Early MSR Projects: Molten Salt Fuels and Technology Gaps

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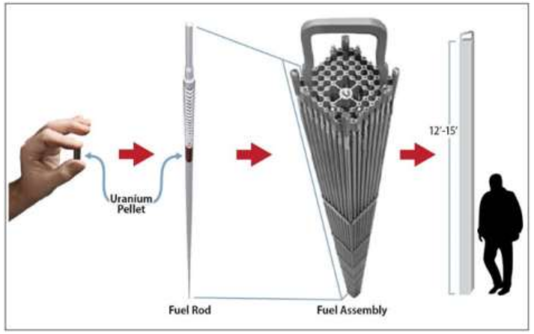
- 2015-2020 - ROSATOM's representative (diplomatic) to the International Organizations in Vienna and Adviser of Rosatom's CEO
- 2011-2015 - IAEA Deputy Director General, Head of the Department of Nuclear Energy
- 2006 - 2011 - Director General of the Research Institute of Atomic Reactors (RIAR) in Dimitrovgrad, Russian Federation,
- 1982 – 2011 – different positions in RIAR (engineer, researcher, head of laboratory, head of division, deputy DG).
- Graduated in chemistry from Moscow State University in 1982, PhD from 1998

- Main areas of R&D activity: the nuclear fuel cycle subjects, including fast reactors nuclear fuel technology and performance, **pyro-processing for manufacturing and recycling of nuclear fuel, high level wastes, molten salt chemistry of actinides,** radionuclide technologies, research reactors applications, design, safety and other aspects of nuclear R&D facilities.
- Expert on international cooperation, political and strategic aspects of nuclear energy as well as nuclear infrastructure.
- Co-author of more than 170 scientific publications, and invited professor and lecturer

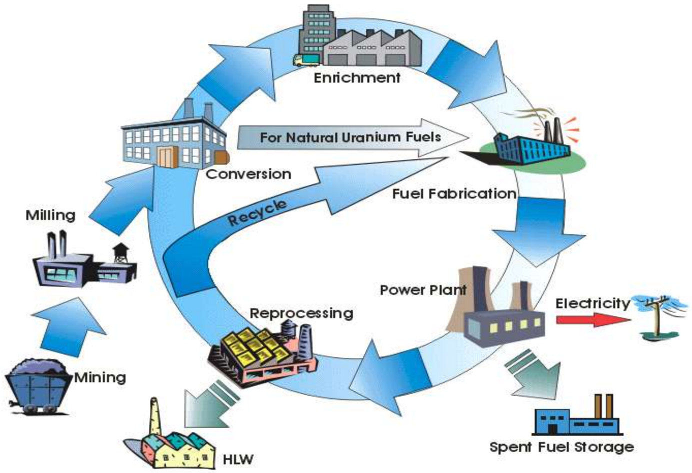
Preamble

Nuclear Fuels Design. Coolants for Nuclear Reactors



Traditional approach for current, advanced and innovative nuclear power reactors:

- Main principal for Nuclear fuel design - **solid,** with **stable cladding**, avoiding interraction with coolant.
- Manufacturing, reprocessing and refabrication are **technologically separate** lines outside of nuclear reacotrs
- Main principal for coolant selection – **clear liquid** (water or other dielectric, liquid metals, molten salts, etc.) or gas. **No interactions** with structural materials.

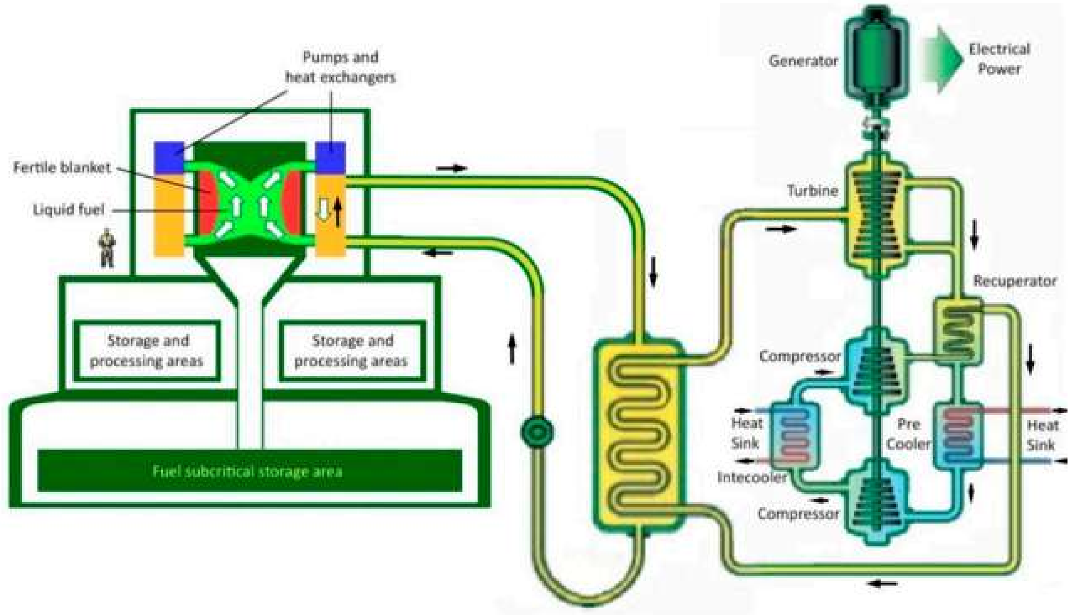


Molten salt fuel – ionic liquid (high melting point) with dissolved fissile and fertile nuclear materials (U, Pu Th, minor actinides)

Molten Salt Reactors – GenIV technology

- MSR – is only one experimentaly demonstated conception where reactor and fuel cycle combined as one system
- MSRs: systems with liquid fuel, molten fluoride (chloride) salts, circulating in primary circuit
- Actinide-free molten salt in secondary circuit
- Online fuel reprocessing instead of fuel fabrication - (partial removal of fission products and correction of actinides content)
- Operation either as breeders (Th-U or U-Pu cycle), as nuclear waste incinerators (transmuters)
- Thermal (graphite moderator) or Fast neutron spectrum
- Typical fuel : Fluorides of actinides dissolved in a carrier salt, such as $^{7}LiF\text{-}BeF_2$ or chlorides of actinides dissolved in a carrier salt, such as $LiCl\text{-}NaC\text{-}MgCl_2$.

Nuclear reactor and Closed Fuel cycle in “one package”



What are “Molten Salts”

What are “Molten Salts”, actually?

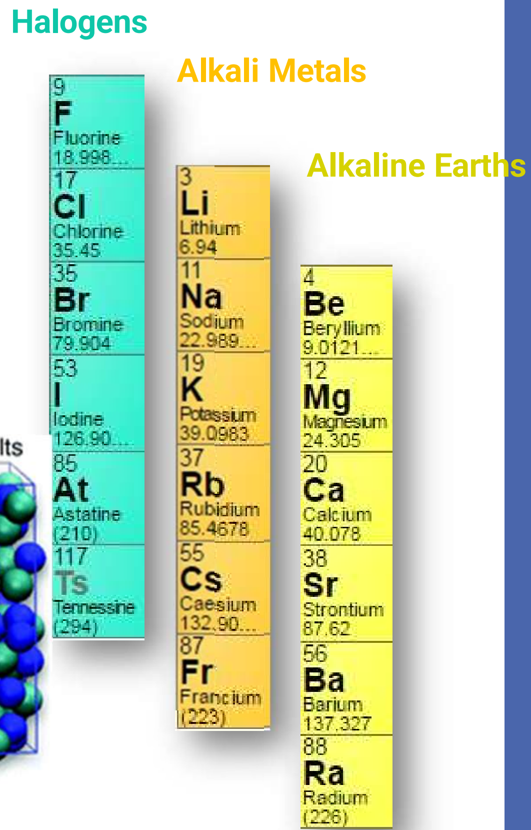
Salts are **ionic compounds** formed from a combination of **electronegative** and **electropositive ions**

At elevated temperatures salts **liquefy** and transform into ionic liquid are termed “molten salts”

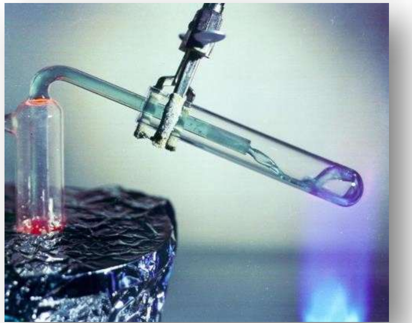
Molten salts are media for many chemical technologies – Al, Mg, Li, Na, Ca and REE production, refining of REE, material treatment, coating, etc.

Pyro-process for nuclear fuel recycling is molten salt technology too.

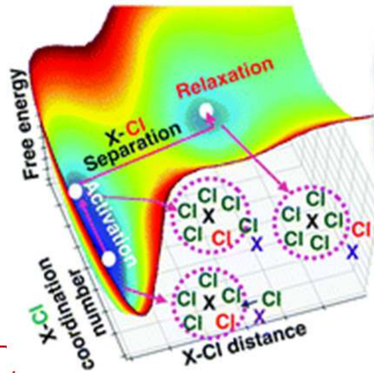
Halide salts are ionic compounds formed from the combination of a **halogen ion** (anion - electronegative) and electropositive ion (cation) – commonly, but not exclusively, **alkali metals** or **alkaline earths**



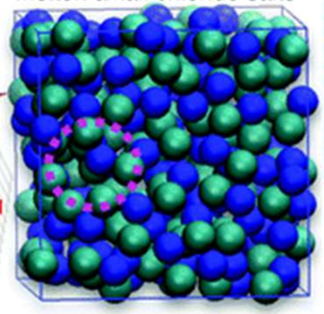
Examples: LiF, BeF_2, $MgCl_2$, NaCl (table salt), ZrF_4, ThF4, UF_4, UCl_3



(left) Solid “Frozen” and (right) Liquid “Molten” $2LiF\text{-}BeF_2$ salt



Molten alkali chloride salts

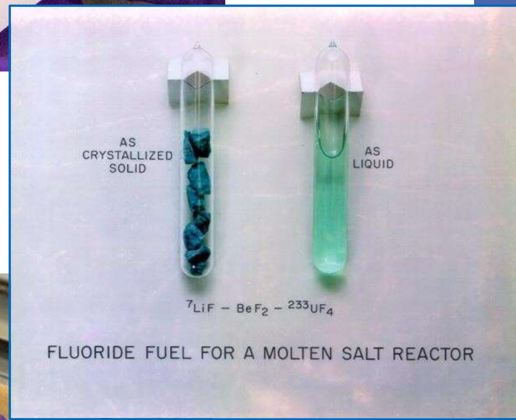


What is so special about salts ?

Compared to water, liquid metals, or gas ?

- High solubilities for Th, U and Pu compounds
  (= we can make nuclear fuel out of them !)
- High melting point and high boiling points
  (= Low vapor pressure at operating temperatures)
- Low chemical potential energy
  (= no vigorous reactions with air or water)
- Large heat capacity, low conductivity, relatively high viscosity
- Compatible with Ni-base alloys, graphite and some ceramic
- Stable thermodynamically, no radiolysis at high temperature
- Compatible with chemical processing
  (= MSRs can have "native" fuel reprocessing)

FLiBe Salt

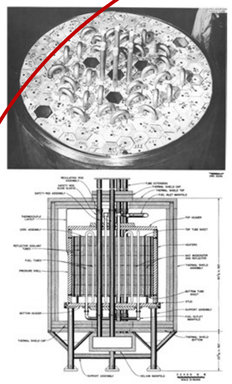


Plutonium Chloride

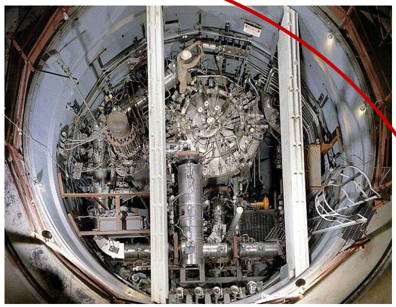
Molten Salt Reactors - Unique Experiments

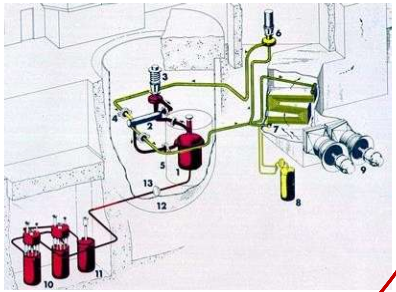
Basis for further development of MSRs

Brief History of MSR Experiments



Aircraft Reactor Experiment (ARE), USA

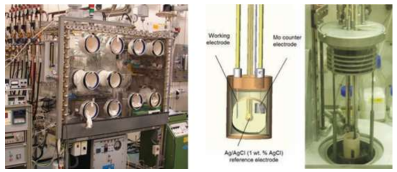




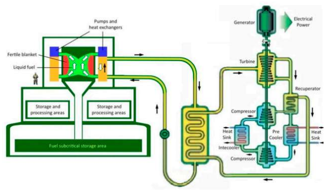
Molten Salt Reactor Experiment (MSRE), USA

MSBR Project, USA

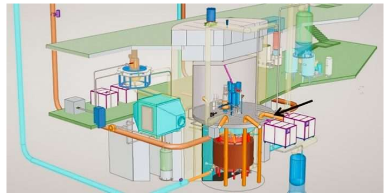
$LiF/BeF_2/ThF_4/UF_4$



Fuji MSR



GenIV



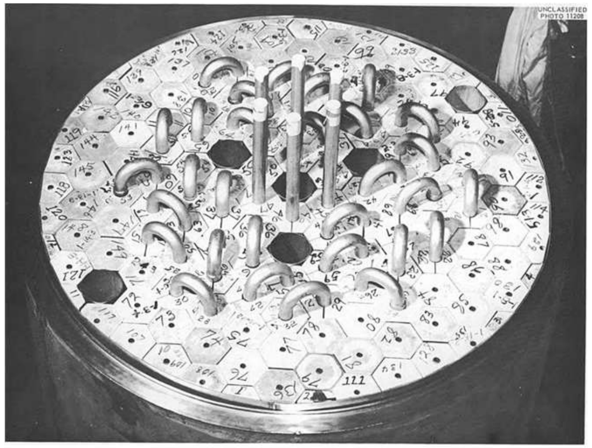
TMSR-LF1 Test Reactors, China

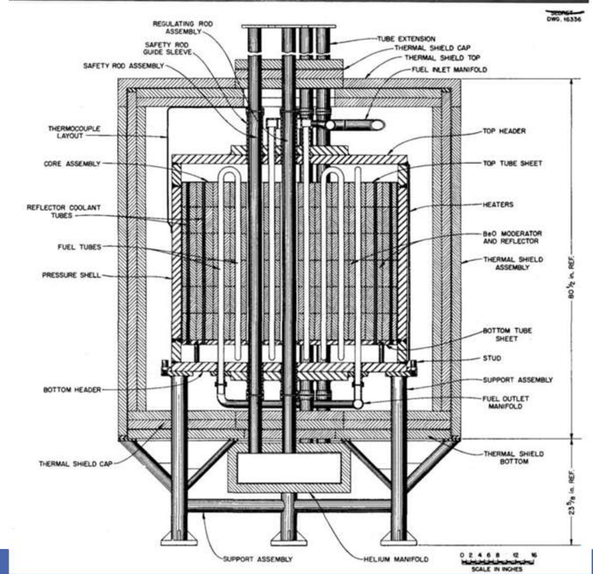
R&D R&D R&D R&D R&D

1954 1965-69 1970s 1980s 1990s 2000s 2010s 2020s

History: Aircraft Reactor Experiment (ARE)

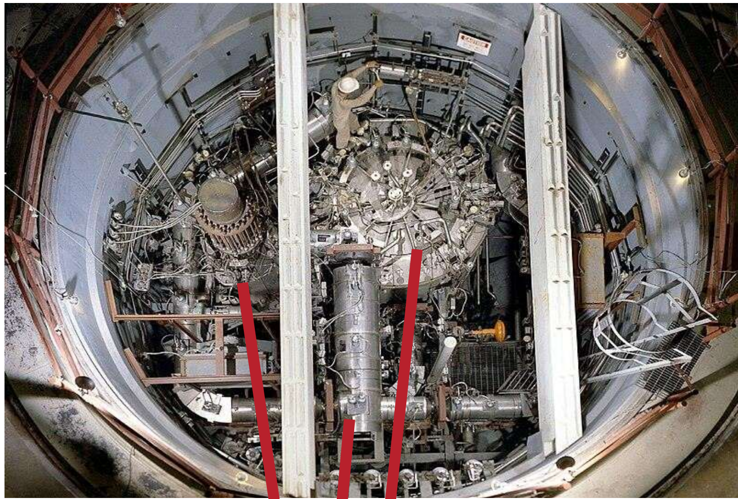
- **First molten salt reactor** was built (1954) for military purposes (aircraft nuclear propulsion) at Oak Ridge National Laboratory
- Beryllium Oxide (BeO) moderated, **NaF-ZrF_4-UF_4** fueled
- Liquid sodium (Na) and helium (He) cooled
- Structural material - Inconel 600
- Operated for 10 days (3 EFPD) up to 2.5 MW_{th} power
- Reached temperatures of ~800°C, a record at the time
- Demonstrated the feasibility of operating a MSR
- Exhibited no measurable Xenon reactivity effect !
- Demonstrated the practicality of **load-following** by **varying heat extraction on the secondary side**

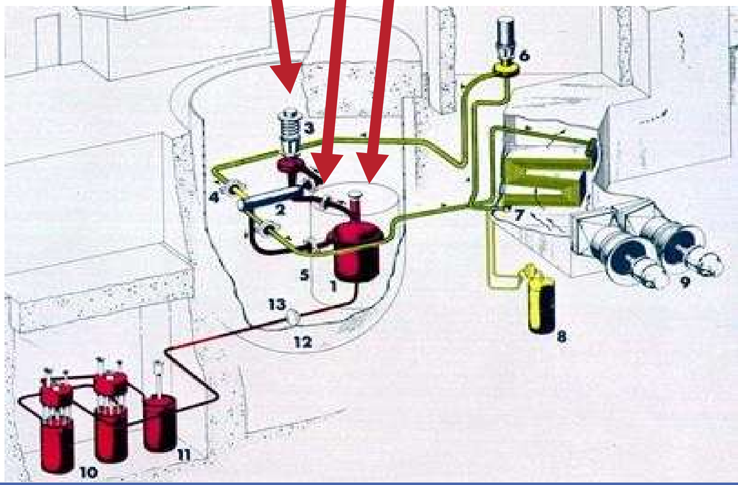




History: Molten Salt Reactor Experiment (MSRE)

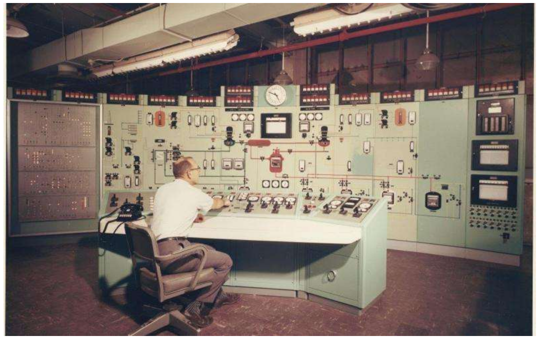
- **Second molten salt reactor** was built (1965) at ORNL
- Graphite moderated, ^{7}**LiF-BeF$_2$-ZrF$_4$-UF$_4$** fueled
- LiF-BeF$_2$ and air-cooled
- Structural material was INOR-8/Hastelloy N
- Operated for 4 years up to 7.4 MW$_{th}$ power
- Operated at a max. nominal fuel temperature of 650°C
- Demonstrated the use of **drain tanks, off-gas system**
- Demonstrated corrosion management by **redox control**
- Demonstrated relatively good behavior of Hastelloy N
- Produced **precious experimental data for validation**





MSRE Parameters and Operational statistics

- **Power**: 7,4 MW (thermal)
  output: 92.8 GWh
  equivalent full-power: 11,555 h
- **Fuel salt**: fluoride, cations: 65% Li-7, 29.1% Be, 5% Zr, 0.9% U
  melting temp: 434 C
  weight: 5,107 kg
  inlet temp: 635 C
  outlet temp: 663 C
  flow rate: 1514 l/min
  fuel pump circulating: 19,405 h
- **Coolant salt**: fluoride,
  cations: 66% Li-7, 34% Be
  weight: 6,940 kg
  coolant pump circulating: 23,566 h
- **Moderator**: nuclear graphite
- **Container**: Hastelloy-N (Ni-based, Mo-16%, Cr-7%, Fe-5%,+Co,W,Si,Mn,Al,Ti,Cu,C)



- **First fuel**: U-235

loading: 150 kgs depleted U and 90 kgs weapon grade U (93% U-235) - ~33% U-235
first critical: 1 June 1965
thermal output: 72,441 MWh
critical hours: 11,515 h
full-power output equivalent: 9,006 h

- **Second fuel**: U-233

loading: 33,26 kg (91,5% U-233), totaly ~36kg with ~84,3% U-233,
critical: 2 October 1968
thermal output: 20,363 MWh
critical hours: 3,910 h
full-power output equivalent: 2,549 h

- **Shutdown**: December 1969

MSRE demonstrated key MSR technologies

Salt chemistry was well-behaved

- *Almost* no corrosion
- UF_4 to UF_3 ratio maintained
- Molten salts are stable under reactor conditions

Hastelloy N performed adequately

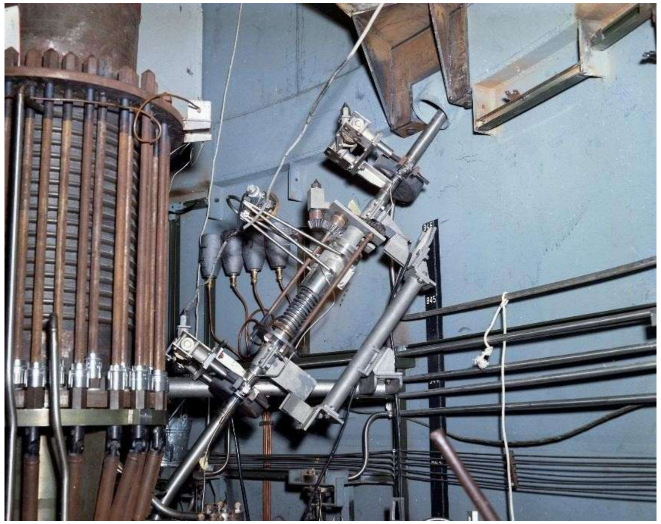
- Little change: ultimate strength, yield strength, creep rate
- Rupture ductility and creep rupture life in specimens reduced

Nuclear performance closely paralleled predictions,

Fission products mainly behaved as anticipated

- Noble gases Xe and Kr efficiently stripped out in off-gas system
- Most other FPs remained in fuel salt

On-line and chemistry adjustment performed well



MSRE Sampler-Enricher Mechanism

MSRE Sample, Enricher Capsules

MSRE Example: Fuel off-gas system

- Salt circuits need to be **inertized** to avoid **air** or **water ingress**
- Gaseous/volatile compounds must be handled

Source : ORNL/TM-0728

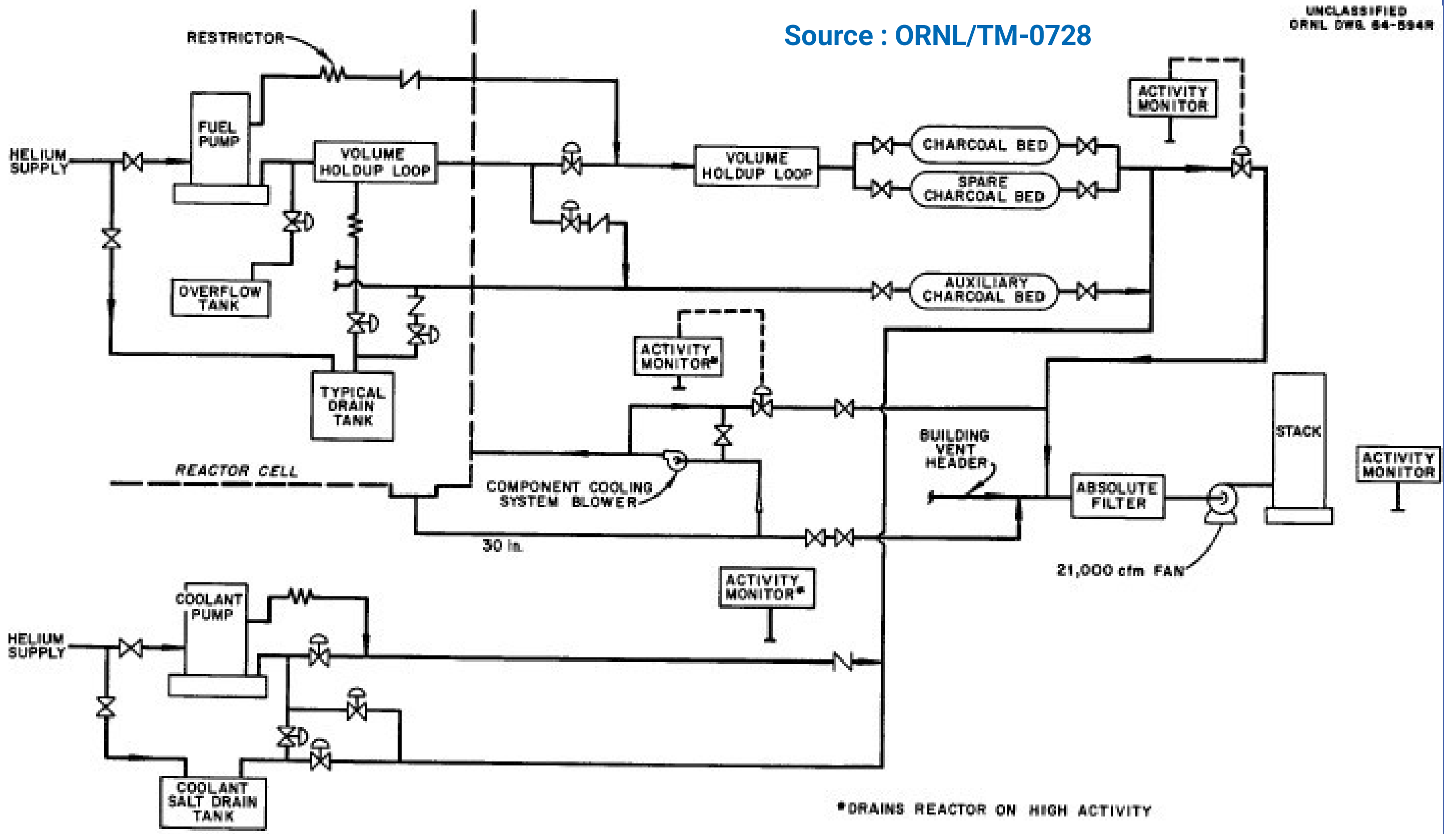
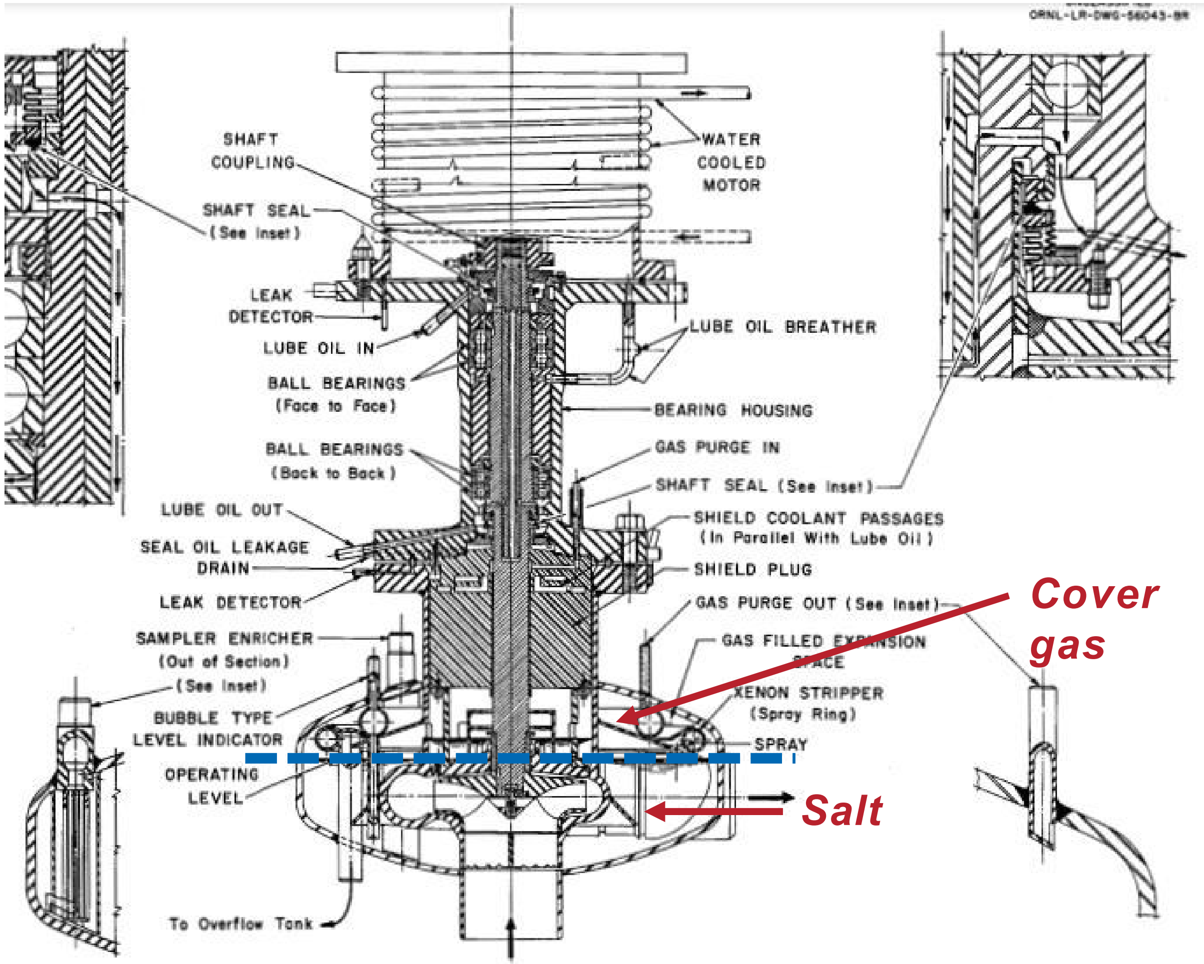


Fig. 12.1. Schematic Diagram of Off-Gas System.

MSRE Example: Fuel off-gas system and pump bowl

- The MSRE pump also plays the role of **expansion tank**
  - Like the PZR in a PWR, accommodates for **changes in volume** of the fuel with temperature
- Provides the fuel circuit with cover gas for inertizing
- Provides free surface for removal of gaseous FP



Source : ORNL/TM-0728

MSRE Example: Fuel salt drain tanks

- Fuel salt drain tank is the vessel for **removal salt in any operational** reasons (e.g., maintenance).
- Such drain tanks can also be used as **safety systems**, as the **fuel can be drained by gravity** from the core into the tank.
- This can be used as a **reactivity control method** and/or alternate **decay heat removal method**.
- Ways of draining include pumping stoppage, frozen valves, siphons, active valves…

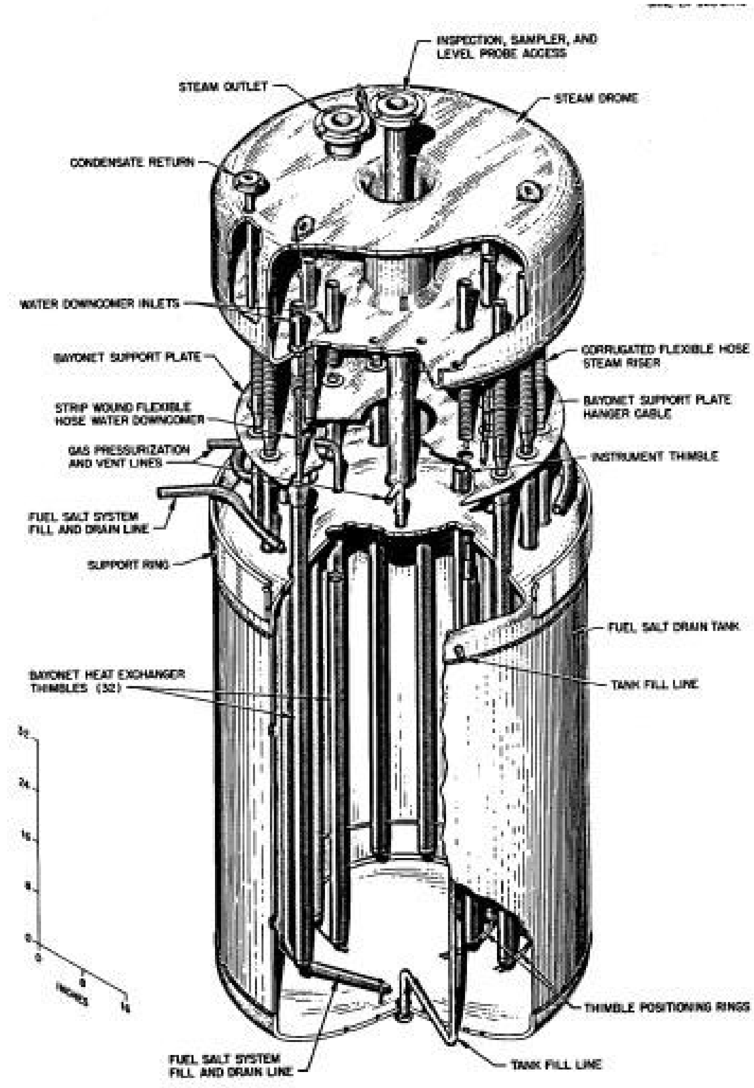


Fig. 2.6. Fuel-Salt Drain Tank.

Source : ORNL/TM-0728

Some important results of MSRE

Distributed source term & decay heat - A unique aspect of MSRs:

Some FP elements were **leaving the salt to the cover gas** or **deposit in the fuel circuit.** This means that both the **source term** and the **decay heat** would be **distributed.** Not only does it need a dedicated safety approach, but also:

- **Specific physic-chemical models for codes** (including severe accidents)
- **Experimental data to validate the models** (which may be difficult to obtain!)

Natural circulation & stability - Impact of internal heat generation

Natural circulation can be **unstable** in certain situations.

Internal heat generation (for example in a **liquid fuel with decay heat**) tends to **increase the size of the unstable domains**

Particular attention should be paid to the physics of such systems, as well as experimental and code validation

Fuel density reactivity feedback - Reactivity effect of fuel dilatation

- The fuel being **liquid**, it dilates **more** with temperature than a solid (approx. **by a factor of 10**);
- Provided the fuel can expand **freely**, fuel expansion **reduces the fuel density in the core**;
- The **reactivity effect** is thus strongly **negative**;
- Moreover, the power is deposited mostly **directly in the fluid** (because it is fuel), and thus the feedback is **fast** (but **not instantaneous)**

Problems and difficulties in MSRE (1)

MSRE *did* encounter issues during operation

MSRE was shut down 225 times, but only 58 were planned

Reactor vessel progressively embrittled due to neutron damage

Drain tank isolation freeze valve cracked during its final cycle due to a field modification

- Stiffening the air-cooling housing prevented pipe flexing
- Xenon, iodine, krypton, and noble metals detected in reactor cell

Small, continuous leak of oil into fuel pump caused foaming

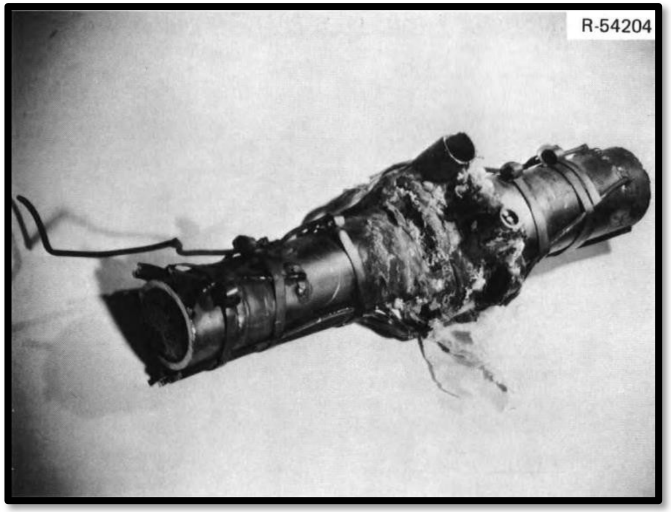
- Periodic plugging of off-gas system
- Fixed with larger filter

Pump-entrained gas caused sporadic (about 10 times/h) increases in reactor power (~5–10%) for a few seconds

Control rod failed scram test due to snagging on thimble

Experimental ‘rod-jogger’ stuck control rod in out position during a pseudo random binary sequence test

Power level ramped up then decreased without intervention



Bottom of cracked freeze valve

Problems and difficulties in MSRE (2)

Some other technological points of MSRE (for further solutions in MSR Projects):

- The reactor's power level: MSRE **was designed to produce 10 MW** of heat, but it was reached only **7.4 MW**.
- Unplanned interruptions were due to various technical problems, including:
  - **"chronic plugging" of the pipes** that led into charcoal beds intended to capture and remove radioactive materials so the reactor could operate;
  - **failures of the blowers** that removed the heat produced in the reactor;
  - **fuel draining** through the so-called "freeze valve safety system" .
- While **Hastelloy-N did not get significantly corroded** (at least during 4 years of operations) it had two significant problems which could lead to the component failing:
  - material had trouble managing stresses. It became brittle, for example.
  - material developed cracks on surfaces exposed to the fuel salt. Both of these
- **Sedimentation (plate-out) of some "noble" FPs** on reactor and circuit surfaces.

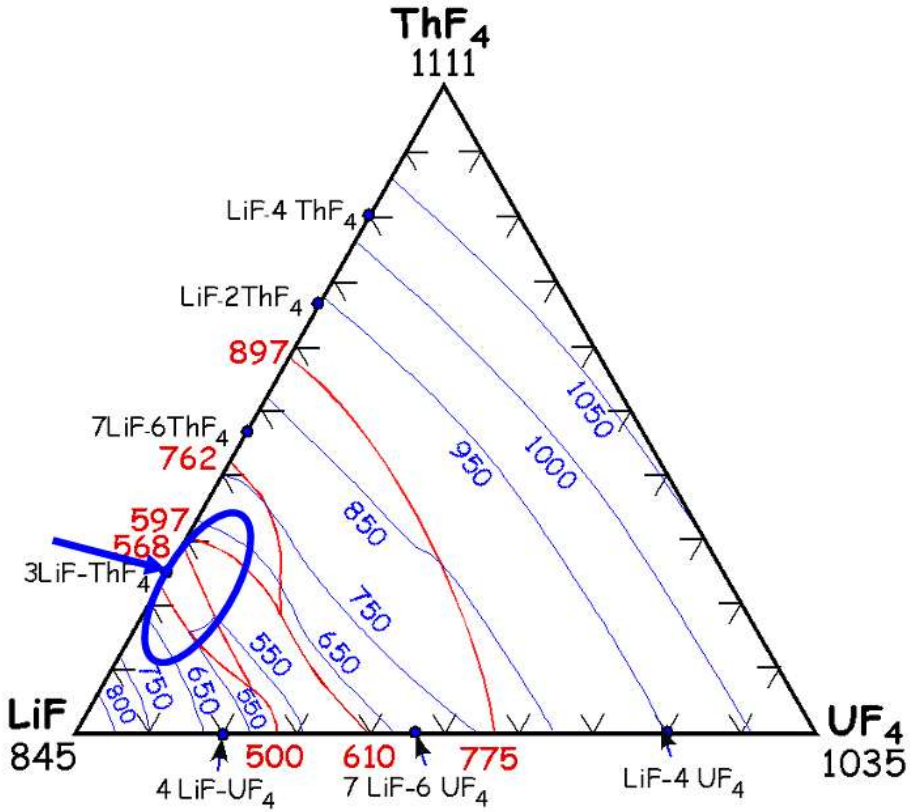
- *Interpretation of FPs behavior were made on basis of 1960s knowlage*
- *Uranium fluorides and FPs behavior during storage period of solid drained salt fuel*

Molten salt physics and chemistry (1)

Selection of salt mixture

- Selection of halide anion (F or Cl) and cation (usually, alkali or alkali-earth metals)
- **Electrochemical compatibility** with the container material
- Mixtures with **low melting point.**
- Phase diagrams are used to determine the **behavior of the mixtures** in terms of melting, boiling, etc.
- Phase diagrams can also **give solubility limits as function of the temperature.**
- Cooling **above the solubility limit** may lead to **precipitation of species**

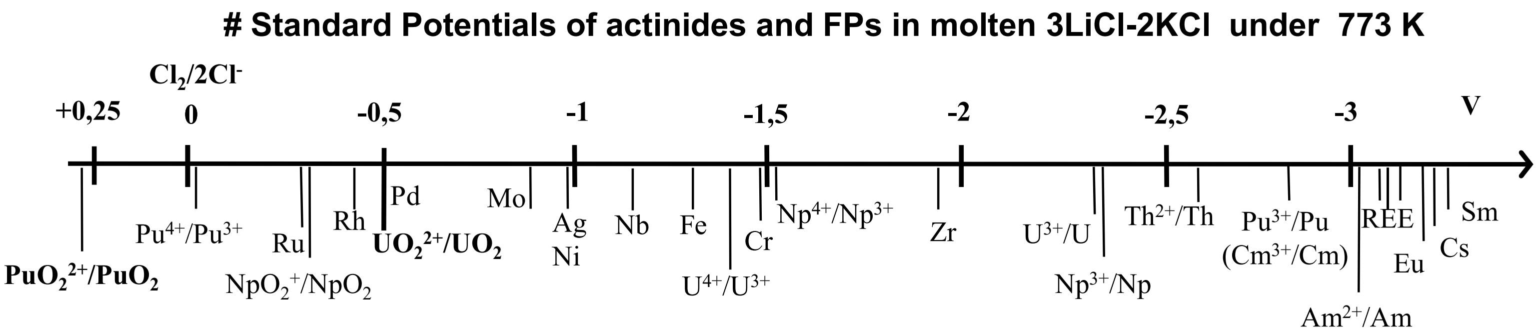
Phase diagrams



Molten salt physics and chemistry (2)

Redox potential control – very important

- **Redox potential** is a measure of a substance's tendency to gain or lose electrons, expressed in volts, indicating its ability to act as an oxidizing or reducing agent.
- Electrical potential in the cell formed by the salt and the container material follows the **Nernst equation**

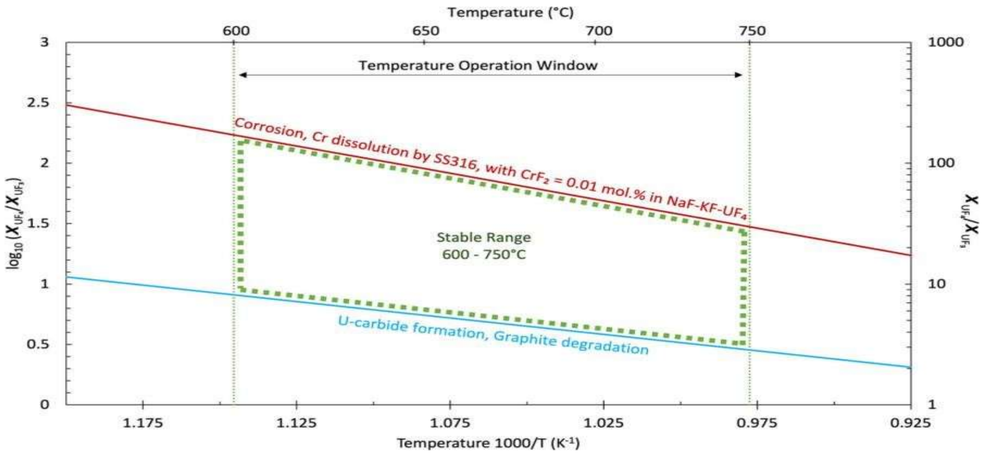


- Depending on t**he redox potential** of the salt, elements will have different oxidation states
- Depending on the t**emperature**, **pressure**, and oxidation **state**, an element will behave **differently**
- This means that there is an **additional variable to take into account** in the safety assessment

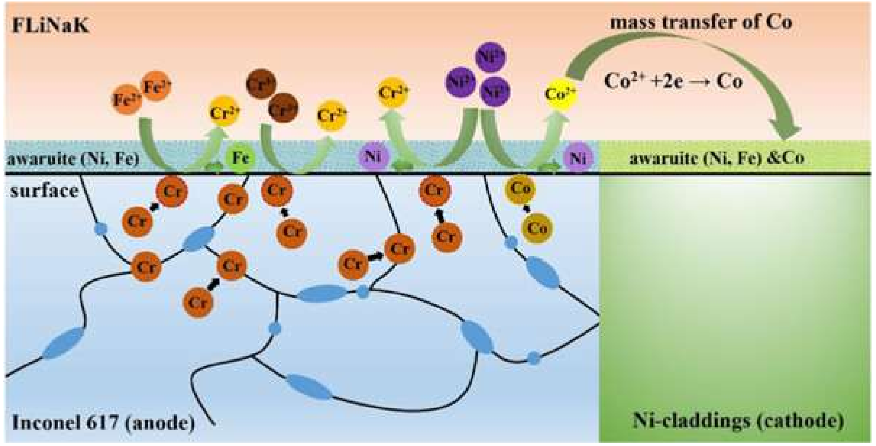
Molten salt physics and chemistry (3)

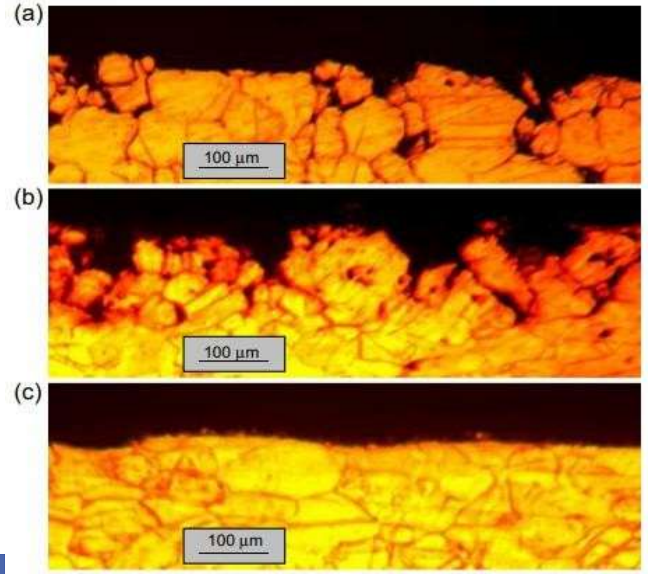
Redox potential control and corrosion

- Influences **corrosion rates** and **what types of materials** will be stable in or in contact with the salt
- In particular, it influences the **behavior of salt components**, in particular fission products



Source (1) : doi.org/10.1016/j.electacta.2024.144647
Source (2) : doi.org/10.1016/B978-0-08-100906-2.00005-7





No load
U(4)/U(3) = 500

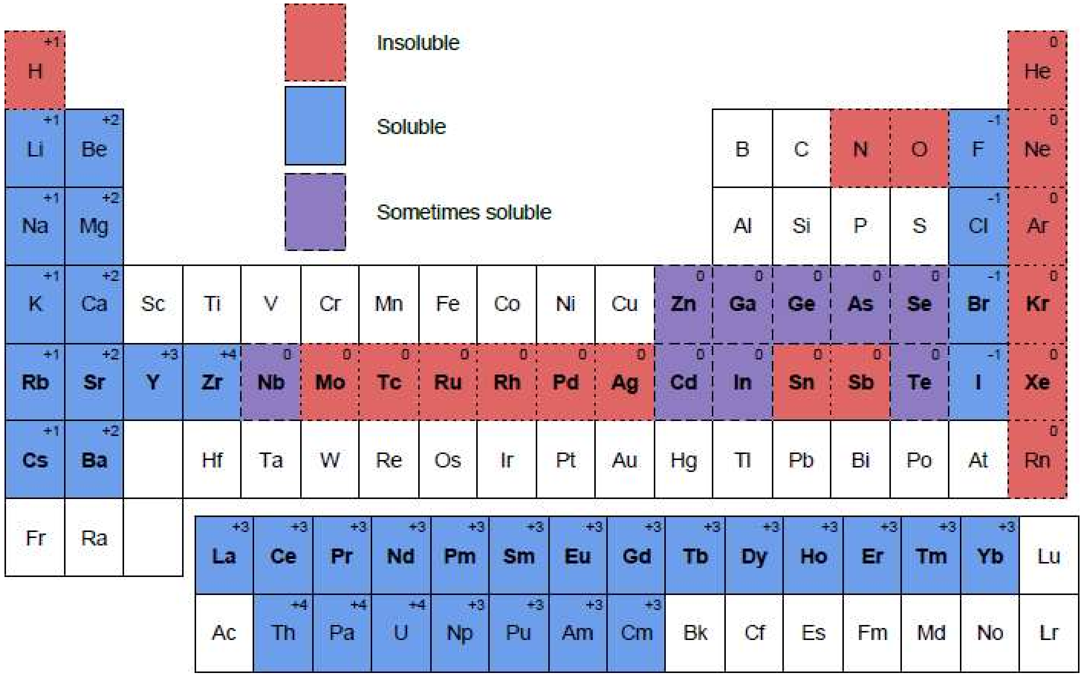
Loaded
U(4)/U(3) = 500

Loaded
U(4)/U(3) = 100

Actinides and Fission products behavior in molten salt

Depends on the FP element, redox potential, temperature…

- Some FPs are **gaseous** and **insoluble** in the salt: they will leave the salt naturally (e.g., Xe, Kr, etc.);
- Some FPs form **volatile halides**: part of them evaporate in the cover gas (e.g., ZrCl4), part of them in the salt.
- Some FPs are **solid** and **insoluble** (electrochemically noble): they can likely deposit (plate out) in the fuel circuit (e.g., Mo, Nb, Ru etc.);



- Some FPs are **liquid** and **insoluble** (electrochemically noble) or form **unstable halides**: they can transport with the salt as droplets or deposit in the circuit (e.g., Sb);
- Other FPs form stable halides (cations and anions) and **remain in the salt** (e.g., Cs, Ba, lanthanides, I, Br).

FPs solubility changes along decay chain

A few elements are very sensitive to redox changes:

Nb behavior changed during MSRE operation after addition of Be°

Transitional (*soluble* → *gas* → *soluble*) decay example:

24 sec. half-life *4min. half-life* *6 % cumulative ^{235}U fission yield*

$^{137}I \rightarrow {}^{137}Xe \rightarrow {}^{137}Cs$

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	*Zn*	*Ga*	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	*Cd*	*In*	Sn	Sb	Te	I	Xe
Cs	Ba	La-Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	*Hg*	*Tl*	Pb	Bi	Po	At	Rn
Fr	Ra	Ac-Lr															

soluble “insoluble” sometimes soluble

Fission continuously alters redox conditions

When U or Pu atom fissions, the available electrons will rearrange on each fission product to satisfy its oxidation state requirements and produce either net oxidizing or reducing conditions in the melt :

- For ^{235}U (as UF_4) **four** F ions are released.

  FPs require <4 and thus there will be an **excess** of F ions with **net oxidizing conditions**
- For ^{239}Pu (as PuF_3) **three** F ions are released.

  FPs require >3 and thus there will be a F ion **deficit** with **net reducing conditions**

Salt	Fission Product	Oxidation State (Z)	Yield (Y) [atoms]	F atoms reacted (Y*Z)
Fluoride Salt (UF_4)	Br, I	-1	1.5	-1.5
	Kr, Xe	0	60.6	0
	Rb, Cs	1	0.4	0.4
	Sr, Ba	2	7.2	14.4
	Lanthanides, Y	3	53.8	161.4
	Zr	4	31.8	127.2
	Nb	0	1.4	0
	Mo	0	20.1	0
	Tc	0	5.9	0
	Ru	0	12.6	0
	Total F atoms reacted out of 400 available			301.9

MSRE periodically added metallic Be (strong reducing agent) to maintain UF_4/UF_3 ratio

- Fuel salt redox changes with decay of retained fission products and escape of gaseous fission products

Use of a circulating redox buffer (U^{4+}/U^{3+}) provides means to maintain redox condition

Sources: Baes (fluoride salts), Harder (chloride salts)

MSRE demonstrated corrosion control via redox

Source: ORNL/TM-4174

- Surveillance specimen of graphite and Hastelloy-N were introduced in MSRE
- As can be seen on the picture, numbers scratched on the surface are **still visible**
- And this after 15300 hours in salt at 650°C !
- **Corrosion control by redox control** is demonstrated, but it needs to be implemented **practically** for a reactor

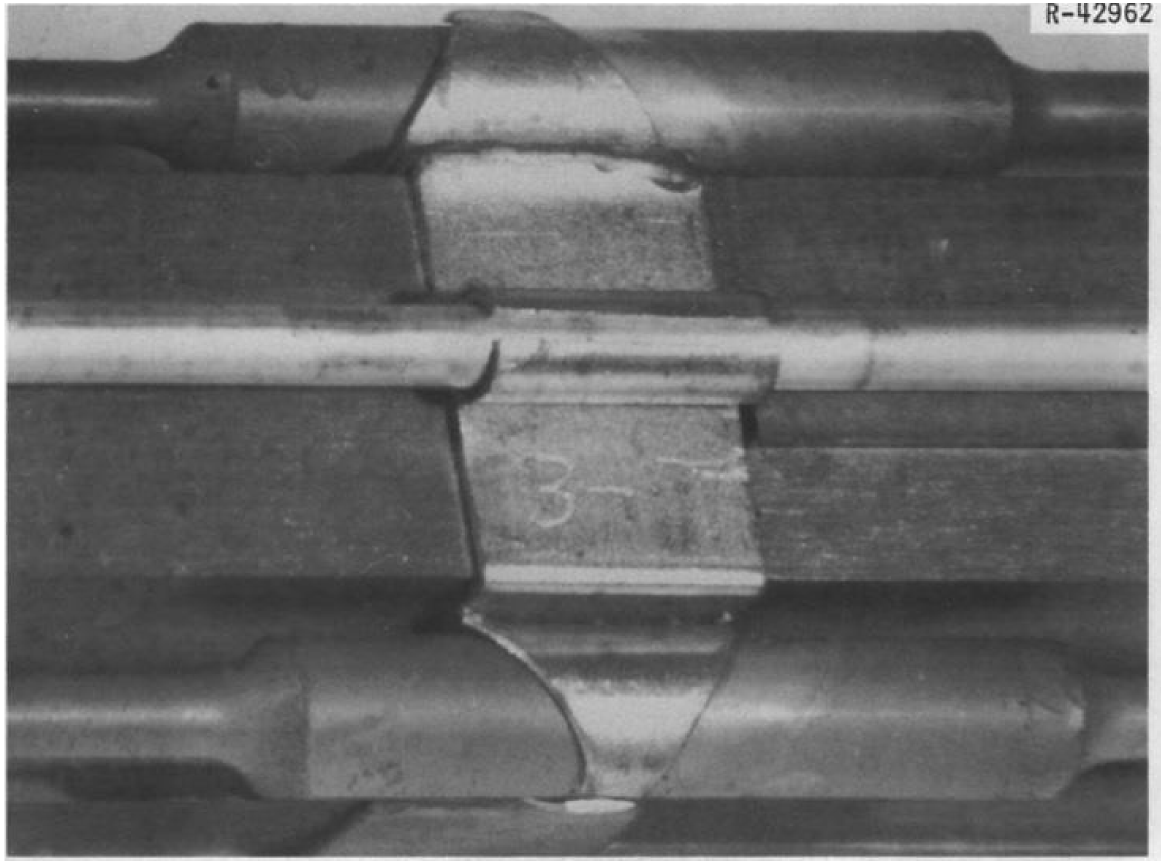


Fig. 1. Graphite and Hastelloy N Surveillance Assembly Removed from the Core of the MSRE After 72,400 Mwhr of Operation. Exposed to flowing salt for 15,300 hr at 650°C.

Results of MSRE – from the past to the present and the future

Alvin Weinberg's message for future

Director ORNL **Alvin Weinberg's** explanation revolved around the historical domination of nuclear power by physicists. In a letter to **Glen Seaborg**, he wrote, "***our problem is not that our idea is a poor one — rather it is different from the main line, and has too chemical a flavor to be fully appreciated by non-chemists***."

MSRE demonstrated the feasibility of Molten Salt Reactors implementation!

BUT....

During last 60 years requarements reletad nuclear reactor technologies and performances changed completetly:

- Strenthening of Nuclear and Radiation safety standards
- Envaironmental protection approches and standards

„new regulatary world"

- Non-prolifaration and safeguard control
- Nuclear security

„new global security regim"

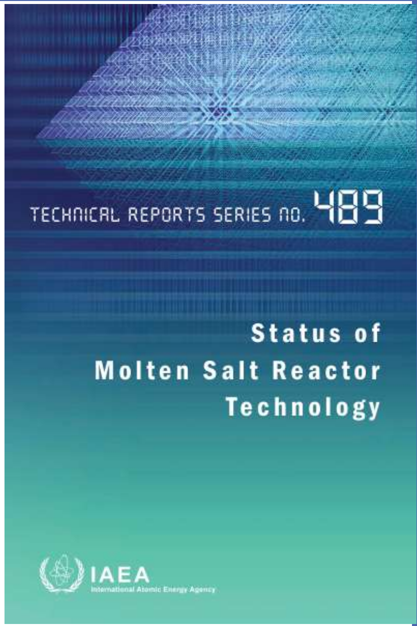
Molten Salt (and Ionic liquids) physical chemistry deeply studied in 1970-2000s – new understanding

Understanding of Advantages and Technical Challenges of MSR Technology based on MSRE results

- Safety advantages
  - Near atmospheric operating pressure.
  - Radioisotope retention.
  - Chemically inert and not flammable.
  - Removal of gaseous fission products.
  - No irradiation damage or mechanical failure of fuel.
  - Strong negative reactivity temperature feedback.
  - Molten salt as a heat sink.
  - High boiling point.
  - Excellent neutron economy.
  - Flexible fuel cycle.

- Economic advantages
  - High temperature.
  - Effective load following.
  - High resource utilization.
  - Fuel qualification.

- TECHNICAL CHALLENGES
  - associated with reactor physics. associated with salt chemistry and materials science.
  - associated with engineering design of all main and auxilary systems



Technology Gaps of MSR
(based on MSRE results)

A. Power Cycles
- Efficiency.
- Freeze protection.
- Tritium control.
- Chemical reactions.

B. Fuel Inventory
- Fuel salt inventory.
- Fuel salt processing.
- Heat exchanger size.
- Tritium control.

C. Noble Metal Plate-Out
- Better understanding of the physical processes.
- New materials.
- Improved separation methods.

D. Fuel Storage

E. High-Level Waste (HLW) Form
- Conversion to nonfluoride waste forms.
- Fluoride HLW form.

F. Peak Reactor Temperature

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Molten-Salt-Reactor Technology Gaps

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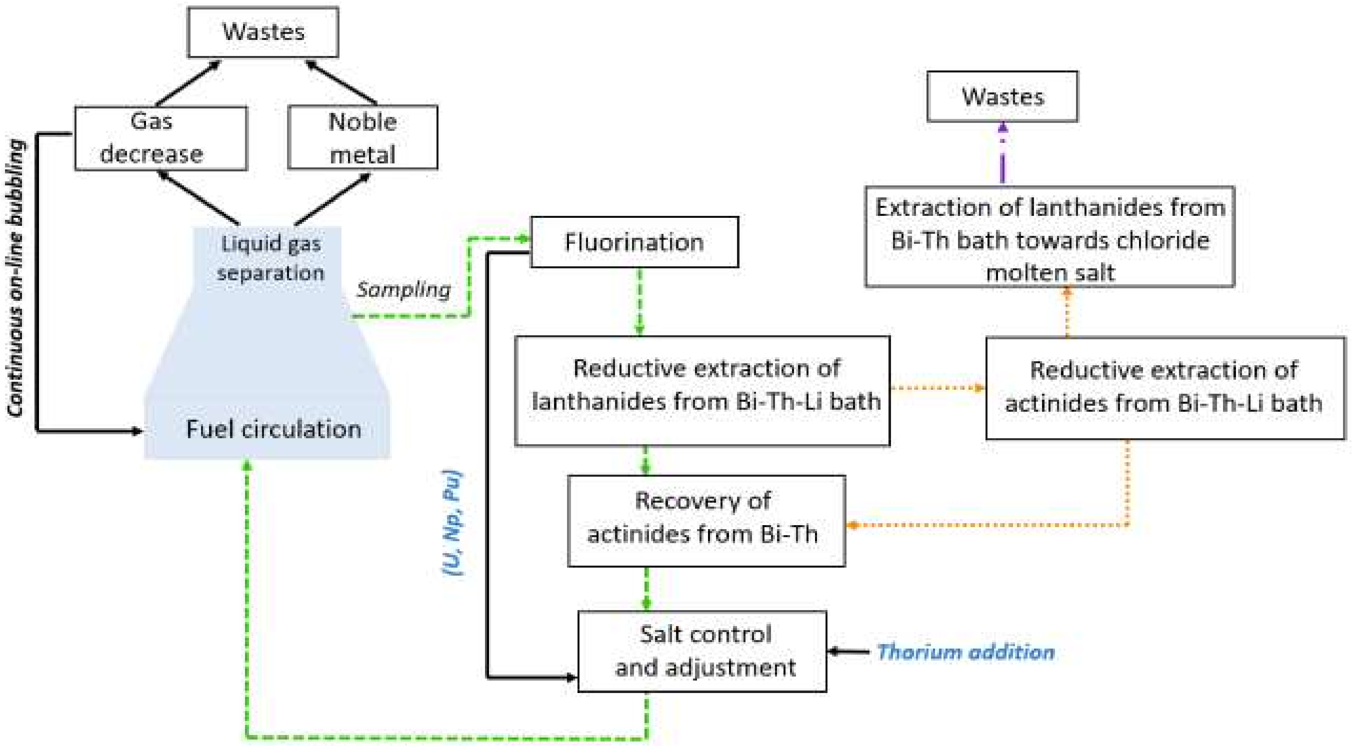
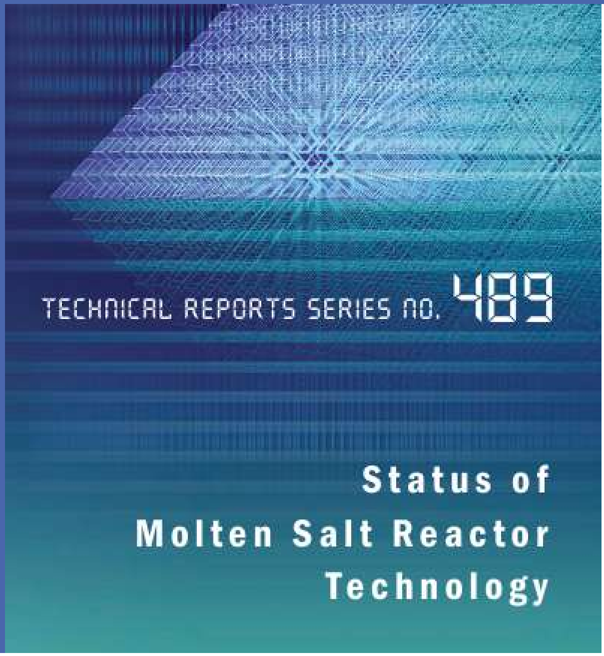
MSR fuel treatment: Process flow and primary waste stream for recycling of MSR fuel



- Noble gases and Noble Metals fission product can be removed online by special traps.
- For complete reprocessing of salt: some pyro-process could be applied as molten salt electrolysis, precipitation or fluorination methods.
- No complete understanding and models for behavior of fission product's atoms in ionic liquids after actinides fission.
- Waste form should be specially selected

INPRO/WIRAF Study



CURRENT RESEARCH AND DEVELOPMENT

RECENT PROGRESS IN MOLTEN-SALT REACTOR DEVELOPMENT *

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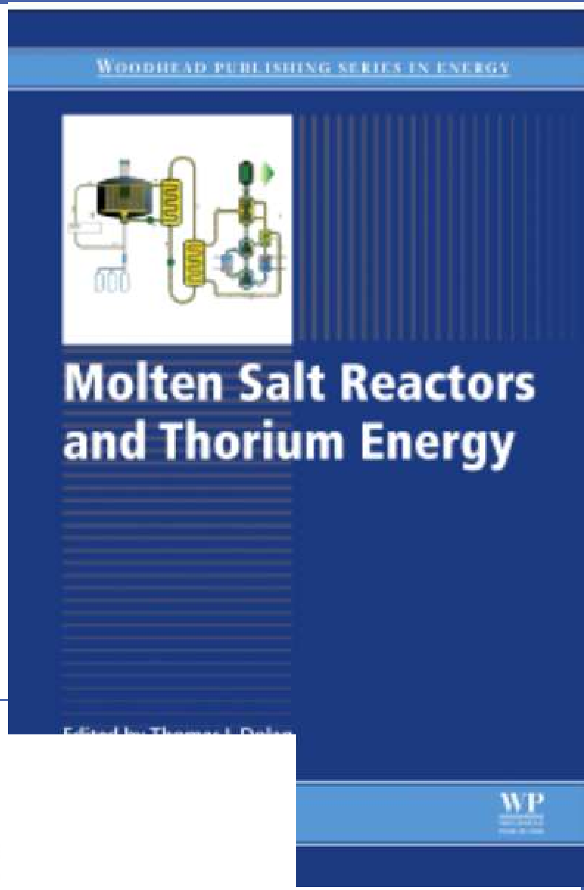
ABSTRACT. This article reviews progress made in the development of molten-salt breeder reactors since an overall review of molten-salt reactor technology was written about two years ago. Emphasis is placed on three areas in which advancements during this period have been most significant. These are: (1) the final period of operation of the Molten-Salt Reactor Experiment, in which the reactor operated on ^{233}U but additions of plutonium were made to obtain experience with it as a MSR fuel; (2) the modification of Hastelloy N, the preferred container and structural material for MSRs, to give it resistance to radiation damage at the full range of conditions expected in a power reactor; and (3) the development of an improved process for the rapid removal of rare-earth fission products from the fuel salt.



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MOLTEN-SALT REACTORS—HISTORY, STATUS, AND POTENTIAL

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Molten-salt breeder reactors (MSBR's) are being developed by the Oak Ridge National Laboratory for generating low-cost power while extending the nation's resources of fissionable fuel. The fluid fuel in these reactors, consisting of UF_4 and ThF_4 dissolved in fluorides of beryllium and

fuel appeared to offer several advantages, so experiments to establish the feasibility of molten-salt fuels were begun in 1947 on "the initiative of V.P. Calkins, Kermit Anderson, and E.S. Bettis. At the enthusiastic urging of Bettis and on the recommendation of W.R. Grimes, R.C. Briant adopted molten fluoride salts in 1950 as the main

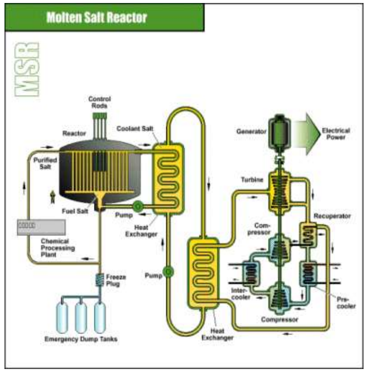
MSR and Fuel: R&D needs

Tasks for future

- Consideration of different severe accidental scenarios -non criticality, but related to FPs release.
- Development of the model of FPs transformation and behavior in Molten salt
- Study of corrosion mechanisms including welding's as potentially dangerous surfaces for corrosion or development of protection

Potentially useful R&D

- Study of molten salt properties and FPs behavior
- Performance of integral and separate effect tests to validate safety performance
- Improved neutronic and thermal-hydraulic models and tools
- Study of materials aspects associated with MSR
- Demonstration of tritium management technologies
- Salt redox control technologies for avoiding of corrosion
- Demonstration of surveillance and maintenance technologies for high irradiation conditions, such as MSR containments



Conclusions

Experimental projects on early MSR (MSRE mailnly) demonstatted:

- The fundamental possibility of technology
- The choice of materials
- The ability to manage of MSR and salt fuel

MSRE is and will be the base for further research and development.

Experimental data from earlz MSR were obtained, processed and interpreted by “old” outdated models and concepts.

Experimental studies are required on real MSR test systems with irradiation and circulation of fuel

The unconventional concept of a nuclear reactor that is at the same time a chemical reactor must be implemented – a new understanding of molten salt fuel and all aspects of safety and operability.

Thank you!

The presentation contains illustrations from publicly available publications.
Special thanks to Artemiy Konstantinov (IAEA/NSNS) for help in preparation of slides!

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