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Fuel handling and waste issues for Molten Salt Reactors

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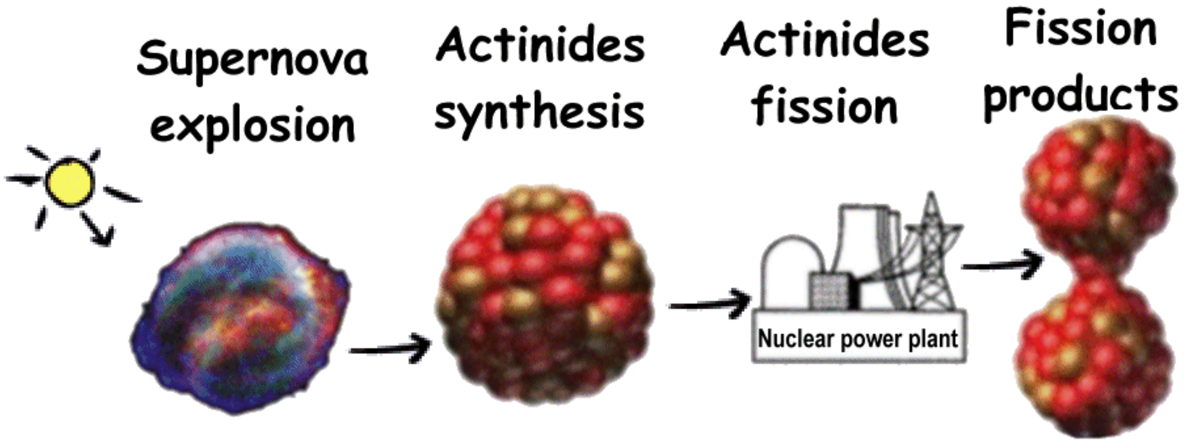
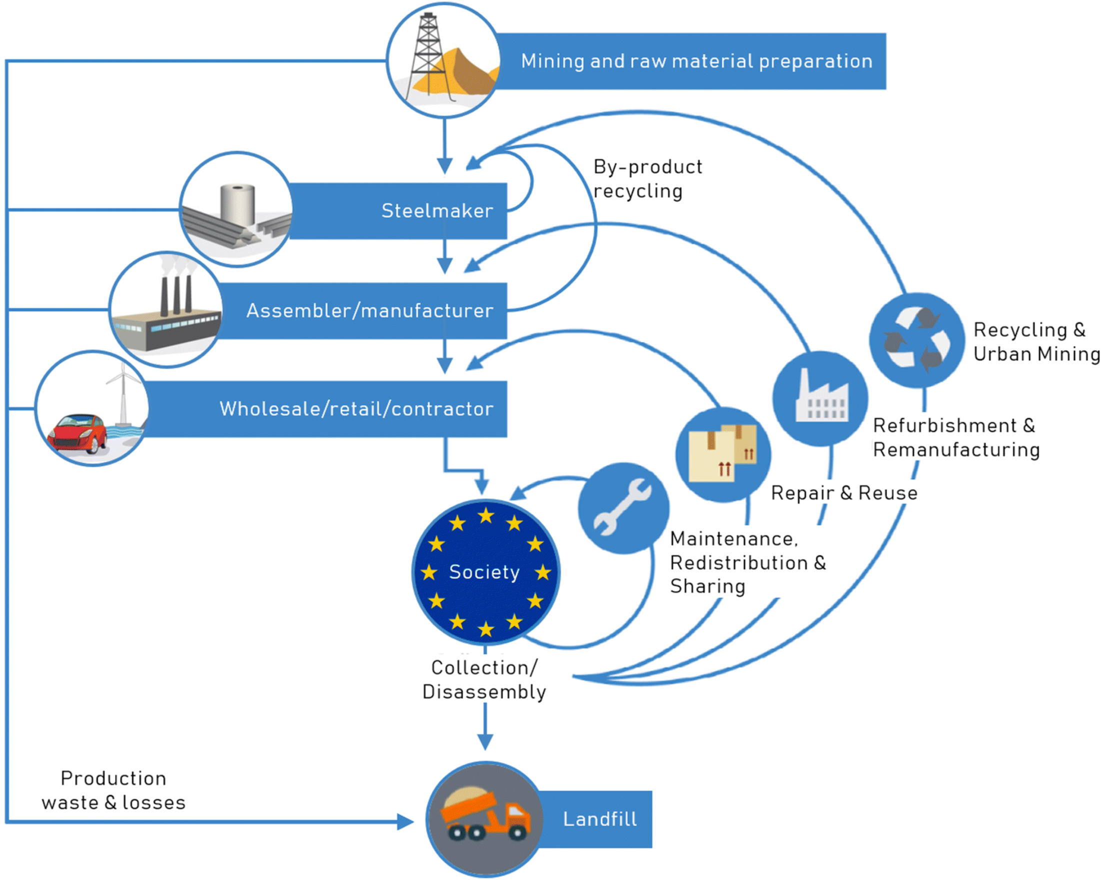
Fuel cycle, Closed cycle, and Closed fuel cycle

- **Fuel cycle** is a process chain to obtain **energy**.
- In **closed cycle** some substances (**re**)**cycle** and does not leave the cycle.
- **Closed cycle** is applicable to **resources**.
- **Nuclear fuel cycle** is generally **open**,
- but could be closed for actinides (**also in MSRs**).



"Open" nuclear fuel cycle

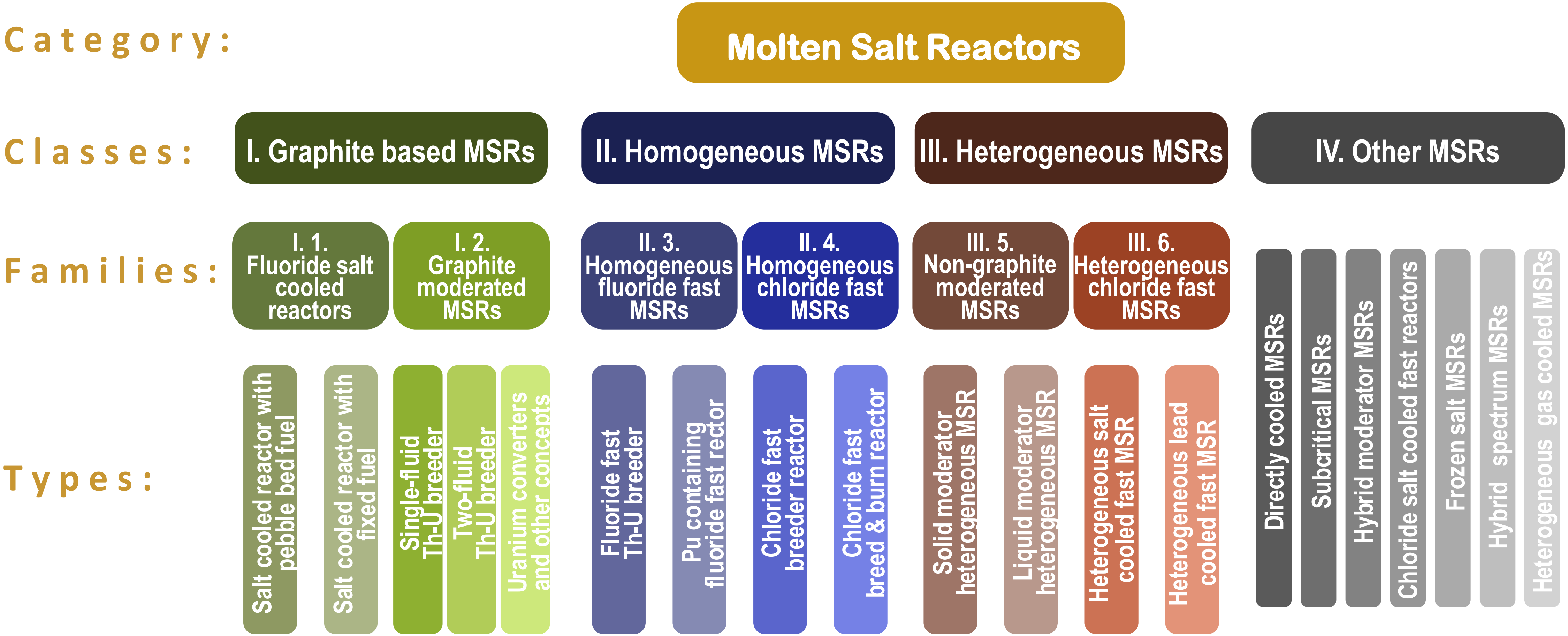
The actual waste are e.g. lanthanides.



Circular economy for natural resources.

EMF (2017) The circular economy—a wealth of flows. Ellen McArthur Foundation, Cowes

Molten Salt Reactors as a reactor category



Category: Molten Salt Reactors

Classes:

- I. Graphite based MSRs
  - I. 1. Fluoride salt cooled reactors
    - Salt cooled reactor with pebble bed fuel
    - Salt cooled reactor with fixed fuel
  - I. 2. Graphite moderated MSRs
    - Single-fluid Th-U breeder
    - Two-fluid Th-U breeder
    - Uranium converters and other concepts
- II. Homogeneous MSRs
  - II. 3. Homogeneous fluoride fast MSRs
    - Fluoride fast Th-U breeder
    - Pu containing fluoride fast rector
  - II. 4. Homogeneous chloride fast MSRs
    - Chloride fast breeder reactor
    - Chloride fast breed & burn reactor
- III. Heterogeneous MSRs
  - III. 5. Non-graphite moderated MSRs
    - Solid moderator heterogeneous MSR
    - Liquid moderator heterogeneous MSR
  - III. 6. Heterogeneous chloride fast MSRs
    - Heterogeneous salt cooled fast MSR
    - Heterogeneous lead cooled fast MSR
- IV. Other MSRs
  - Directly cooled MSRs
  - Subcritical MSRs
  - Hybrid moderator MSRs
  - Chloride salt cooled fast reactors
  - Frozen salt MSRs
  - Hybrid spectrum MSRs
  - Heterogeneous gas cooled MSRs

Families:

Types:

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Adopted from: IAEA Technical Report Series, Status of Molten Salt Reactor Technology, document in preparation, International Atomic Energy Agency, 2021.

5 major fuel cycle types foreseen for MSR

Resources utilization: **<1% of nat. U**

I. **Enriched uranium burning** *(^{235}U burning)*
  - Enrichment level can range from 0.7% till 20%.
  - The cycle is generally open and “waste” intensive.
  - However, irradiated U and generated Pu can be recycled.

II. **Closed Th-U cycle** *(^{232}Th burning in closed cycle)*
  - Actinides recycling in a breeder reactor fueled by ^{232}Th.

<95% of ^{232}Th

III. **Closed U-Pu cycle** *(^{238}U burning in closed cycle)*
  - Actinides recycling in a breeder reactor fueled by ^{238}U.

<95% of nat. U

IV. **Breed-and-burn U-Pu cycle** *(^{238}U burning in open cycle)*
  - Open cycle, Ac. are not recycled, but the reactor acts as a breeder.
  - “Waste” intensive cycle, however fuel can be reused by another reactor.

cca 20-30% nat. U

V. **Synthetic actinides burning**
  - Cycle dedicated to minimization of existing synthetic actinides.

not relevant

- **Combination or transition between above cycles**
  e.g. actinides from I. or V. can acts as an initial
  or add on fuel for II. - IV. or vice versa.

5 fuel cycle performance parameters

I. **Breeding capability**
  - How many neutrons can be captured by ^{232}Th or ^{238}U so that the reactor is still critical.
  - BTW: Uranium enrichment reduces ^{238}U capture, hence also the breeding capability.
  - It is about neutron economy.

II. **Achievable burnup**
  - Is limited by Fission Products (FPs) neutron capture and by fuel irradiation stability.
  - Depends on initial reserve of fissile material and its renewal (breeding capability).

III. **Initial fissile mass**
  - It is determined by neutron economy and spectrum type of the reactor.
  - Higher burnup may impose higher initial fissile mass reserve.

IV. **Means of criticality maintenance**
  - Ac. irradiation and FPs creation results in reactivity oscillations / swing.
  - Compensation option for reactivity swing differ between reactor types.

V. **Transmutation capability**
  - "Neutron costs" and "speed" of synthetic actinides fission.
  - Synthetic Ac. compatibility with the fuel and fabrication process.

MSR: possible absence of structural materials

Radiation stability of the salt

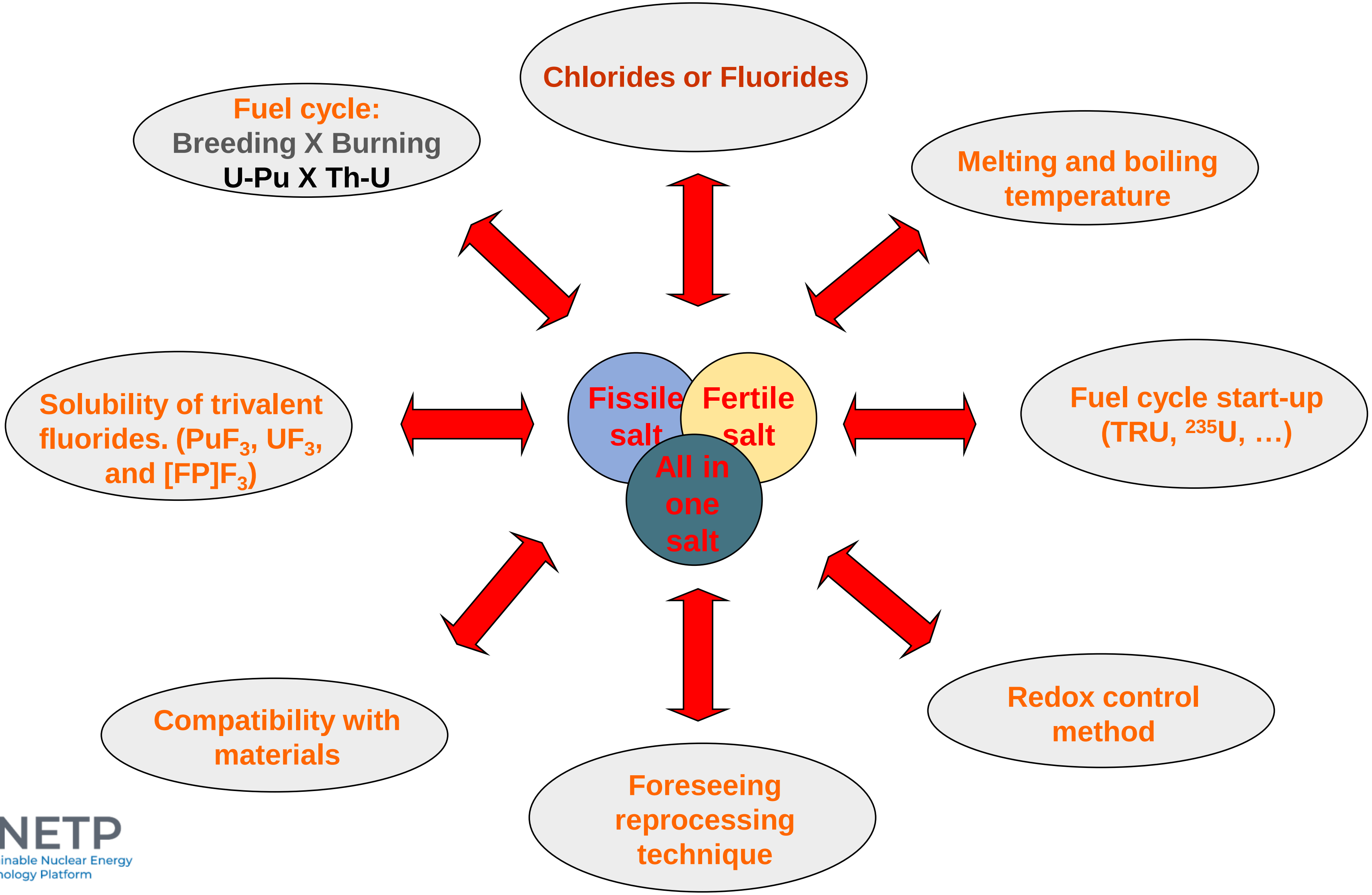
Online refuelling and removal of some FPs

Possible liquid fuel reshaping / draining

Absence of fabrication
Solubility of actinides?

Krepel, J., Self-sustaining breeding in advanced reactors: Comparison of fuel cycle performance, Encyclopedia of Nuclear Energy, (Greenspan, E., Ed.), Elsevier, 2021.

Selection of the salt



Chlorides or Fluorides

Fuel cycle:
Breeding X Burning
U-Pu X Th-U

Melting and boiling temperature

Solubility of trivalent fluorides. (PuF_3, UF_3, and $[FP]F_3$)

Fissile salt

Fertile salt

All in one salt

Fuel cycle start-up (TRU, ^{235}U, …)

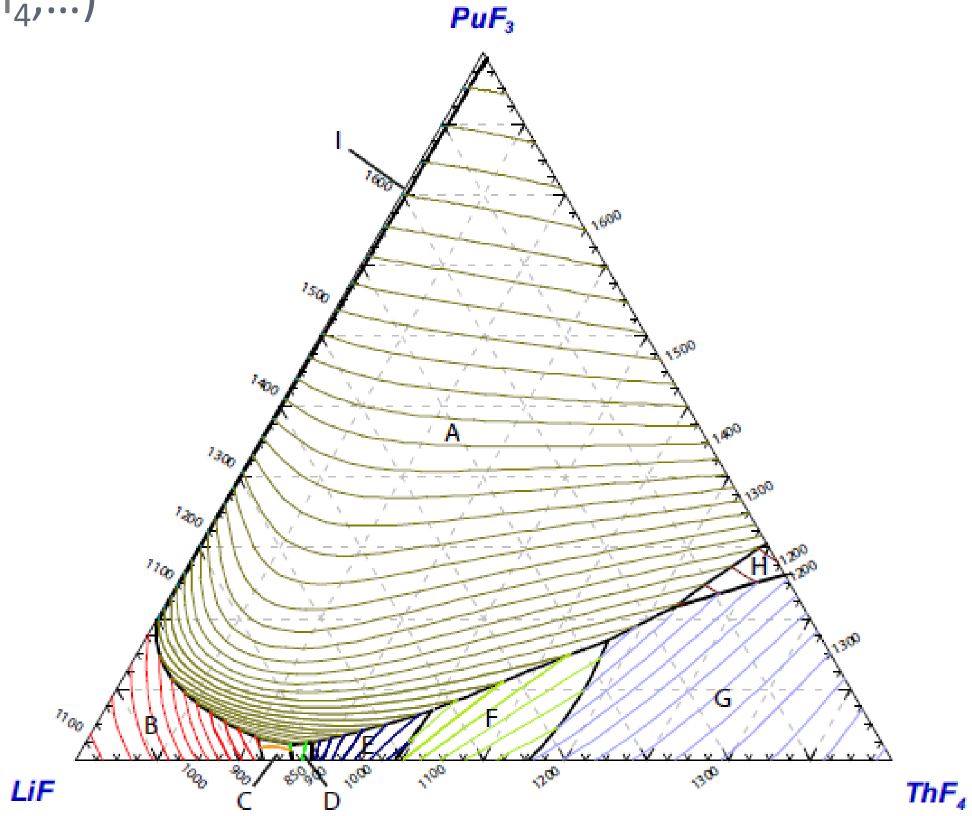
Compatibility with materials

Foreseeing reprocessing technique

Redox control method

Salt melting temperature as major parameter

- Typically eutectic mixture of carrier salts (LiF, BeF_2, NaF, LiCl, NaCl,…) and actinides salts (ThF_4, UF_4, PuF_3, $PuCl_3$, UCl_3, $ThCl_4$,…)
- **MSRE** salt, **$T_{melt.}$=432 ° C**
  65%LiF - 29.1BeF_2 - 5%ZrF_4 - 0.9%UF_4
- **MSBR** , Th-U equilibrium cycle, **$T_{melt.}$=500 ° C**
  71.7%LiF - 16%BeF_2 - 12%ThF_4 - 0.3%UF_4
- **MSFR**, Th-U equilibrium cycle, **$T_{melt.}$=560 ° C**
  78%LiF - 17.6%ThF_4 - 4%UF_4 - 0.2%PuF_3
- **MSFR**, Pu started Th-U cycle, **$T_{melt.}$=625 ° C**
  78%LiF - 16%ThF_4 - 6%PuF_3
- **MCFR**, Pu started U-Pu cycle, **$T_{melt.}$=565 ° C**
  60%NaCl - 35%UCl_3 - 5%$PuCl_3$
- **MCFR**, Pu started Th-U cycle, **$T_{melt.}$=425 ° C**
  55%NaCl - 39%$ThCl_4$ - 6%$PuCl_3$
- Generally **solubility limits** (e.g. PuF_3) and **actinides density** compete with **melting temperature**.

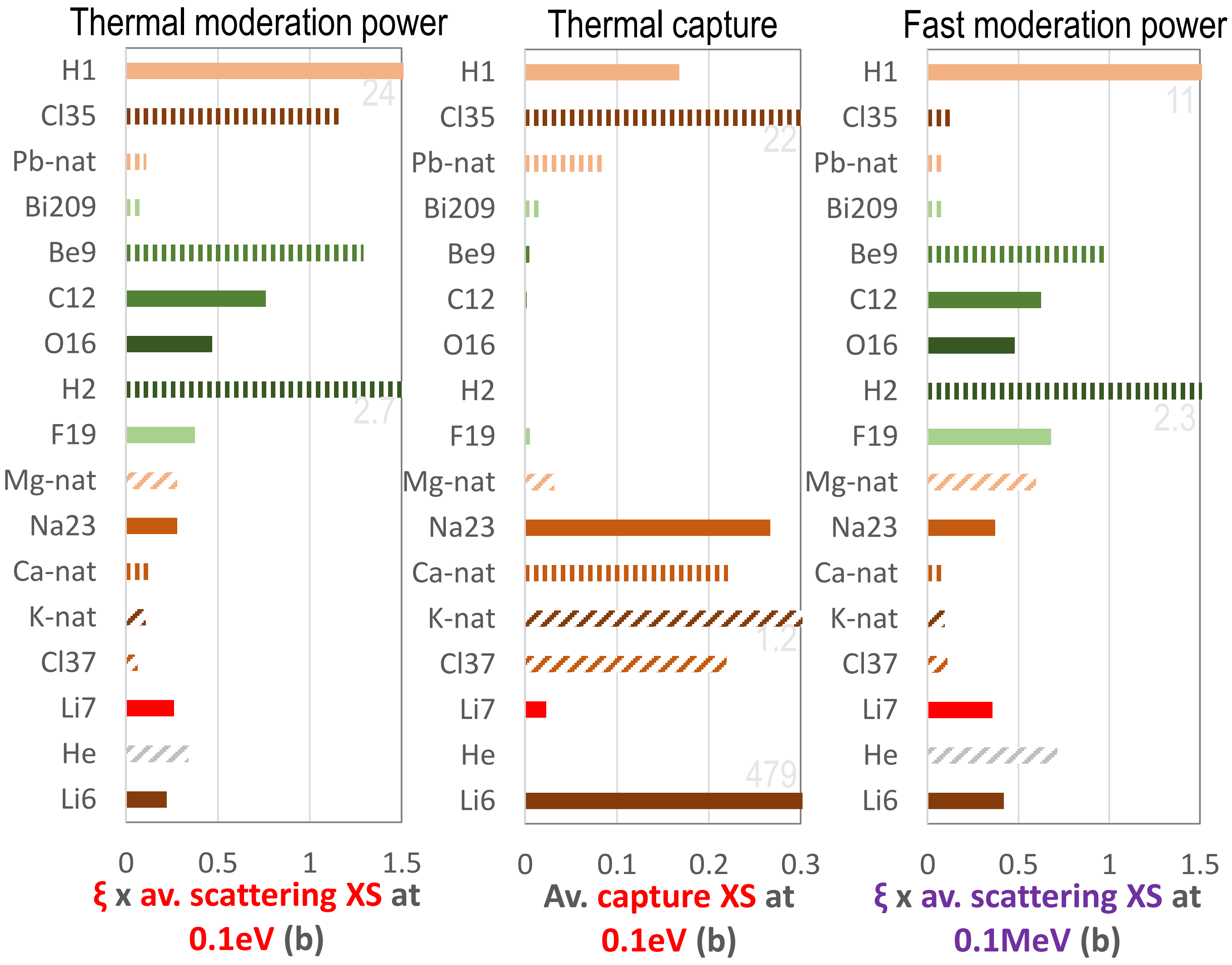


LiF-ThF_4-PuF_3 ternary phase diagram w/ fixed 1% mol UF_4 concentration

E. CAPELLI et al., "Thermodynamic Assessment of the LiF–ThF4–PuF3–UF4 System," J. Nucl. Mater., ***462****, 43 (2015).*

Basic nuclear properties of salt nuclides

Chlorides salts: 1) high capture in thermal spectrum,
2) very limited scattering in fast spectrum.



ξ x av. scattering XS at 0.1eV (b)

Av. capture XS at 0.1eV (b)

ξ x av. scattering XS at 0.1MeV (b)

	Moderator:	Suppressing fast neutrons:	Breeding in thermal spectrum:	Breeding in fast spectrum:
H1	Yes*	Yes	No	No
Cl35	No	No	No	No
Pb-nat	No	No	No	Yes
Bi209	No	No	No	Yes
Be9	Yes	Yes	Yes	No
C12	Yes	Yes	Yes	No
O16	No	No	Yes	Yes
H2	Yes	Yes	Yes	No
F19	No	Yes**	Yes	Yes***
Mg-nat	No	Yes**	Yes	Yes
Na23	No	No	No	Yes
Ca-nat	No	No	No	Yes
K-nat	No	No	No	Yes
Cl37	No	No	No	Yes
Li7	No	Yes**	Yes	Yes
He4	No	Yes	Yes	Yes
Li6	No	Yes**	No	No

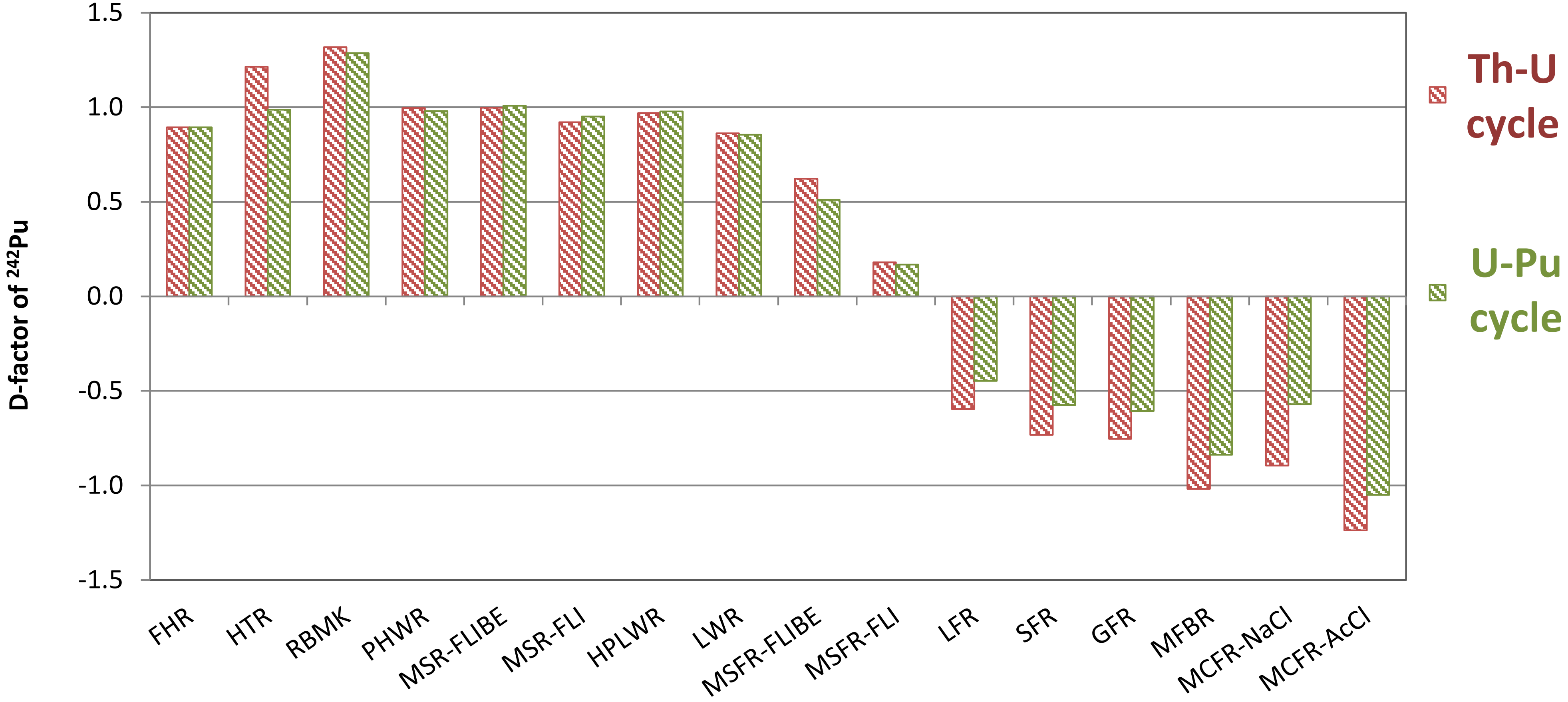
*Substantial capture XS

**Broad Scattering resonances around 0.1MeV

***However spectrum is quite soft.

Neutron balance of ^{242}Pu burning (D-factor)

D-factor: how many neutrons it costs to burn one atom in given reactor.



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Krepel, J., Self-sustaining breeding in advanced reactors: Comparison of fuel cycle performance, Encyclopedia of Nuclear Energy, (Greenspan, E., Ed.), Elsevier, 2021.

Liquid versus solid fuel

https://www.saldo.ch/artikel/d/tankstellen-strecken-benzin-und-diesel-mit-guenstigem-biosprit/

http://www.chemistryexplained.com/Ce-Co/Coal.html

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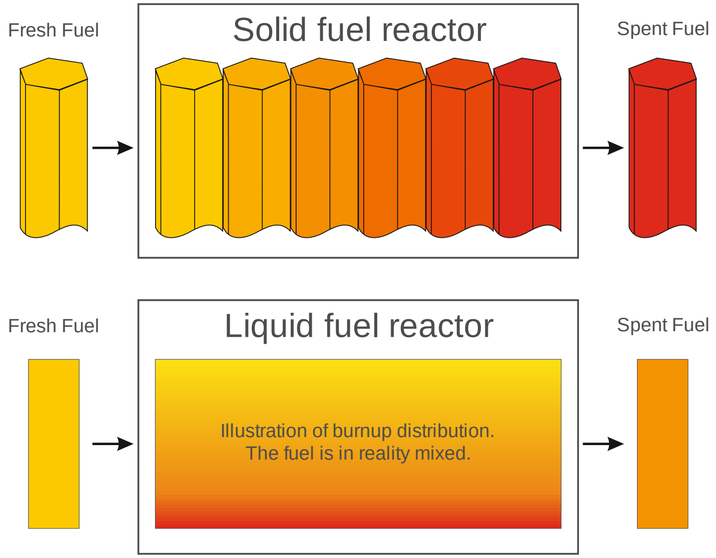
Fuel manipulation

In solid fuel reactors the fuel can be:

1. Loaded in to the core.
2. Discarded from the core.
3. Reshuffled (optional).
4. Reprocessed ex-situ, with years of delay.



Basic operation with the liquid fuel:

1. Salt refilling into the core. (direct impact to the core neutronics).
2. Salt removal from the core. (no direct impact to the core neutronics).
3. Salt cannot be “reshuffled”, unless the reactor has more than one salt fluids.
4. Salt cleaning inside of the core. (direct impact to the core neutronics).
5. Salt cleaning outside of the core. (no direct impact to the core neutronics).

Security & safeguards

- Liquid fuel is not divided into discrete and numbered assemblies.
- In-situ fuel treatment may include techniques, where weapon great materials are accessible (e.g. volatilization of ^{233}Pa and its decay to ^{233}U).
- At the same time, development of the MSR with the respective reprocessing methods is not the simplest way of obtaining these materials.
- The fuel treatment steps can be organized so, that the removal is impossible or demanding.
- The quality of the fissile material could be deteriorated by denaturation (addition of other isotopes of the same element).

Criticality safety

- In **solid fuel fast breeder** reactor, the coolant **void effect** is **positive**.
  (positive coolant density or actually coolant temperature effect)

- It may be **positive** also in reactor with **separated coolant** and moderator (**PHWR, RBMK**).

- In **fast solid fuel** reactors the **fuel compaction** can introduce **positive reactivity**.

- **Thermal solid fuel** reactor may suffer by **Xenon poisoning**.

- Solid fuel convertors (e.g. LWR) need **initial reactivity excess** in fresh fuel, which is **compensated by absorber** (in fuel itself, **in coolant and in control rods**).

- **Liquid fuel MSR** can be designed as **a breeder with negative void effect**.
  (fuel and coolant are coupled = Doppler and respective density effects are also coupled)

- **Liquid fuel compaction** = collection can be **avoided by design**.

- **Xenon** as a gas is **can be removed** from liquid fuel reactor.

- **Reactivity excess** is not necessary, liquid fuel **composition can be controlled** / adjusted.

Fuel irradiation

In solid fuel reactors the irradiation time is limited by:

1. Cladding lifetime.
2. Fissile element load in convertors or burners (breeders can be self-sustaining).
3. Gaseous FPs pressure.
4. Core poisoning by FPs neutron capture.

In liquid fuel reactor:

1. There is no cladding in some MSR families.
2. Fissile elements can be continuously added if needed.
3. Breeders are self-sustaining and fertile elements can be continuously added.
4. Some FPs can be continuously removed form the core.
5. Remaining FPs are still poisoning the core by neutron capture.

In MSR breeder the **only reason** for fuel reprocessing is **FPs poisoning**.

Gaseous fission products

In solid fuel:

1. There is **dedicated plenum** in the fuel pin to collect gaseous FPs.
2. The gas **pressure** in the pin is **growing** with burnup.

In liquid fuel reactor:

1. Dedicated plenum or collector for the gaseous FPs can be included in the primary circuit (fuel circuit).
2. There are also concepts with dedicated **off-gas system** to **prevent pressurization.**
3. This system can be passive (gas suction) or active (Helium sparging).
4. Active system requires set of filters to separate He and FPs, at the same time, it can help to remove semi-noble metals.
5. In both cases safe storage or **immobilization of the FPs** will be needed.

Reprocessing techniques and strategies

Fuel salts components:

1. Carrier salt (LiF, NaCl,…)
2. Fertile actinides (^{232}Th and ^{238}U).
3. Fissile actinides (^{233}U and ^{239}Pu).
4. Minor actinides (MA).
5. FPs.

Salt treatment / reprocessing techniques:

- Gaseous and volatile FPs removal (off-gas system).
- Metallic FPs removal (sponge filter or by off-gas sys.).
- Molten salt / liquid metal reductive extraction.
- Electro-separation processes.
- Compound evaporation or possibly precipitation.
- Fluoride volatilization techniques, fluorination of the molten salt mixture.

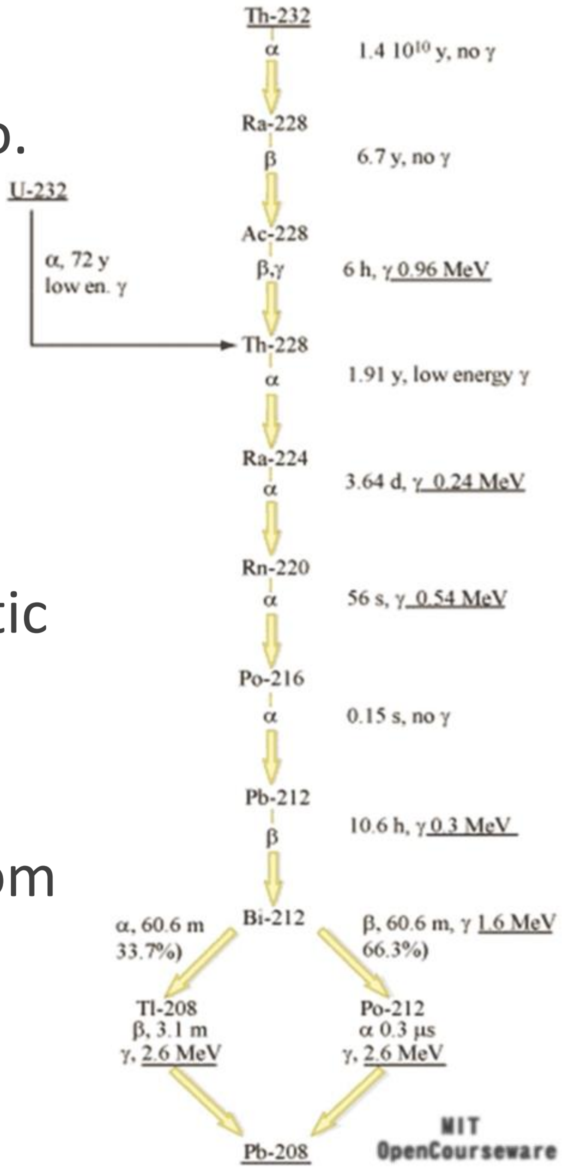
Reprocessing strategies:

Salt removal from the core	Removed salt share	Fissile fuel recycling	Fissile fuel return after reprocessing	Carrier salt cleaning	Carrier salt return after reprocessing	Reprocessing waste immobilization
Continuous or Batch-wise	From 0.1% to whole salt volume	In-situ or Ex-situ	ASAP or with months or years of delay	In-situ or Ex-situ	ASAP or with months or years of delay	In-situ or Ex-situ

Recycling & fabrication issues

- **Th-U cycle issue**: Hard gamma, e.g. from ^{212}Bi, ^{208}Tl, and ^{212}Po.
  - Pyrochemical reprocessing methods can simplify the treatment of radioactive molten salts (**dust free, remote operation,…**).

- **U-Pu cycle issue**: Minor actinides (e.g. Am, Cm) emits energetic alpha and Cm also neutrons. It is difficult to **fabricate** solid **pellets** with **internal heat source**.
  - Molten salt as liquid fuel can accommodate heat source from MA in U-Pu cycle (**no fabrication,** liquid is **simpler to cool**).



MIT OpenCourseware

Waste treatment

- MSR waste could have several forms.
- Off-gas system may include gaseous and metallic FPs.
- Reprocessing unit may include separated lanthanides.
- In some cases fuel salt will be not treated during the operation.
- The discharged salt can be immobilized by e.g. Synroc technology.
- This “synthetic rock” is based on crystalline or mineral phases.

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