CURRENT CZECH R&D IN THORIUM MSR TECHNOLOGY AND FUTURE DIRECTIONS

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Outline:

- Arguments for R&D of MSR technology
- Existing R&D projects supported by the Ministry of Industry and Trade
- Development in reactor physics, structural material and fuel cycle technology
- Current status and future directions

R&D of GEN IV reactor systems and P&T

- Decision to develop selected technologies of MSR system and to resume on the ORNL effort from 1960s was made in 1999
- Ministry of Industry and Trade (MoIT) has been supporting the R&D program since 2000
- National P&T program has been also based on MSR technology and fluoride pyrochemical partitioning

- It was relatively exceptional activities in EU *(Main European approach to GEN IV and P&T is focused on FR and ADS)*

Main sense of the Czech focusing on MSR R&D and main aims of our effort

Sense and background

- Basic principles of MSR were successfully verified in 1950s and 1960s in ORNL → MSR is viable technology
- ^{232}Th – ^{233}U fuel cycle within MSR is environmentally much more friendly approach than ^{238}U – ^{239}Pu fuel cycle (closed fuel cycle, no production of higher actinides)
- MSR technology could be acceptable for non-superpower countries (very low concentration of fissile ^{233}U in the fuel salt, use of ^{233}U with excellent physical protection)

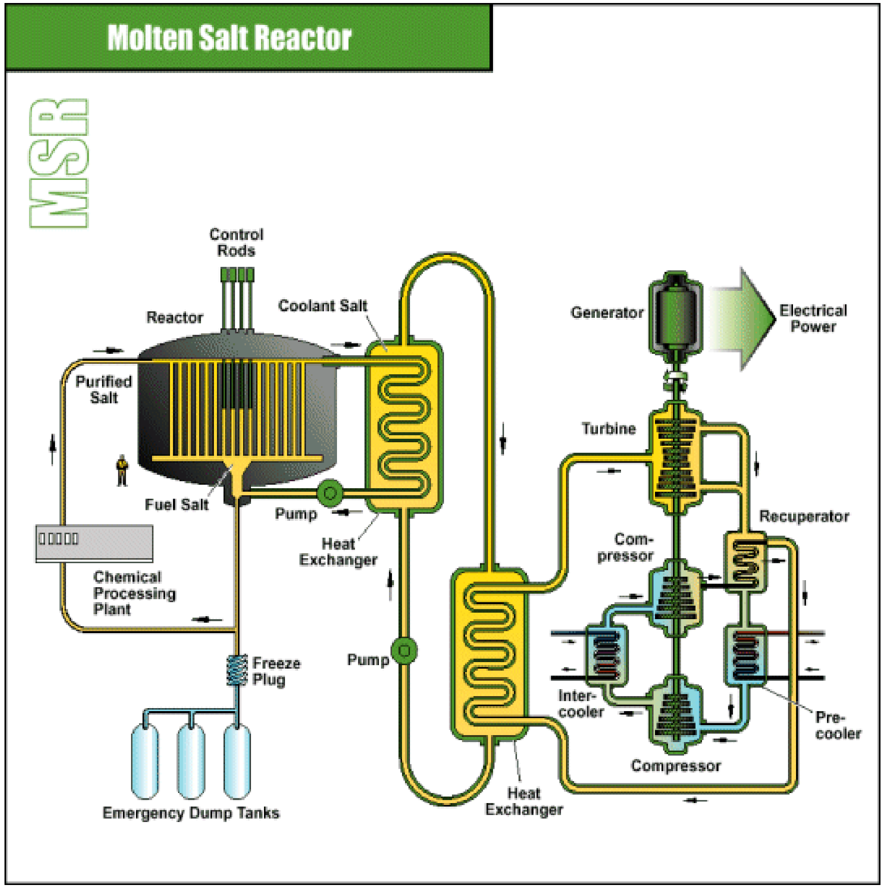
Main aims

- To appropriately contribute to the knowledge of MSR reactor physics, core design and safety, structural material development and Th – U fuel cycle
- To focus on R&D of technologies applicable within the MSR on-line reprocessing of liquid fuel
- To verify experimentally selected important areas of MSR technology and to solve existing bottlenecks

Czech Republic is too small country to be able to develop some GEN IV reactor system completely, but we can be an important partner for final collaborative development of MSR.

MSR – non classical reactor type

- **Specific features of MSR comes out from the use of liquid (molten-salt) fuel circulating in the primary circuit.**
- **MSR can be operated either as thorium breeder (in thermal or fast/resonance spectrum) within the ^{232}Th – ^{233}U fuel cycle or as actinide transmuter (in resonance/epithermal spectrum) incinerating transuranium fuel.**
- **Typical fuel: fluorides of actinides dissolved in fluoride carrier salt.**



R&D project devoted to MSR system technology

MoIT project: "Nuclear system SPHINX with molten fluoride salts based liquid nuclear fuel" (2004 - 2008)

- R&D project devoted to MSR technology covering theoretical and experimental activities in MSR physics, fuel cycle chemistry, molten salt thermohydraulics, structural material development and testing of apparatuses for molten fluoride salt media
- Solved by consortium of institutions and companies including NRI Řež, ŠKODA JS (Nuclear Machinery), Energovýzkum Ltd., Faculty of Nuclear Sciences and Physical Engineering of the CTU in Prague

Subsequent MoIT project: "Fluoride reprocessing of GEN IV reactor fuels" (2006 – 2012)

- R&D project devoted mainly to the MSR fuel cycle technologies
- Solved by NRI Řež

Participation in selected EURATOM projects devoted to MSR technology (MOST, ALISIA, EVOL)

Participation in SSC MSR of the Generation Four International Forum in the frame of the EURATOM membership

Czech R&D projects devoted to MSR system technology

Nuclear system SPHINX with molten fluoride salts based liquid nuclear fuel

- R&D project devoted to MSR technology covering theoretical and experimental activities in MSR physics, fuel cycle chemistry, molten salt thermohydraulics, structural material development and testing of apparatuses for molten fluoride salt media

Fluoride reprocessing of spent fuel of GEN-IV reactors

- R&D project devoted to pyrochemical fuel cycle technologies focused mainly to fluoride separation processes suitable for MSR technology including thorium - uranium fuel cycle and separation of transuranics

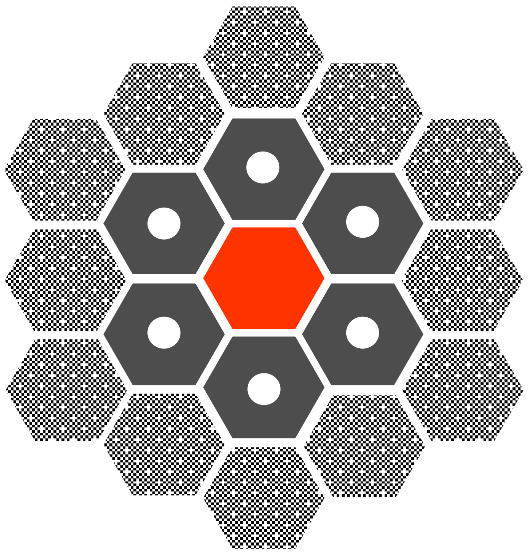
SPHINX project work-packages:

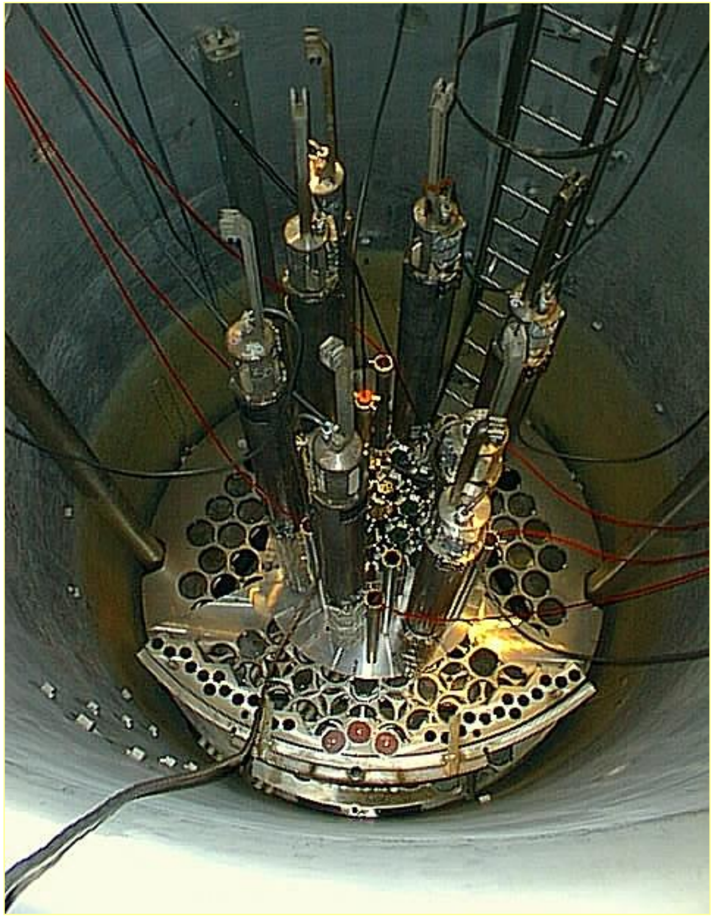
- WP1 – MSR core and primary circuit
- WP2 – MSR fuel cycle technology
- WP3 – Experimental MSR core and its control system
- WP4 – Secondary circuit and its components
- WP5 – Structural materials for MSR technology
- WP6 – System study of MSR – SPHINX
- WP7 – Experimental program SR-0

Selected results of SPHINX project

WP1 – MSR core and primary circuit
WP3 – Experimental MSR core and its control system
WP7 – Experimental program SR-0

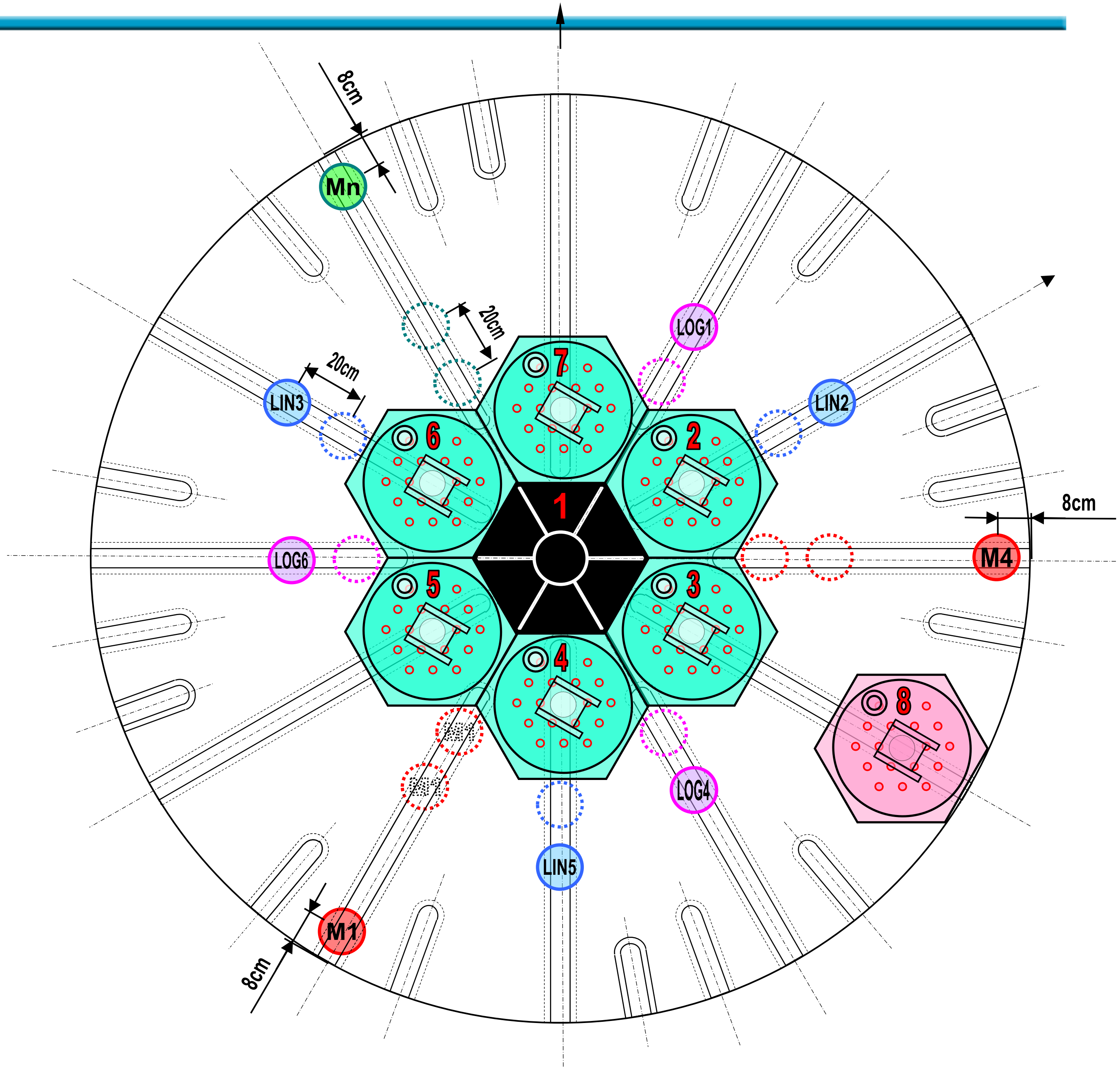
- Development of modified computer codes for calculations of neutronic characteristic of MSR systems
  - **OGAR code - calculation of the evolution composition during the burnout**
- Experimental research of MSR neutronics based on instrumented BLANKA probes irradiation (EROS)





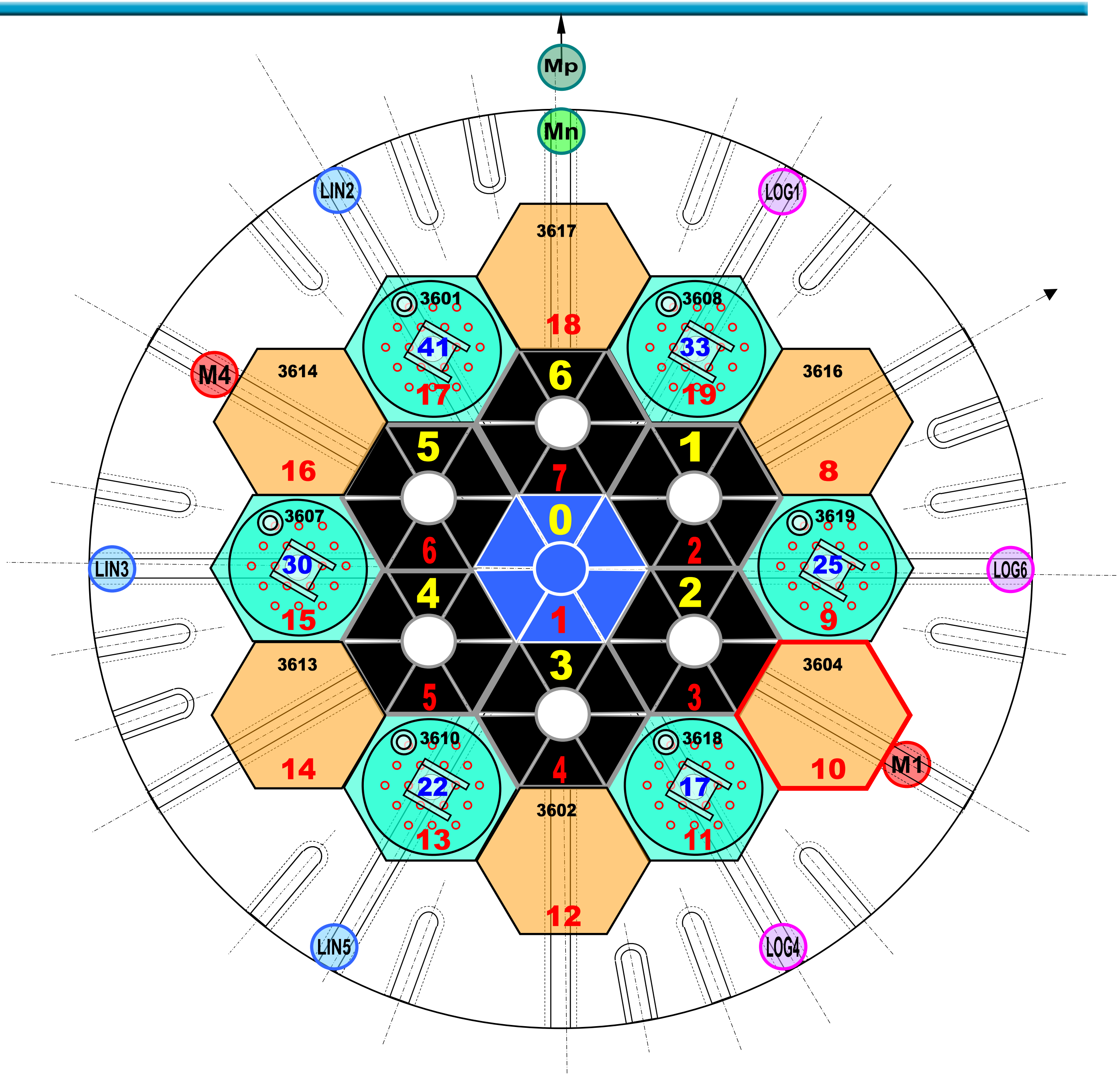
Geometries of SPHINX in LR-0 reactor

- First configs with 1 exp. channel
- Filled by
  - Graphite
  - LiF-NaF 60 / 40 % mass
  - Graphite with salt in center cyl.

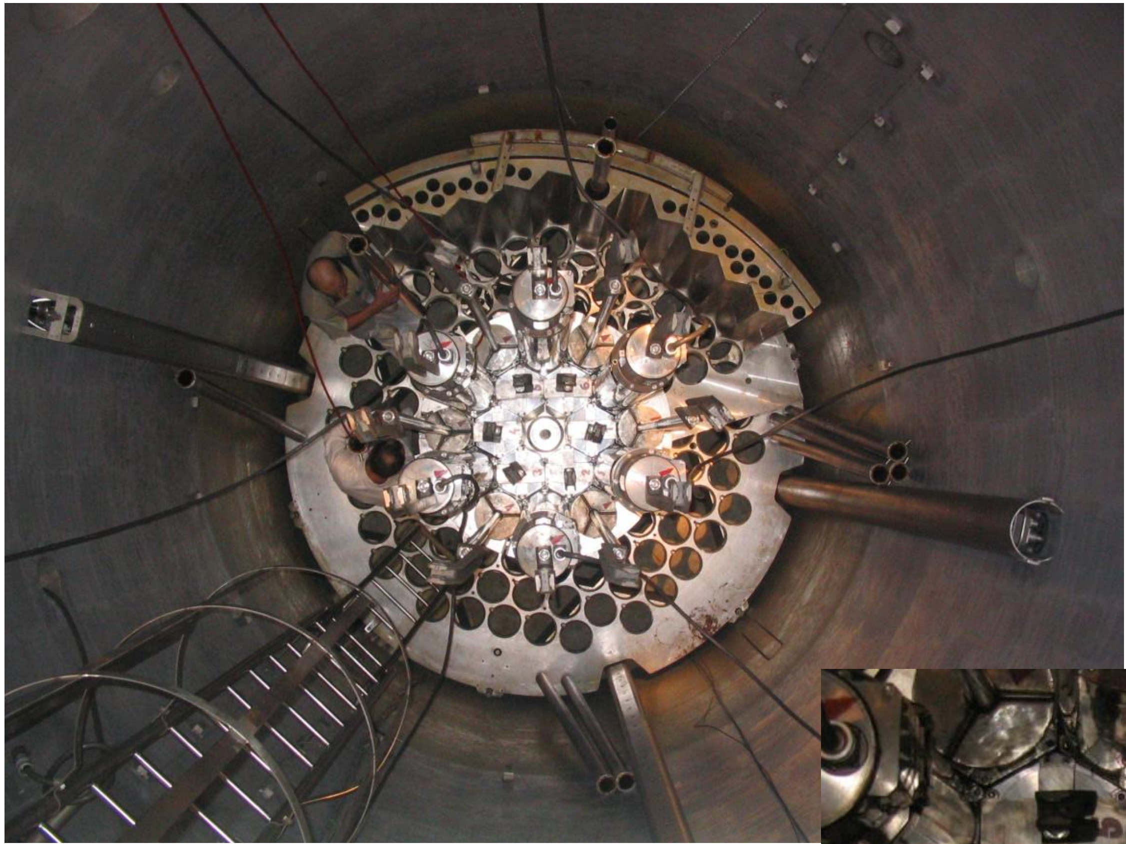


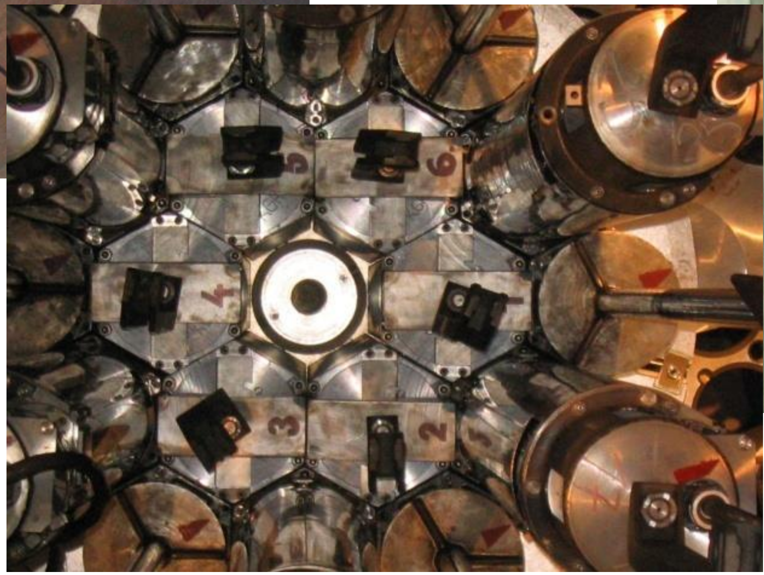
Geometries of SPHINX in LR-0 reactor

- Configs with 7 exp. channels
- 2 variants with or without salt cylinders in peripheral graphite channels



SPHINX inserted zones in LR-0 reactor





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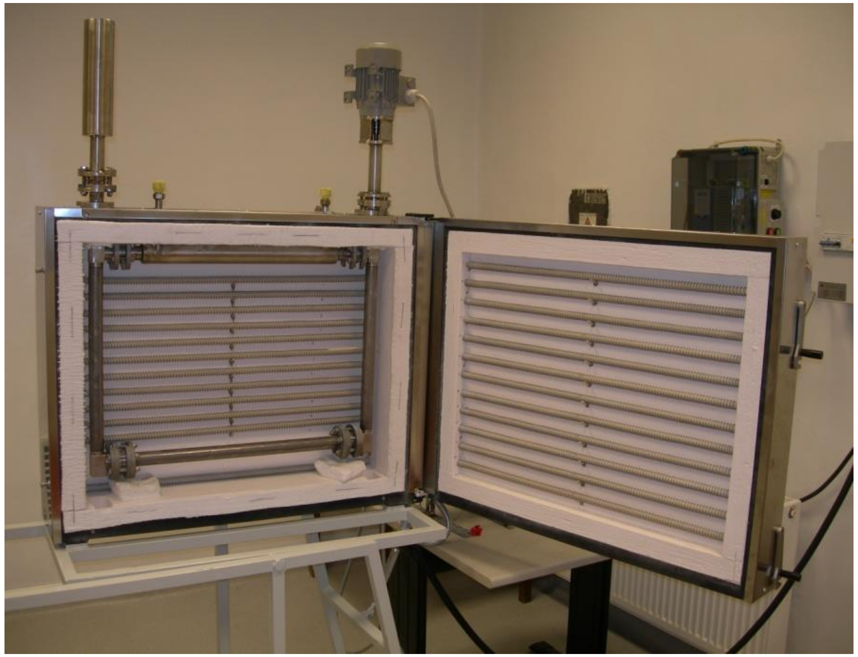
Selected results of SPHINX project (cont.)

WP2 – MSR fuel cycle technology

- Development and verification of liquid fuel processing – molten salts containing UF_4 and ThF_4
- Development of Fluoride volatility process for transuranium fuel processing and Electrochemical separation technology
- Basic studies of molten fluoride salt thermodynamics



Selected results of SPHINX project (cont.)

WP5 – Structural materials for MSR technology

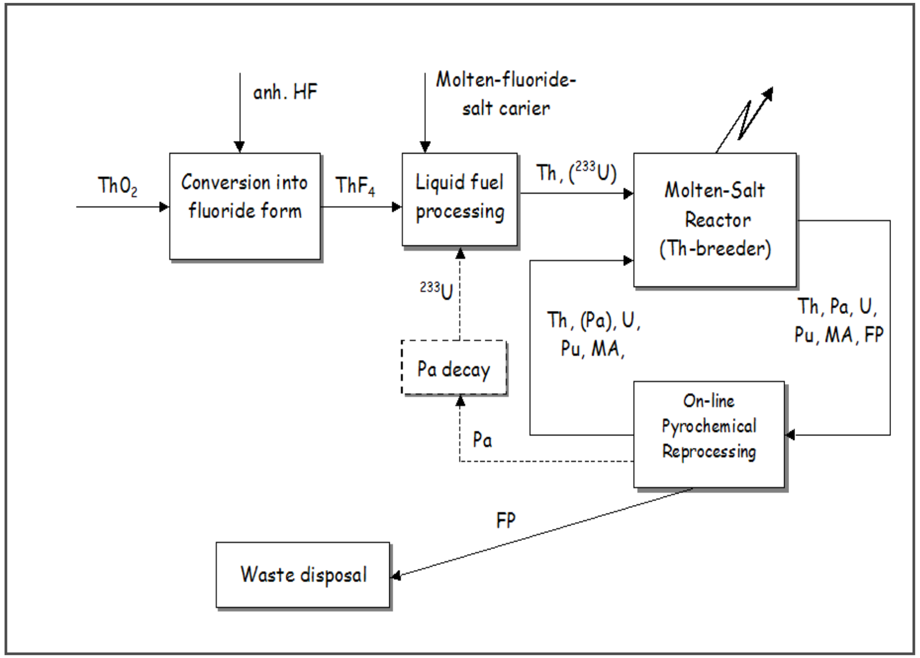
- Development of high content nickel alloy MONICR for molten-salt technology
  - **Corrosion, irradiation and metallographic tests**
  - **Design and manufacture of selected equipment and apparatuses**

Collaboration with ŠKODA JS and COMTES FHT

Selected results of SPHINX project (cont. 3)

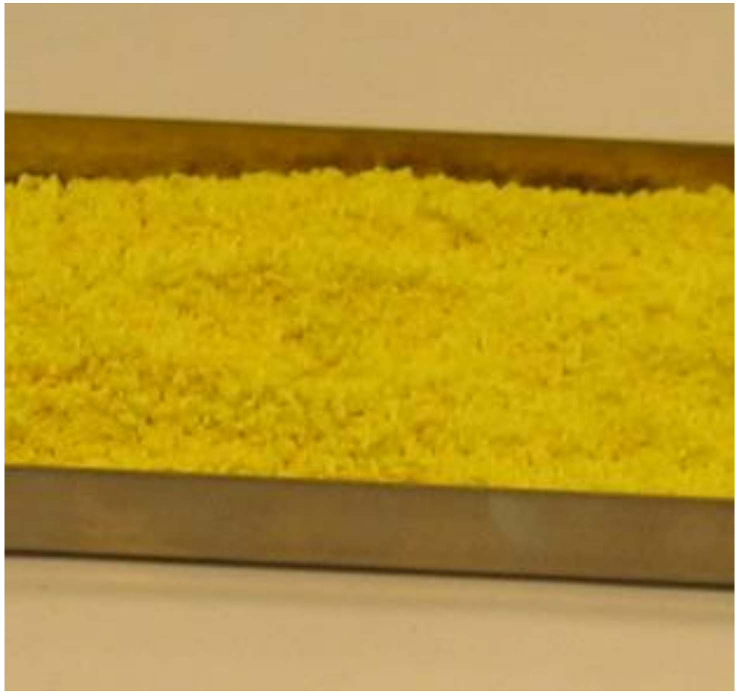
WP4 – Secondary circuit and its components
WP6 – System study of MSR - SPHINX

- Calculation and design studies of MS heat exchangers and use of gas turbine systems for generating electricity
- System studies of MSR – Th breeder (single-fluid and double-fluid system)
- Non-proliferation aspects of MSR fuel cycle technology



MSR fuel processing: Preparation of UF_4

- $9(NH_4)_2U_2O_7(s) \rightarrow 2N_2(g)+14NH_3(g)+6U_3O_8(s)+15H_2O(g)$
- $U_3O_8(s)+2H_2(g) \rightarrow 3UO_2(s)+2H_2O(g)$
- $UO_2(s)+4HF(g) \rightarrow UF_4(s)+2H_2O(g)$



$(NH_4)_2U_2O_7$

UF_4

MSR fuel processing: Preparation of ThF_4

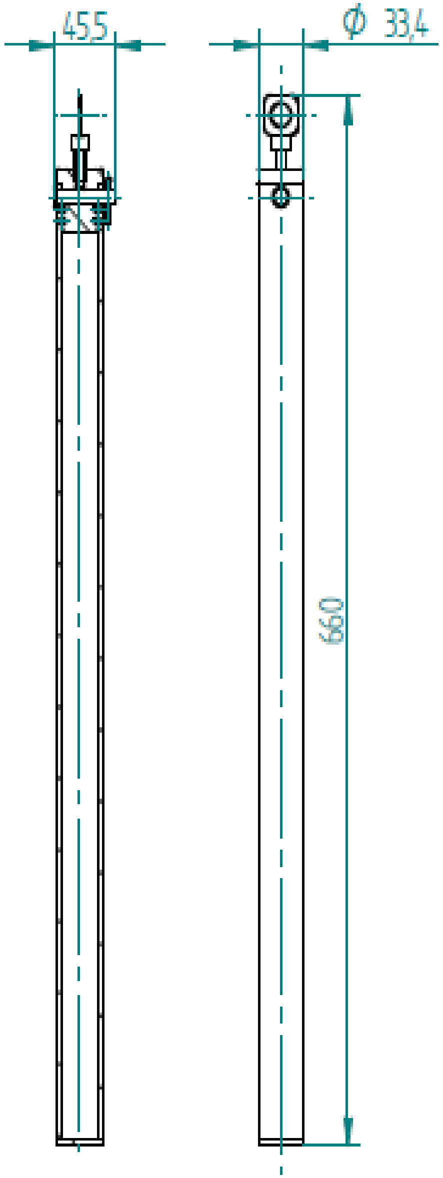
- $ThO_2(s) + 4HF(g) \rightarrow ThF_4(s) + 2H_2O(g)$

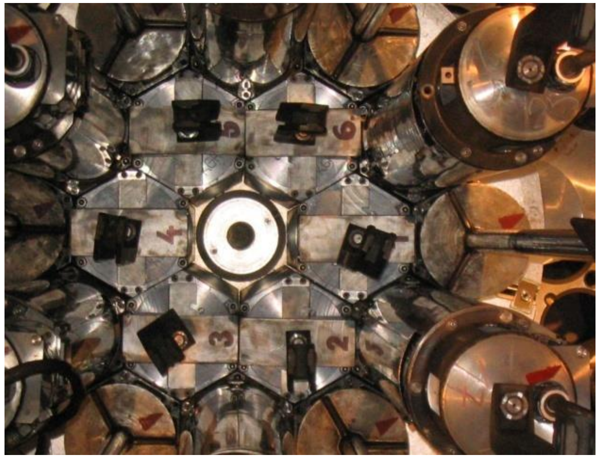
ThO_2

ThF_4

AMPULA for LR-0 inserted zone

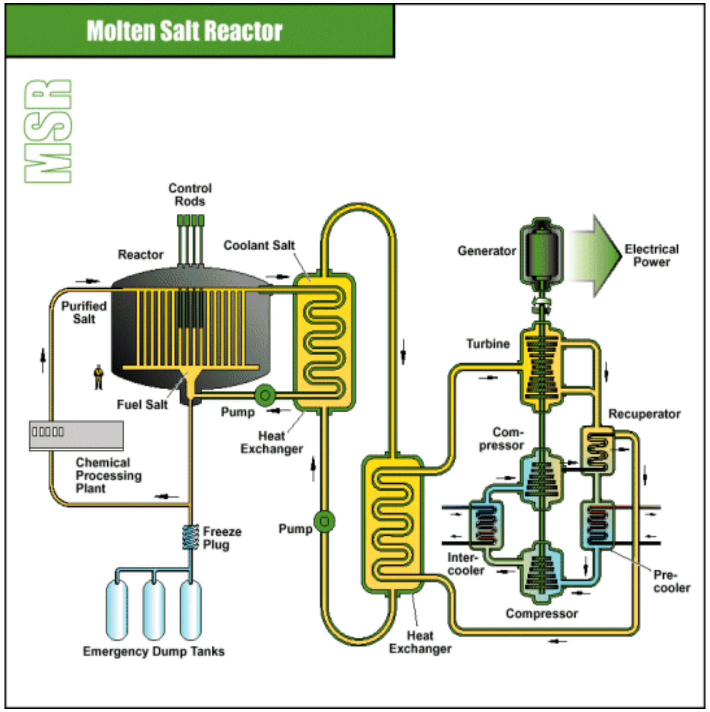
- **Inserted probe for irradiation experiments in experimental LR-0 reactor and in school VR-1 reactor**



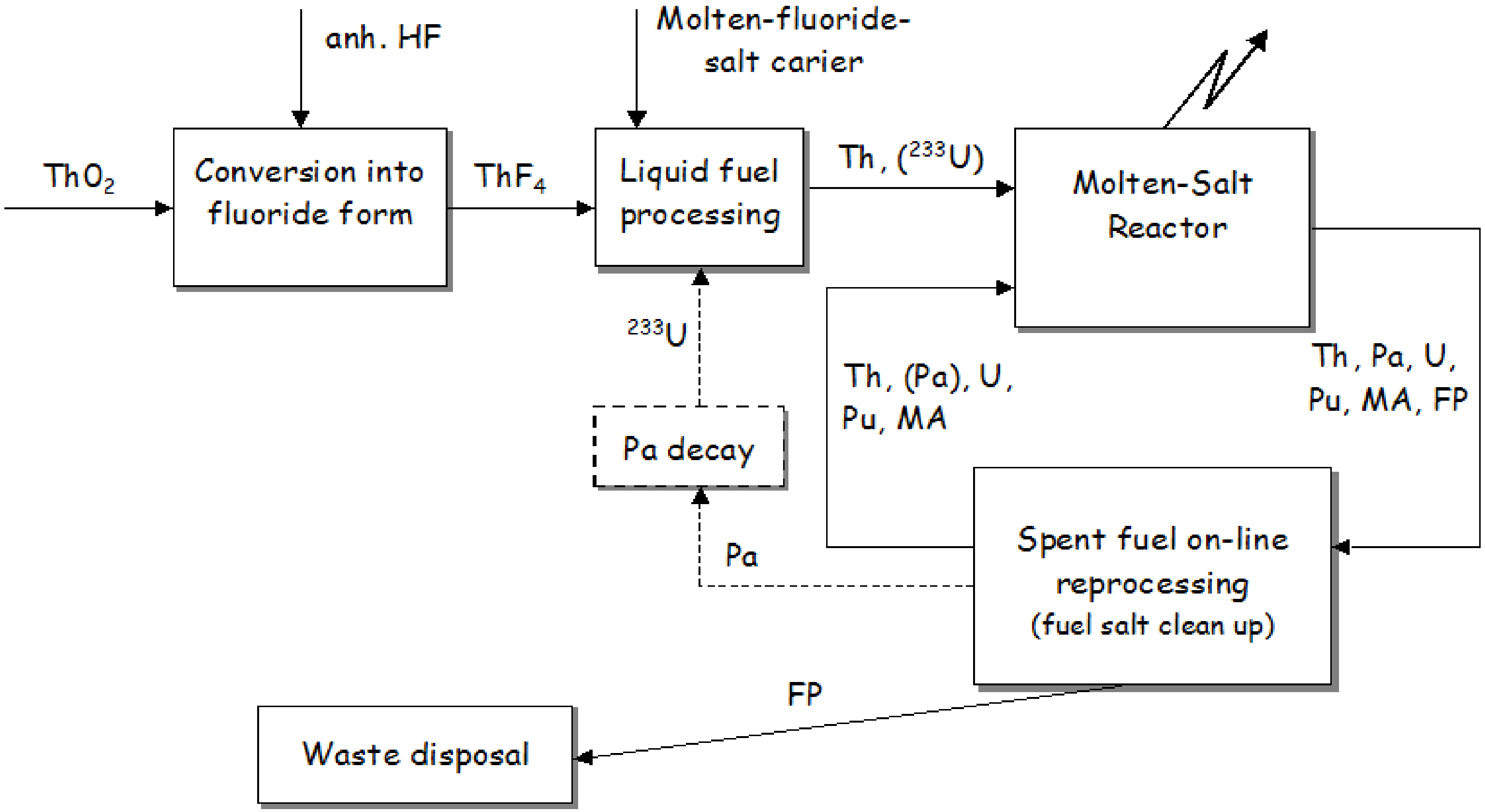


MSR – thorium breeder

- MSR is the only reactor system from the GEN IV reactor family for which the thorium fuel is considered.
- ^{233}U is the only fissile material in the thorium – uranium fuel cycle
- MSR – Th breeder with higher breeding cannot be operated without the on-line reprocessing
- Typical fuel: ThF_4 and UF_4 dissolved in ^{7}LiF – BeF_2 carrier molten salt.



Fuel cycle technology in MSR system



Mission of MSR fuel on-line reprocessing

To keep the reactor in steady-state conditions by continuous cleaning-up of the primary (fuel) circuit salt.

- **To clear away neutron poisons like Xe, Kr, lanthanides**
- **To extract freshly constituted fissile material or its precursors**
  - Desirable reaction in the reactor core (MSR – Th-breeder):

    $^{232}Th\ (n,\gamma) \rightarrow {}^{233}Th(\beta^-) \rightarrow {}^{233}Pa(\beta^-) \rightarrow {}^{233}U$
  - Undesirable reaction: $^{233}Pa(n,\gamma) \rightarrow {}^{234}Pa(\beta^-) \rightarrow {}^{234}U$
- **To secure refilling of fresh fuel into fuel (primary) circuit**

  (reprocessing technology is connected with fresh fuel feeding)

Technologies proposed for on-line reprocessing

Reprocessing technologies proposed for MSR fuel cycle are generally pyrochemical, majority of them are fluoride technologies. This is caused by the fact that MSR fuel is constituted by a mixture of molten fluorides and the technology has to be resistant to a very high radioactivity of fuel entering the process.

Main separation techniques proposed for on-line reprocessing of MSR fuel:

- **Fluoride volatilization processes**
- **Molten-salt / Liquid metal extraction processes**
- **Electrochemical processes**
- **Gas extraction process (He-bubbling)**

R&D on electrochemical reprocessing of MSR fuel in Řež
- separation of actinides and fission products

R&D on Electrochemical separation of An/Ln from molten fluoride media

Development of separation technology suitable for on-line reprocessing of MSR fuel

Carrier salt of MSR primary (fuel) circuit:

$^{7}LiF\text{-}BeF_2$ (called FLIBE) or $^{7}LiF\text{-}BeF_2\text{-}NaF$

However, FLIBE is insufficiently electrochemically stable.

Carrier salts proposed for electrochemical separation processes:

$^{7}LiF\text{-}BeF_2$ or $^{7}LiF\text{-}BeF_2\text{-}NaF$ *(limited use)*

LiF-NaF-KF (called FLINAK)

$LiF\text{-}CaF_2$

Electrochemical separation processes under development:

Cathodic deposition method

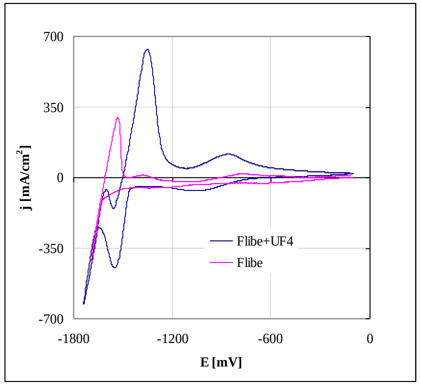
Anodic dissolution method

Laboratory research on electrochemical separation from fluoride molten-salt media

- The electrochemical measurements have been carried out in a laboratory nickel electrolyser under inert atmosphere in a three-electrode set-up.

- A specially designed reference electrode based on the Ni/Ni^{2+} red-ox couple was developed.

- Linear Potential Sweep Cyclic Voltammetry Method has been used as the measurement technique.



Evaluated red-ox potentials of selected actinides and lanthanides in various carrier salts

Red-ox couple	LiF-NaF-KF	$LiF-CaF_2$	$LiF-BeF_2$	LiF-NaF
	Ref. Ni/Ni^{2+} in FLINAKu	Ref.. Ni/Ni^{2+} in $LiF-CaF_2$	Ref. Ni/Ni^{2+} in $LiF-BeF_2$	Ref. Ni/Ni^{2+} in LiF-NaF
U^{3+}/U^{0}	-1.75	-1.90	-1.4	-1.4
U^{4+}/U^{3+}	-1.20	-1.40		(-0.8) – (-1.25)
U^{5+}/U^{4+}	+0.40	-		not-detected
U^{6+}/U^{5+}	+1.40	-		not-detected
Th^{x+}/Th^{0}	~ -2.00	-1.70	out of window	-
Th^{4+}/Th^{x+}	-0.70	-		-
Nd^{2+}/Nd^{0}	< -2.05	-2.00	out of window	out of window
Nd^{3+}/Nd^{2+}	~ -1.00	not-detected		-1.3
Gd^{2+}/Gd^{0}	< -2.05	-2.10	out of window	-1.35
Gd^{3+}/Gd^{2+}	~ -1.00	not-detected		-0.55
Eu^{3+}/Eu^{x+}	~ -0.75	not-detected		-0.2
Eu^{x+}/Eu^{0}	-1.95	< -2.30		out of window
Zr^{4+}/Zr^{x+}	-1.50	-		-
Zr^{x+}/Zr^{0}	-1.80	-		-
Sr^{2+}/Sr^{0}	< -2.05	-		-
La^{3+}/La^{0}	< -2.05	-	out of window	-
Pr^{3+}/Pr^{0}	< -2.05	-	out of window	-
Sm^{2+}/Sm^{0}	-0.8	-	-	out of window
Sm^{3+}/Sm^{2+}	out of window	-	-	-1.2

Separation possibilities of selected actinides and fission products (lanthanides)

Carrier melt	Separation presumed possible	Separation presumed impossible
FLIBE	U / Th U / Nd, Gd, Sm, Eu, Pr	Nd / Gd
FLINAK	U / Th U / Nd, Gd, La, Sm, Eu, Pr, Sr Th / La, Pr, Sr, Zr, Eu	Th / Nd, Gd Nd / Gd U / Zr
$LiF\text{-}CaF_2$	U / Th U / Nd, Gd, Sm, Eu, Pr Th / Nd, Gd, Sm, Eu, Pr U,Th / Nd, Gd, Sm, Eu, Pr	U / Nd Nd / Gd

Mutual link between reactor physics and chemical technology of on-line reprocessing

The mutual link between MSR physics and the on-line reprocessing is extremely strong.

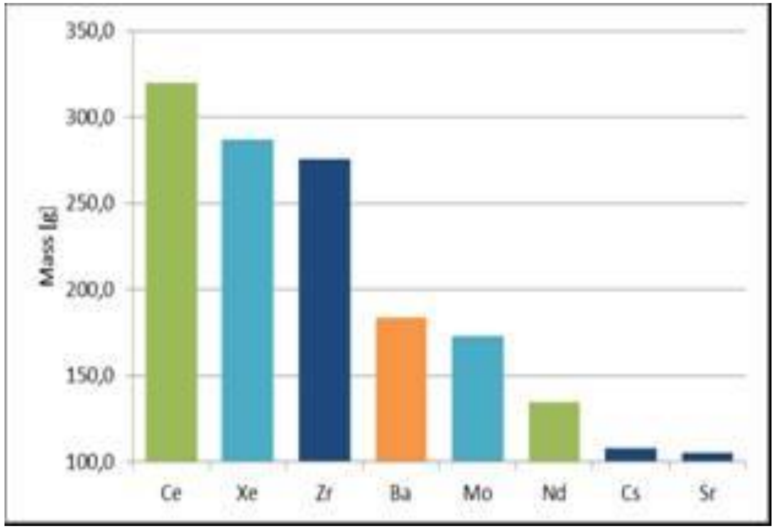
Instantaneous fissile power affects the capacity of on-line reprocessing; the rate of reprocessing has the retroactive effect on the quantity and concentration of fissile material. The rate of protactinium removal affects the breeding factor of the reactor system.

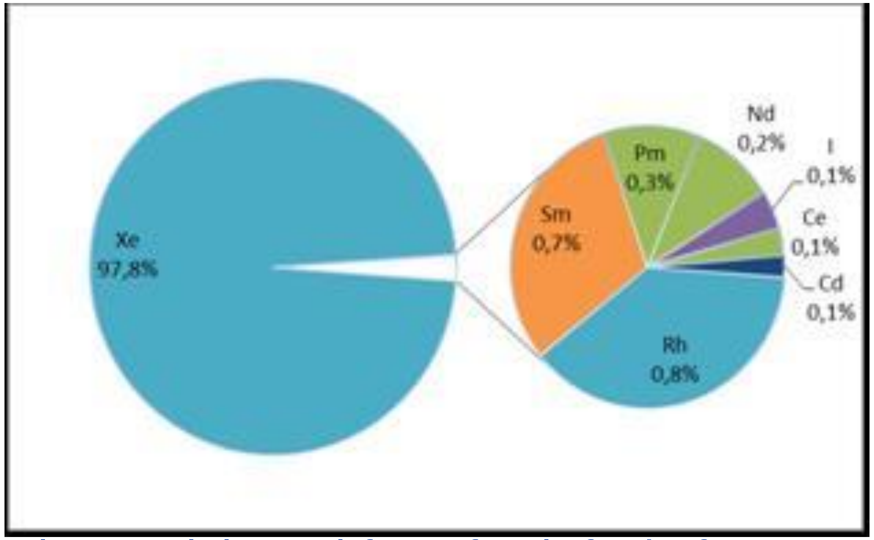
Another interconnection between chemistry and reactor physics is based on the fact that fission reaction is oxidative process, which must be chemically compensated to keep reductive conditions in the fuel circuit to minimize corrosion of structural material (nickel alloys).

If we wish is to maximize the breeding of ^{233}U, then our task is not only to extract protactinium as soon as possible, but also to identify the main neutron poisons among fission products in fuel circuit. It can be done on the basis of their production rates and neutron capture cross section.

The knowledge of these data can finally determine the technology of chemical partitioning and the details of the conceptual reprocessing flow-sheet.

Computation made by Monte Carlo code MCNPX v 2.7 with ENDF/B VII library (two-fluid MSR core configuration)

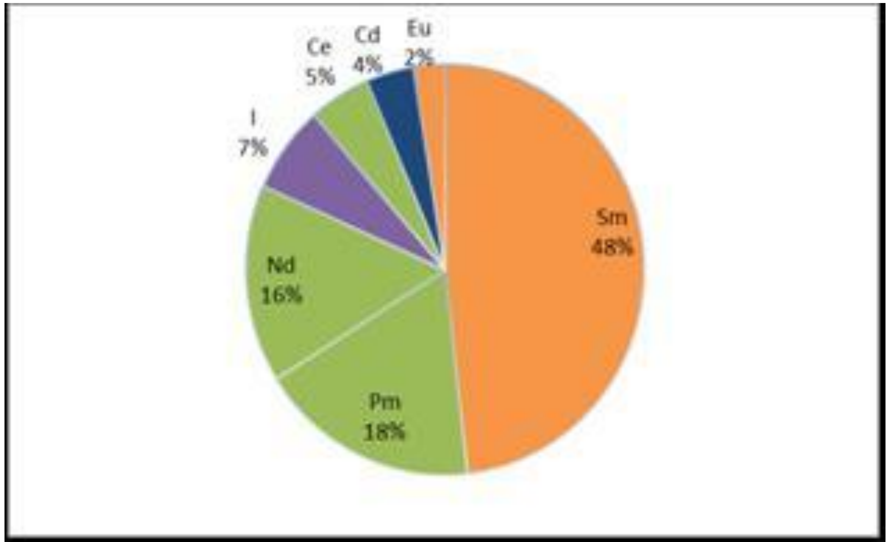




Figures: Mass production of main fission products originated from fresh fuel after one-day reactor operation

Distribution of fission products capture rate leaving the MSR core

Distribution of fission products capture rate entering the reprocessing unit



MSR System studies: Core design

There are two possibilities of the MSR reactor core design:

- One fluid (single fluid) system **– fissile and fertile materials are mixed together**
- Two fluid system **– with separate channels of fissile and fertile fuel components**

One fluid system:

- **Significantly simple construction – relatively low breeding factor (~1.04)**

Two fluid system:

- **Complicated reactor core design – however excellent breeding factor (~1.1 – 1.13)**

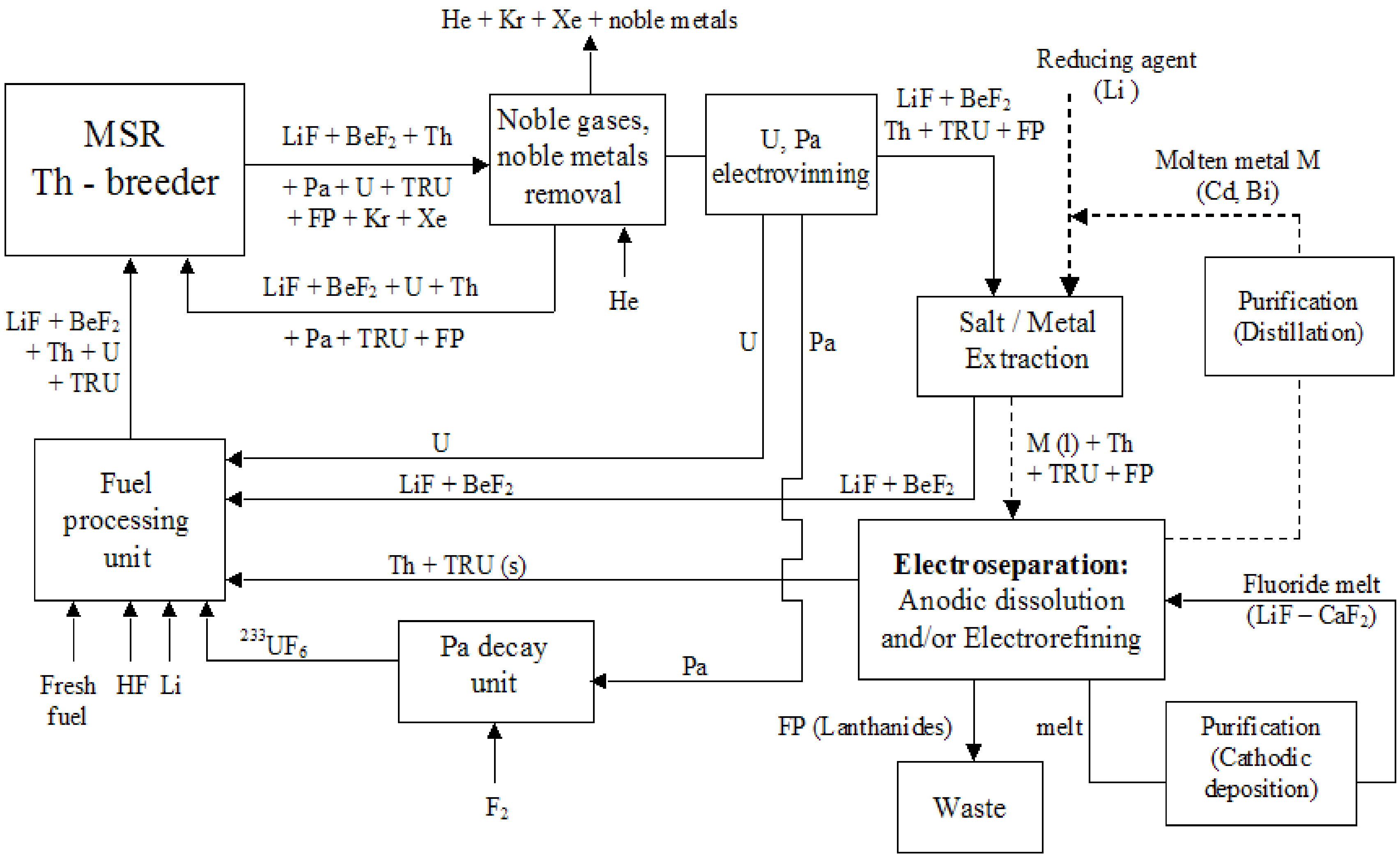
One fluid MSR on-line reprocessing design

- **To extract neutron poisons represented namely by fission products**
- **To separate freshly breeded fissile material ^{233}U or its precursor ^{233}Pa.**

As the fissile and fertile materials are present together in the single primary (fuel) circuit, the reprocessing technology must be able to separate all fuel and fission products components dissolved in the carrier salt.

Conceptual flow-sheet of MSR on-line reprocessing technology

One-fluid system



Two fluid MSR on-line reprocessing design

Fissile fluid circuit:

- **To extract neutron poisons represented namely by fission products**

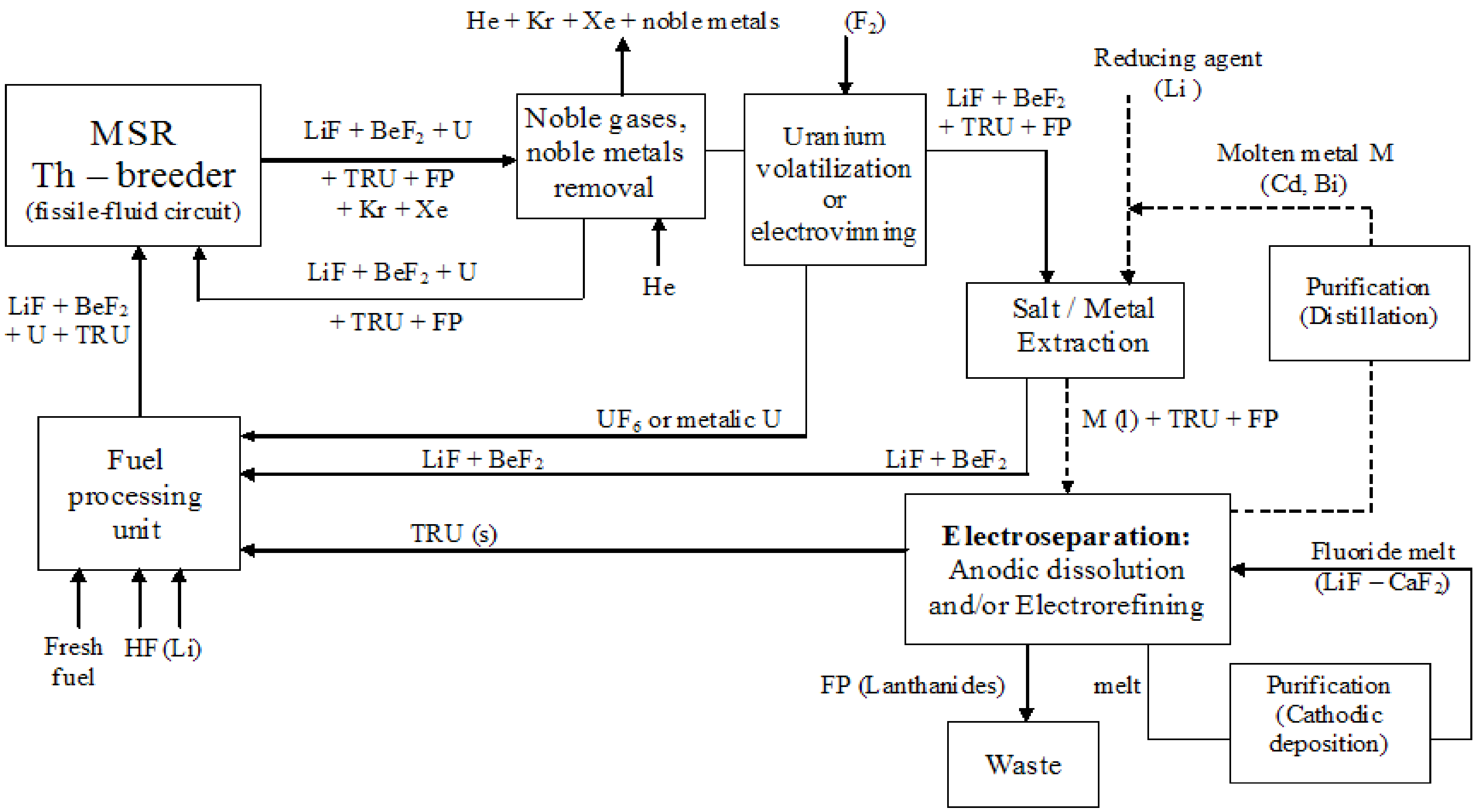
Fertile fluid circuit:

- **To separate freshly breeded fissile material ^{233}U or its precursor ^{233}Pa.**

As the partitioning technology used for the reprocessing of fissile-fluid circuit could be similar to those proposed for the one-fluid system (evidently without the Pa and Th separation), the partitioning of the fertile fluid circuit could be significantly simplified.

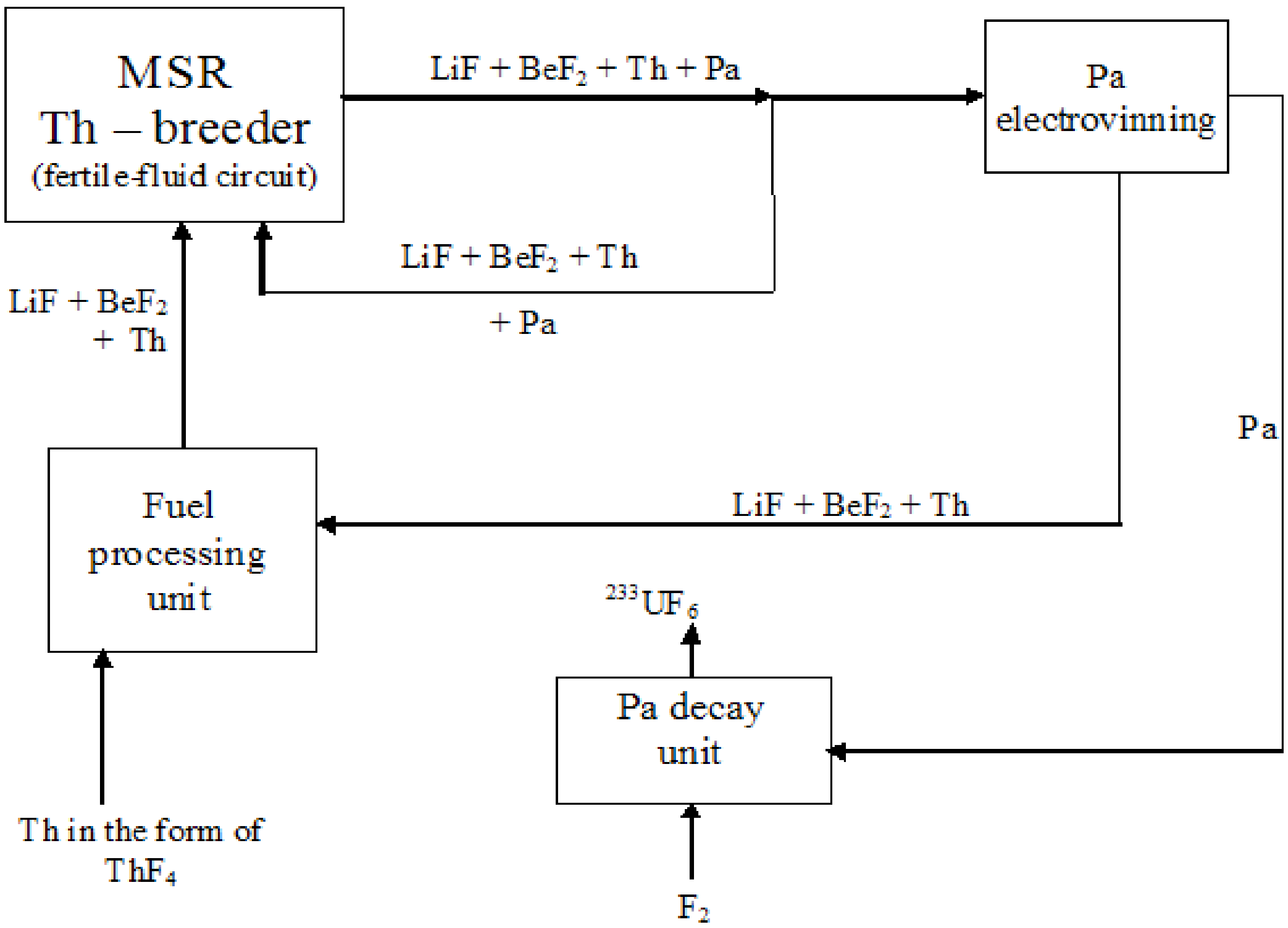
Conceptual flow-sheet of MSR on-line reprocessing technology

Two-fluid system, fissile fluid circuit



Conceptual flow-sheet of MSR on-line reprocessing technology

Two-fluid system, fertile fluid circuit



Current activities and future directions

Main current activities and future plans come from the bilateral agreement concluded between MoIT and US-DOE and focused on the FHR and MSR technology.

Based on the agreement, ORNL transferred about 75 kg of coolant salt (^{7}LiF-BeF_2) to Řež for experimental in-core neutronic studies (Evaluation of reactivity coefficients of FHR/MSR fluoride salt).

A joint program with US-DOE (ORNL) and MIT is now under preparation.

First collaborative project should be focused on the study of FHR/MSR neutronics:

“Evaluation of reactivity coefficients of FHR/MSR fluoride salt”

Conclusions

We have investigated selected areas of MSR system and Th – U fuel cycle technology.

- **reactor physics and chemistry, liquid fuel processing, on-line reprocessing techniques, development of structural materials (Ni alloys), design and manufacture of MS components (loops, impellers, valves, heat exchangers), system studies including (non-proliferation – physical protection)**

Future intentions:

- **New program scheduled for 2014 – 2016 which is under final preparation should take into account the MoIT – US-DOE agreement (MoU) regarding the FHR/MSR systems**
- **Of course, we would like to continue in our existing activities in international collaborative projects under the coordination of GIF, OECD-NEA, IAEA and EC-EURATOM**
- **We are fully aware that our personal, technical and financial possibilities are limited, so we would like to be a good and beneficial partner for the countries which plan to deploy the MSR technology.**

Thank you for your attention

Authors are greatly appreciative for the support of the Ministry of Industry and Trade which made this work possible.