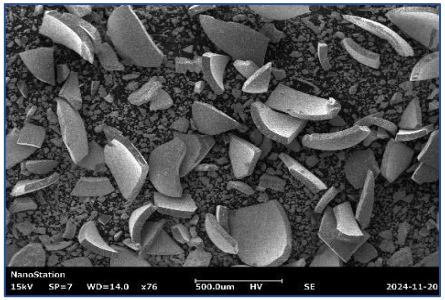




< 1-2 hours
for 1 compact

TRISO integrity remains

KERNEL RECOVERY
pyrocarbon incineration and voloxidation

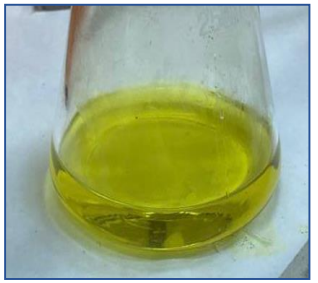


Fuel separates from SiC hulls

Remaining PyC < 0,7-0,8 % of initial PyC content

FUEL RECOVERY
dissolution and clarification

SiC hulls



clarified solution

LOW Chemical Oxygen Demand* = 150 mgO/l

Comparatively, same COD have solutions of: Oxalic acid 3 mmole/l
Or vinegar acid 5 mmole/l

2.4 CARBONACEOUS RADIOACTIVE WASTE IMMOBILIZATION

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GRAPHITE

Both Assembly`s and Compact`s graphite waste streams are estimated to be 3rd class waste

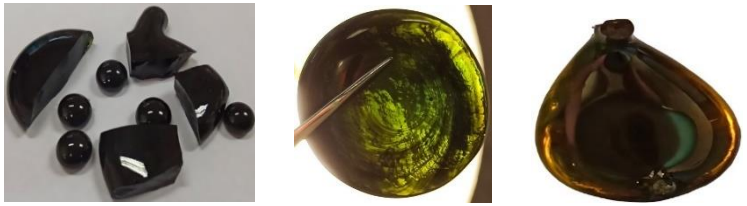
25 % graphite inclusion in Portland cement

PYROCARBON

Mostly follows SiC waste stream

SILICON CARBIDE

Estimated to be 2nd class waste



! **5 % SiC inclusion in borosilicate glass**

Optional: oxidation to SiO_2 (currently in study)

3.1 MOLTEN SALT REACTOR (MSR): AIM AND TASKS

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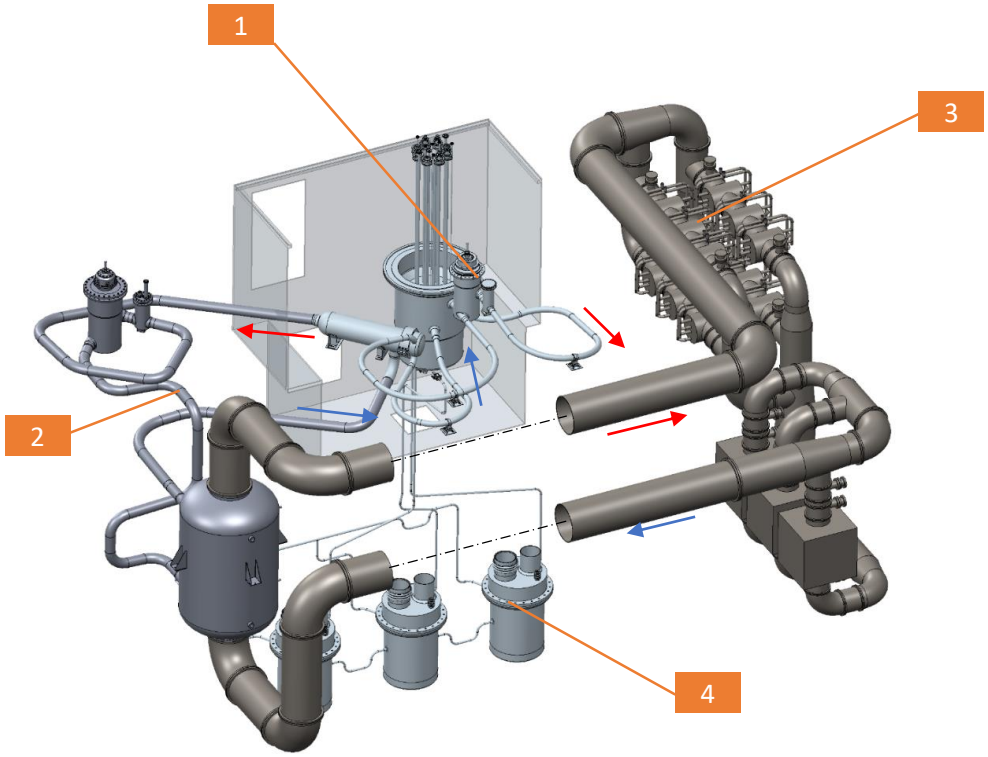
Reducing the volume of minor actinides (MA) from spent nuclear fuel of thermal reactors, due to transmutation of MA in a specialized molten salt reactor (incinerator).

Advantages of MSR

1. Absence of fabrication and re-fabrication of fuel pellets, fuel elements and fuel assemblies, and also absence of transportation of fuel with MA across the country.
2. Undemanding to the isotopic composition of the nuclear fuel.
3. Transmutation of MA does not require expensive operation of the Am/Cm separation

First step of R&D - Research MSR creation.

3.2 PRINCIPAL FLOWSHEET OF THE RESEARCH MSR



1 – fuel circuit; 2 – intermediate circuit; 3 – gas circuit; 4 – passive fuel salt drain system

Thermal power, MW	10
Circulation type	Pump
Fuel salt	73LiF-27BeF_2 +(PuF_3+A_nF_m)
Intermediate circuit salt	66LiF-34BeF_2
Gas circuit coolant	Азот
Flow rate in the fuel circuit, kg/s	141,6
Fuel salt temperature at reactor inlet/outlet, °C	664/700
Service life, years	10

3.3 THE FUEL OF THE RESEARCH MSR

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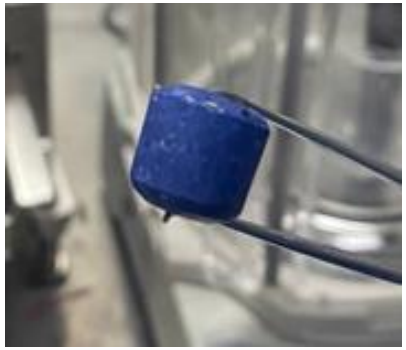
73 mole % of Li - 27 mole % of (BeF_2 + $Pu(Am)F_3$).



PuF_3

T_{melt} = 1430°C

[O] = 1500 - 6000 ppm

$0{,}2PuF_3$ - 0,8LiF

T_{melt} = 748°C

[O] = 35-50 ppm

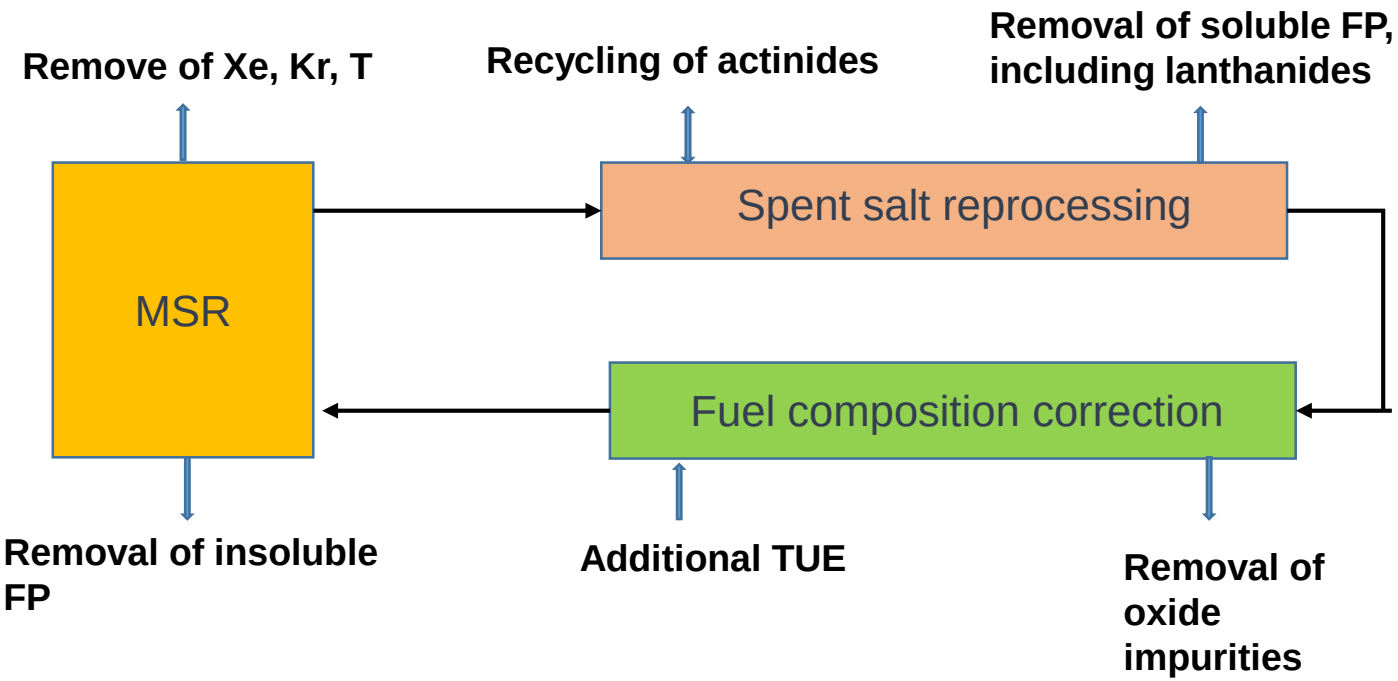
0,73 LiF-0,27 BeF_2+0,1PuF_3

T_{melt} = 650°C

[O] ≤ 100 ppm

3.4 MSR FUEL CECLE

Transmutation of MA into MSR is possible only in combination with a fuel cycle that includes regeneration of fuel salt with fissile material recycling, loading of TUE to compensate burnup and correction of salt composition and oxidation reduction potential



Spent fuel salts reprocessing is being developed based on the reducing extraction technology of actinides into liquid metal.

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THANK YOU FOR

YOUR ATTENTION!

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