Module 3: Overview of Fuel and Coolant Salt Chemistry and Thermal Hydraulics

Presentation on Molten Salt Reactor Technology by:
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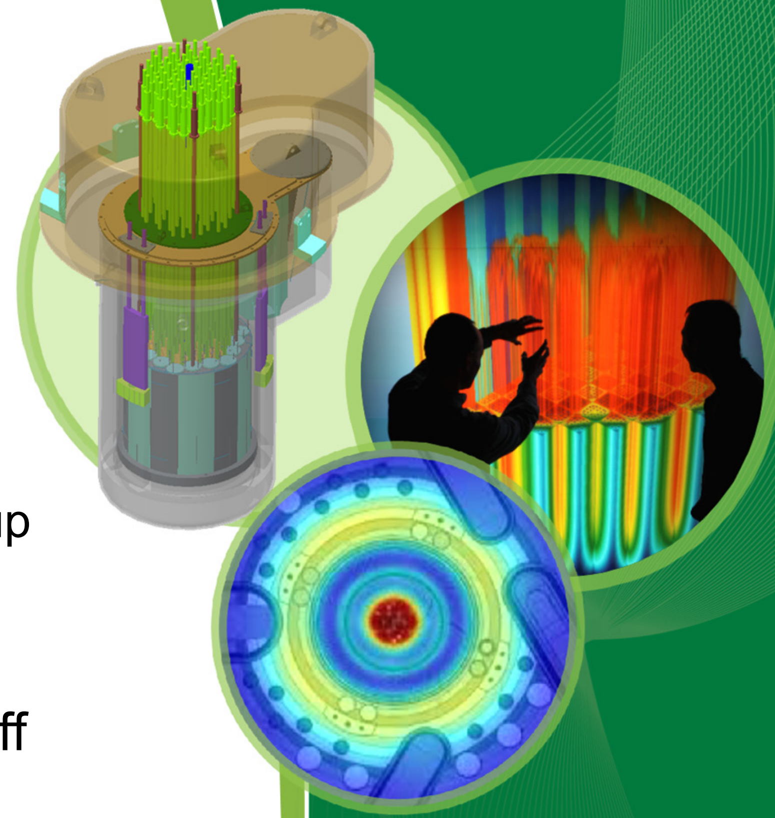

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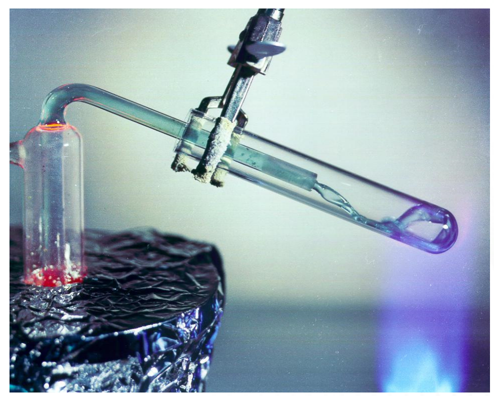
What are “Molten Salts”?

- Salts are ionic compounds formed from a combination of electronegative and electropositive elements
  - At elevated temperatures salts liquefy and are termed “molten salts”
- Halide salts are ionic compounds formed from the combination of a halogen (electronegative) and another electropositive element – commonly, but not exclusively, alkali metals or alkaline earths
  - Examples: LiF, BeF_2, $MgCl_2$, NaCl (aka table salt), ZrF_4, RbF, UF_4, UCl_3

Halogens	Alkali Metals	Alkaline Earths
9 F Fluorine 18.998…	3 Li Lithium 6.94	4 Be Beryllium 9.0121…
17 Cl Chlorine 35.45	11 Na Sodium 22.989…	12 Mg Magnesium 24.305
35 Br Bromine 79.904	19 K Potassium 39.0983	20 Ca Calcium 40.078
53 I Iodine 126.90…	37 Rb Rubidium 85.4678	38 Sr Strontium 87.62
85 At Astatine (210)	55 Cs Caesium 132.90…	56 Ba Barium 137.327
117 Ts Tennessine (294)	87 Fr Francium (223)	88 Ra Radium (226)



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

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Molten Halide Salts Have Attractive Heat Transfer Properties

Coolant (Reactor Concept)	High Working Temperature[a]	High Volumetric Heat Capacity[b]	Low Primary Pressure[c]	Low Reactivity with Air & Water[d]	Coolant & Materials Cost
Water (PWR)	☹	☺	☹	☺	☺
Sodium (SFR)	😐	😐	☺	☹	😐
Helium (GCR)	☺	☹	☹	☺	😐
Salt (FHR/MSR)	☺	☺	☺	☺	☹

[a] High system working temperature desirable for high efficiency power conversion and process heat applications

[b] High coolant volumetric heat capacity enables ~constant temperature heat addition / removal ($\eta_C = 1 - T_C/T_H$ ~ Carnot cycles), compact system architectures, and reduces pumping power requirements

[c] Low primary system pressure reduces cost of primary vessel and piping and reduces energetics of pipe break accidents

[d] Low reactivity with air and water reduces energetics of pipe break accidents

Molten Salts Are Attractive Coolants for Very High Temperatures

<u>*Compared to 20°C water*</u>

Fluorides:

- ~ 2X density
- ~ 1/2X heat capacity
- ~ 1–5X viscosity
- ~ 2X thermal conductivity
- ~ 1X coefficient of expansion as a liquid
- Very low vapor pressure

Chlorides:

- ~ 1 1/2X density
- ~ 1/4X heat capacity
- ~ 1 1/2X viscosity
- ~ 1X thermal conductivity
- ~ 1 1/2X coefficient of expansion as a liquid
- Very low vapor pressure

Characteristics of *Fuel Salts* and *Coolant Salts* Are Available from Review Articles

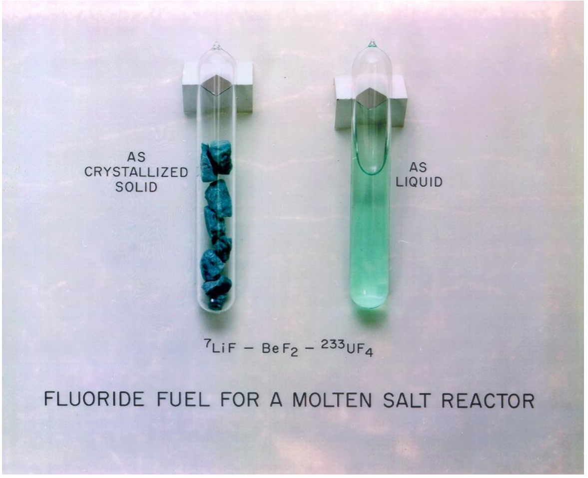
- A fuel salt is a molten salt that contains fissile material
  - C. F. Baes, Jr., "The Chemistry and Thermodynamics of Molten Salt Reactor Fuels," *Journal of Nuclear Materials*, 51 (1974) 149-162
  - W. R. Grimes, "Molten Salt Reactor Chemistry," *Nuclear Applications and Technology*, 8(2) (1970) 137–155
  - B. R. Harder, G. Long, and W. P. Stanaway, "Compatibility and Processing Problems in the Use of Molten Uranium-Alkali Chloride Mixtures as Reactor Fuels," *Nuclear Metallurgy, Metallurgical Society of the American Institute of Mining, Metallurgical and Petroleum Engineers*, 15 (1969) 405-32
- Coolant salts are molten salts with advantageous heat transfer properties
  - D. F. Williams, *Assessment of Candidate Molten Salt Coolants for the NGNP/NHI Heat-Transfer Loop*, ORNL/TM-2006/69

Composition of Fuel Salts Are Tailored to Performance Objectives

- Fuel salts consist of a mixture of
  - Fissile material
  - Fertile material (if used)
  - Solvent (diluent)
    - Lowers melting point
    - Decreases power density
    - Decreases viscosity
  - Fissile oxidation prevention material
    - Preferentially oxidizes to avoid creation of fissile oxide particles due to contamination

$$ZrF_4 + UO_2 \Leftrightarrow ZrO_2 + UF_4$$

  - Fission products (upon use)



Fuel Salts Must Integrate Reactor Physics, Heat Transfer, and Material Compatibility

- Reactor physics requirements
  - Low neutron absorption
    - Thermal neutron absorption is of lower importance for fast spectrum reactors
  - Radiolytic stability under in-core conditions
  - Dissolve fissile materials
- Both chloride and fluoride salts are industrially used as heat transfer fluids
  - High heat capacity, high boiling point, low thermal conductivity fluids
  - Melting point must be below ~525°C
  - Relatively insensitive to fission products
- Both fluoride and chloride salts, under mildly reducing conditions, are reasonably compatible with high temperature structural alloys and graphite

Elements or Isotopes Which may be Tolerable in High-Temperature Reactor Fuels

Material	Absorption Cross Section (barns at 2200 m/sec)
Nitrogen-15	0.000024
Oxygen	0.0002
Deuterium	0.00057
Carbon	0.0033
Fluorine	0.009
Beryllium	0.010
Bismuth	0.032
Lithium-7	0.033
Boron-11	0.05
Magnesium	0.063
Silicon	0.13
Lead	0.17
Zirconium	0.18
Phosphorus	0.21
Aluminum	0.23
Hydrogen	0.33
Calcium	0.43
Sulfur	0.49
Sodium	0.53
Chlorine-37	0.56
Tin	0.6
Cerium	0.7
Rubidium	0.7

Source: Grimes, "Molten Salt Chemistry," *Nuclear Applications and Technology* 8(2) (1970) 137-155.

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Fuel Salts Have Multiple Subclasses

- Thermal spectrum reprocessing optimized fluoride salts
  - FLiBe ($2^7LiF\text{-}BeF_2$) solvent provides optimal neutronic performance
    - Lithium to beryllium ratio selected to minimize melt temperature with acceptable viscosity
    - High tritium production – need isotopically separated lithium
  - $NaF\text{-}ZrF_4$ solvent does not require isotopic separation
    - Much lower tritium production
    - Higher vapor pressure
  - ~1% fissile loading
  - Fertile loadings vary but are typically much higher (~20%)
- Fast spectrum and thermal spectrum, once-through fuel cycle optimized fluoride salts
  - Much higher fissile loading (actinide-rich eutectics)
  - Adequate fissile material content is a significant design challenge
- Chloride salts
  - Enables harder neutron spectrum and enhanced breeding
  - Isotopically separated chlorine preferable - ^{35}Cl from ^{37}Cl
    - ^{35}Cl has a moderate capture cross-section (n,γ) E < 0.1 MeV < E (n,p)

European Fast Spectrum MSR starting fuel composition $LiF\text{-}ThF_4\text{-}UF_4\text{-}(TRU)F_3$ with 77.7-6.7-12.3-3.3 mol%

Chlorine natural isotopic composition
^{37}Cl = 24.23%
^{35}Cl = 75.77%

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Fluoride Fuel Salts Have Substantially More Experimental Data Than Chloride Fuel Salts

- Fluoride salts
  - **Two operating molten salt reactors**
  - Multiple in-pile loops
  - Many capsule tests
  - Fast-spectrum fluoride salts have much less experience
- Chloride salts – laboratory measurements of physical properties
  - No in-core testing of fuel salts
  - Use in pyroprocessing

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Thermal Spectrum Fuel Salt Behaves Similarly to Solvent Salt

- MSRE nominal fuel mixture was

  65 LiF, 29.1 BeF_2, 5 ZrF_4, 0.9 UF_4 (mol %)

- Uranium enriched to 33%

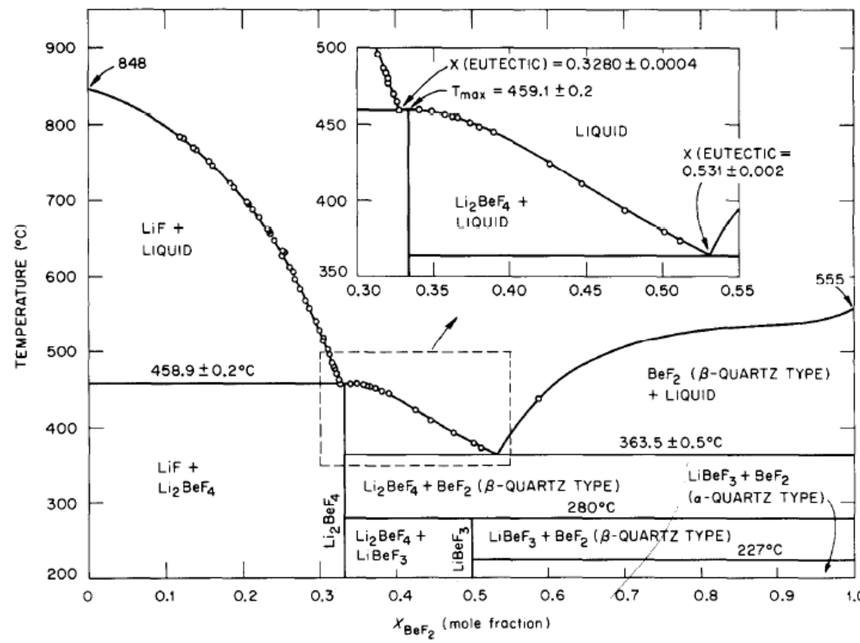
- Uranium trifluoride disproportionates in most molten fluoride solutions

$$4UF_3 \Leftrightarrow 3UF_4 + U^0$$

  – Large UF_4/UF_3 ratio prevents disproportionation

- Isotopically pure ^7Li - nominally 99.993% at MSRE

  – Means to limit tritium production due to large ^6Li cross-section



LiF-BeF_2 Phase Diagram

Source: Benes and Konings, “Thermodynamic properties and phase diagrams of fluoride salts for nuclear applications,” *Journal of Fluorine Chemistry*, 130, 2009.

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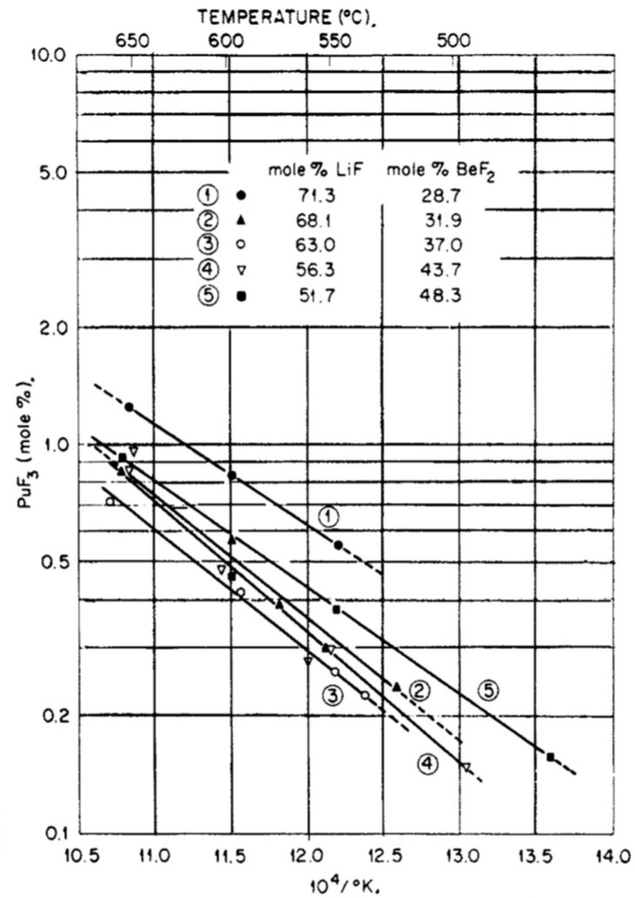
Fluoride Fuel Salts Have Limited Solubility for Actinide Trifluorides

- Fast spectrum systems operate near solubility limits
  - Lanthanide trifluorides compete with actinide trifluorides
    - CeF_3 substantially displaces PuF_3
  - Log of actinide trifluoride solubility is roughly linear versus inverse temperature
- Monovalent solvent fluorides dissolve much higher levels of actinide trifluorides
  - Joint solubility of PuF_3+UF_3 is much less than individual components up to 600°C
  - Solubility has strong temperature dependence
    - Plate out during transients possible
  - Polyvalent fluorides (e.g., ThF_4, UF_4, or BeF_2) substantially reduce solubility



Solubility of PuF_3 in FLiBe

Source: C. J. Barton, "Solubility of Plutonium Trifluoride in Fused-Alkali Fluoride-Beryllium Fluoride Mixtures" *J. Phys. Chem.* , Vol. 64, 1960

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Fuel Salt Properties Will Be Impacted by Fission Products

- Fission products may be gaseous, solid, or dissolved
  - Alkaline and alkaline earth fission products (e.g., Cs and Sr) form stable fluorides (or chlorides)
  - Semi-noble fission products plate out on metal surfaces
    - Potential heat load issue following rapid draining
  - Noble fission products form suspended clusters that may plate out
- May elect to actively strip gaseous fission products
  - Lowers the in-core accident source term
  - Requires cooling fission product traps
  - Bubble formation and collapse results in reactivity burps
- Fluoride salts have been extensively examined
  - Reactors, in-pile loops, capsules
  - Some uncertainty remains - especially about impact of long-term build up of fission products
- Chloride fuel salts almost entirely untested in core environments
  - Potential for development of undesirable compounds and phases

> "I am pleased, without benefit of rack and thumbscrew, to recant. More realistic calculations based on the single-region 'reference design' MSBR heat exchangers indicate that peak afterheat temperatures, while still uncomfortably high, will be much lower than originally anticipated."
>
> J. R. Tallackson, ORNL-TM-3145

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Fission Product 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 — *4-min. half-life* — *6 % cumulative ^{235}U fission yield*

^{137}I → ^{137}Xe → ^{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

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Cover Gas Handling System Is a Key Element of Any MSR

- Distribution of fission products is a central safety issue
  - Reduction of fission products in the core limits potential fuel accident source term
  - Fission products away from core change decay cooling requirements and radionuclide containment requirements
- Cover gas will inevitably contain some fission products
  - Aggressive sparging may result in up to 40% of fission products in cover gas (nearly all of the fission products with gaseous precursors)
  - Results in substantial heat load in short term fission product trap
  - Longer term fission product traps contain much lower levels of activity
- Transition from fission product barrier function to waste handling system along carbon beds is conceptually significant
  - ^{85}Kr emerging from final stage could be vented
- Some fuel salt fissile components have significant vapor pressures
  - UCl_4 boils at 791°C
- Some solvents vaporize incongruently
  - ZrF_4 sublimes resulting in snow-like deposits in exhaust piping

NRG (Petten) Recently Began Irradiation Tests of Fuel Salt Capsules

- SALIENT program is trilateral collaboration between NRG, JRC, and TUD
- Fluoride salts initially
  - Chlorides later stage
- Goals
  - Handling experience
  - Salt–graphite interaction
  - Fission product stability / redistribution
  - Metal particle size distribution
- Longer term
  - Waste route for spent molten salt fuel
  - In-pile molten salt loop for the HFR Petten

Cartoon of potential Petten MSR loop

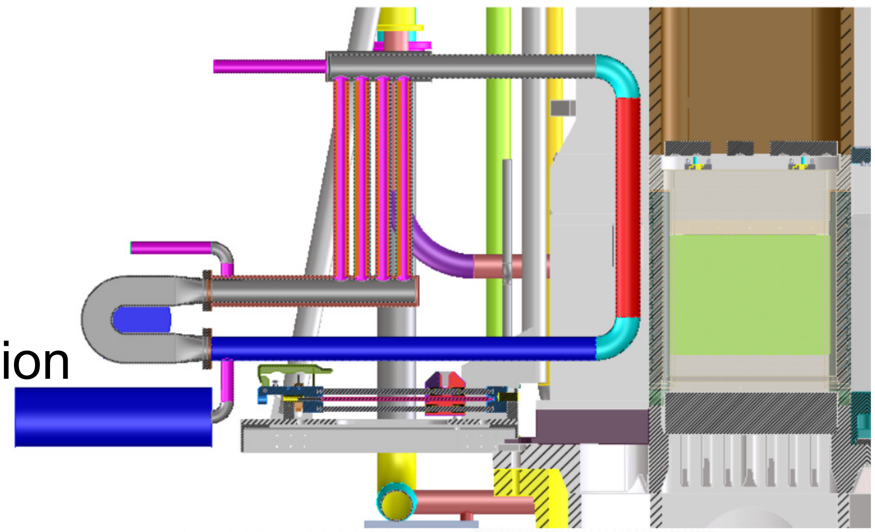


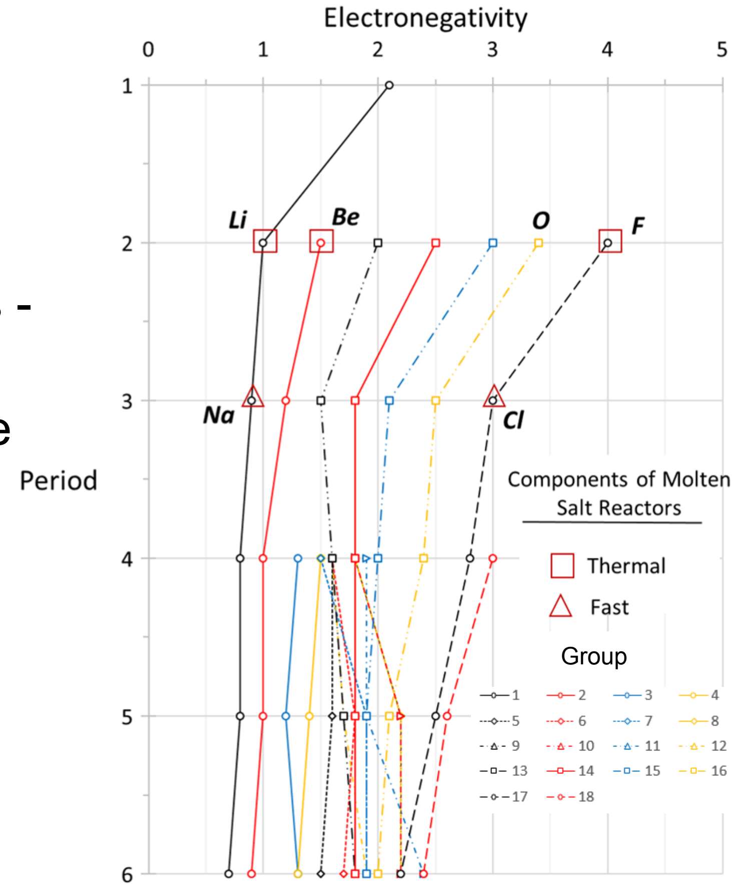
Image provided by NRG; used with permission.

Isotope Separation Is a Significant Issue for Both Fluoride and Chloride MSRs

- Lithium enables optimal reactor physics
  - Lithium-6 is a large cross-section thermal neutron absorber that yields tritium
  - Lithium isotope separation is also necessary for fusion and PWR chemistry control
  - Mercury amalgam-based lithium isotope separation was performed at industrial scale in the 1950s for defense purposes
- Chlorine
  - Absorption reactions in ^{35}Cl both produces ^{36}Cl (long-lived radionuclide) and results in a reactivity penalty
  - Lack of chlorine isotope separation technology was a key element in US decision in 1956 to pursue thermal breeder MSR

Removing Oxygen Is a Key Technology Requirement for Both Fluorides and Chlorides

- Salts containing excess oxygen are much more corrosive
- Hydrofluorination for fluoride salts
  - HF is highly corrosive - performed offline
  - Also removes other electronegative impurities - sulfur and chlorine
  - Ammonium hydrofluoride - NH_4HF_2 alternative
- Carbochlorination for chloride salts – phosgene ($COCl_2$) or carbon tetrachloride used as reactant
  - $MO_2 + CCl_4 \rightarrow MCl_4 + CO_2$
- Oxygen can also be removed from some chloride melts by precipitation as aluminum oxide
  - $AlCl_3 + UO_2 \rightarrow AlO_2 + UCl_3$



Source: Taube EIR-332; p.156

Tritium is Significant Issue For Lithium-Bearing Salts

- Tritium is produced by neutron reactions with lithium, beryllium, and fluorine as well as being a ternary fission product
  - Tritium production levels are similar to HWRs
- Tritium chemical state in salt is determined by redox conditions
  - TF (oxidizing) or T^+ (reducing)
- Above 300°C tritium readily diffuses through structural alloys
  - Heat exchangers represent largest surface area for diffusion
- Escape through power cycle is potential route for radionuclide release into environment

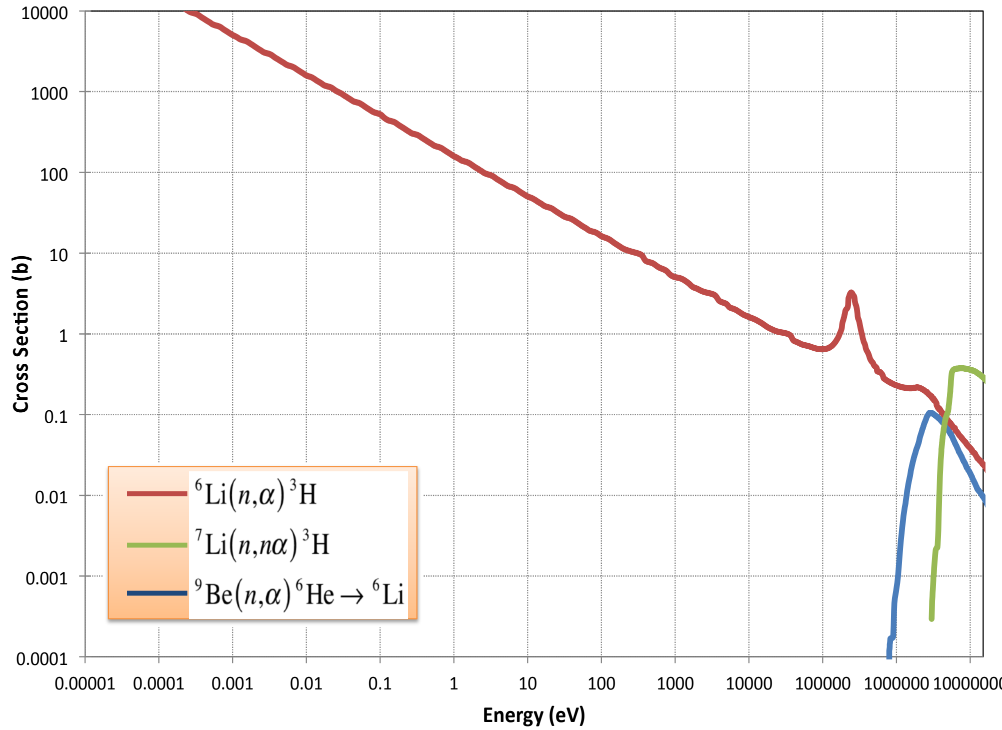


Table 1. Sources and rates of production of tritium in a 1000-MW(e) MSBR[a]

	Production rate (Ci/day)
Ternary fission	31
$^{6}Li(n,\alpha)^{3}H$	1210
$^{7}Li(n,n\alpha)^{3}H$	1170
$^{19}F(n,^{17}O)^{2}H$	9
Total	2420

[a] From Ref. 1.

Source: Mays, ORNL/TM-5759

Tritium Mitigation Methods Include Stripping, Blocking, and Trapping

- Largest technical challenge for stripping is the small diffusion of tritium in salt
  - Necessitates intimate mixing of salt and stripping material
    - Gas sparging or spraying in gas space using fine droplets
    - Turbulent flow (to promote mixing) across large surface area window (e.g., double-walled heat exchanger)
    - Flow through packed bed of absorbers
  - Palladium alloys have highest tritium diffusion coefficient
    - Nickel may be acceptable and is much less expensive
  - Carbon traps tritium at operating temperatures - desorbs at high temperatures (peak storage at ~800°C)
    - Nickel coating carbon improves trapping kinetics
    - Irradiation damage significantly increases number of traps
  - Several lanthanides form stable tritides (e.g., Y or Sm)
- Tritium trapping in coolant salt was demonstrated in NaF-$NaBF_4$ (8–92 mol%) at engineering scale for MSBR

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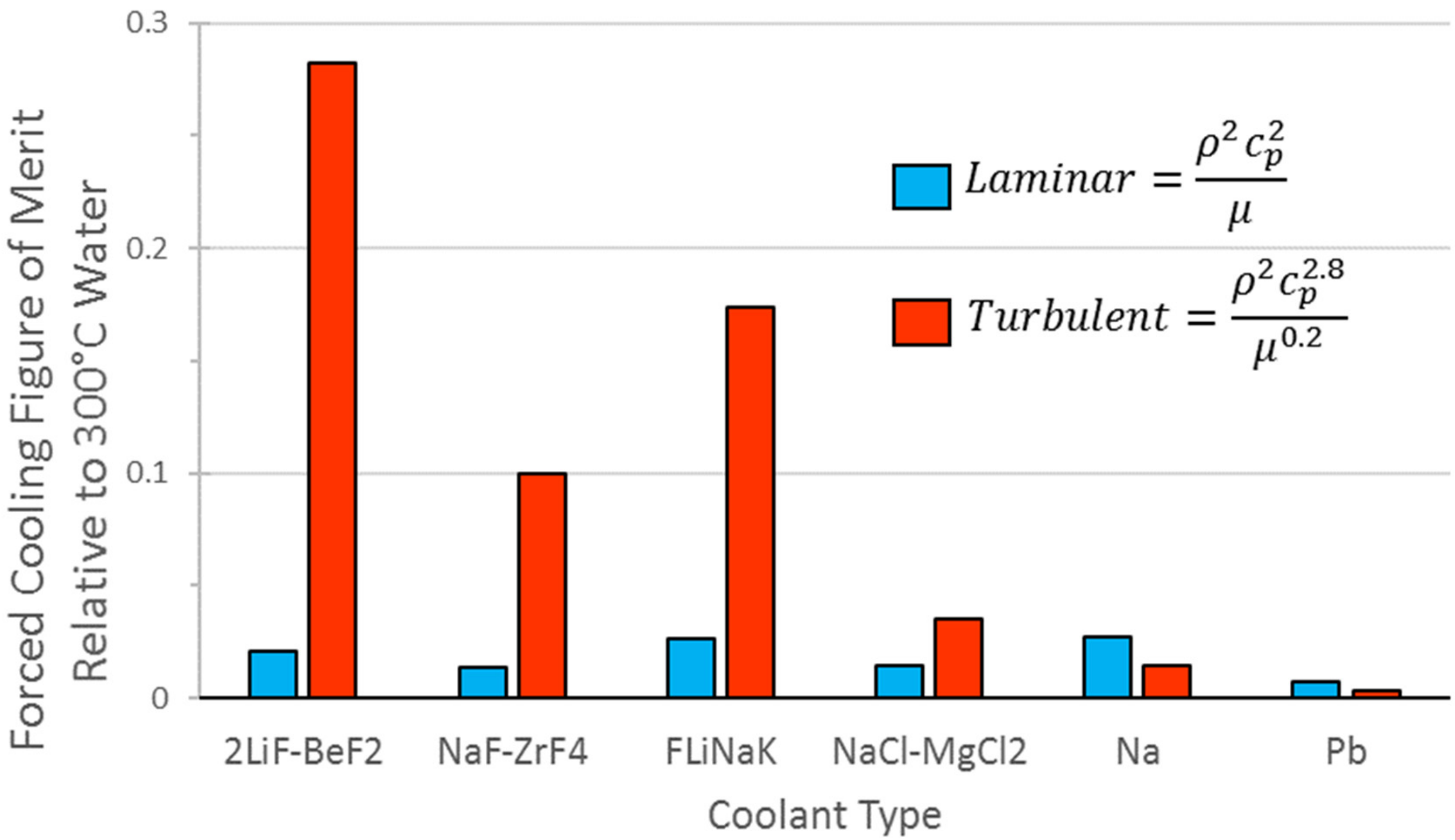
Molten Salts Have Attractive Heat Transfer Properties



- Large heat capacity and low viscosity are key properties

Source:
Nuclear Engineering Handbook 9-90,
D. F. Williams et al., ORNL/TM-2006/12

ρ = density
c_p= heat capacity
μ = dynamic viscosity
β = volumetric expansion coefficient

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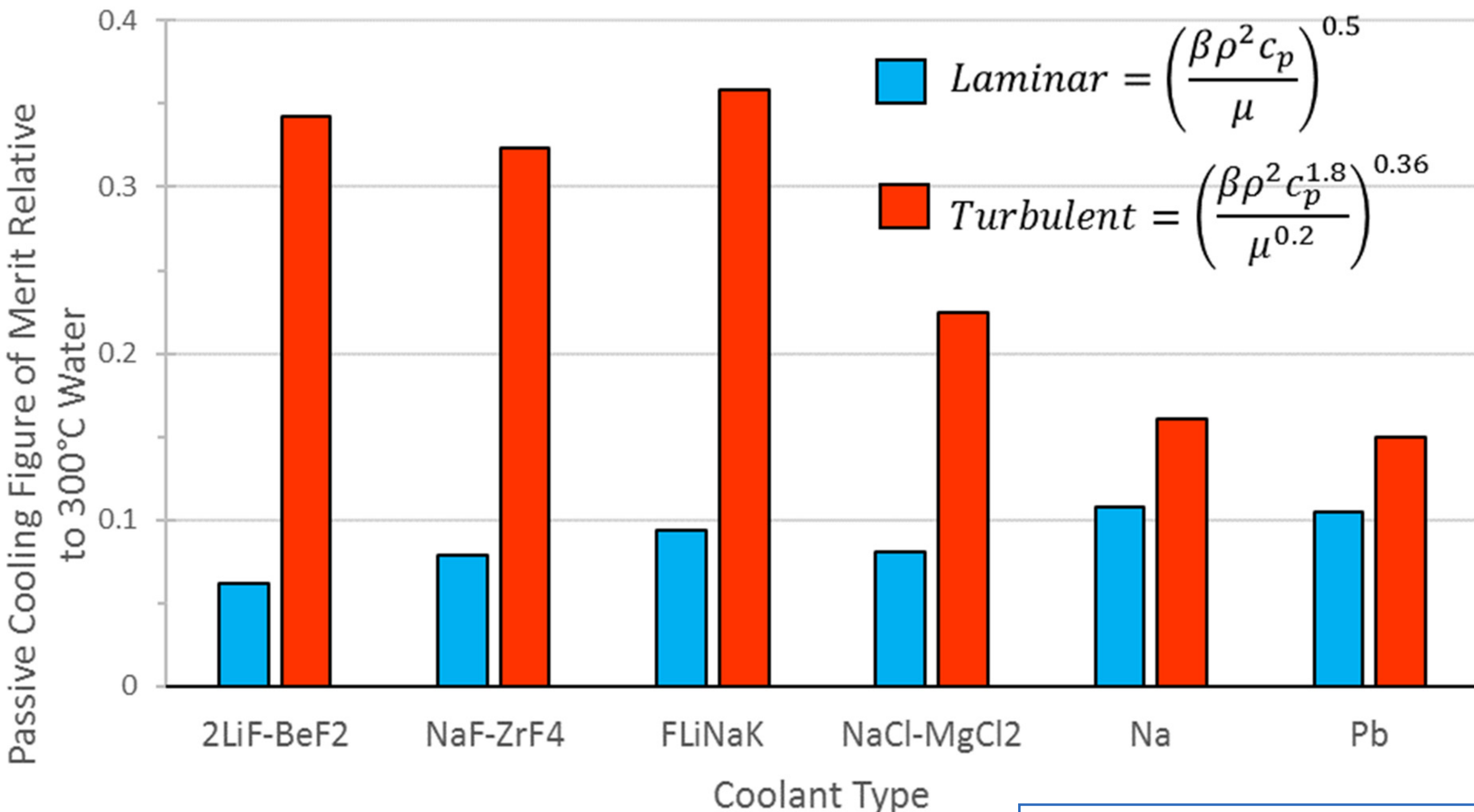
Molten Salt Passive Cooling Characteristics are Favorable



ρ = density
c_p = heat capacity
μ = dynamic viscosity
β = volumetric expansion coefficient

- Volumetric expansion with temperature provides buoyancy driving force

Source:
Nuclear Engineering Handbook 9-90,
D. F. Williams et al., ORNL/TM-2006/12

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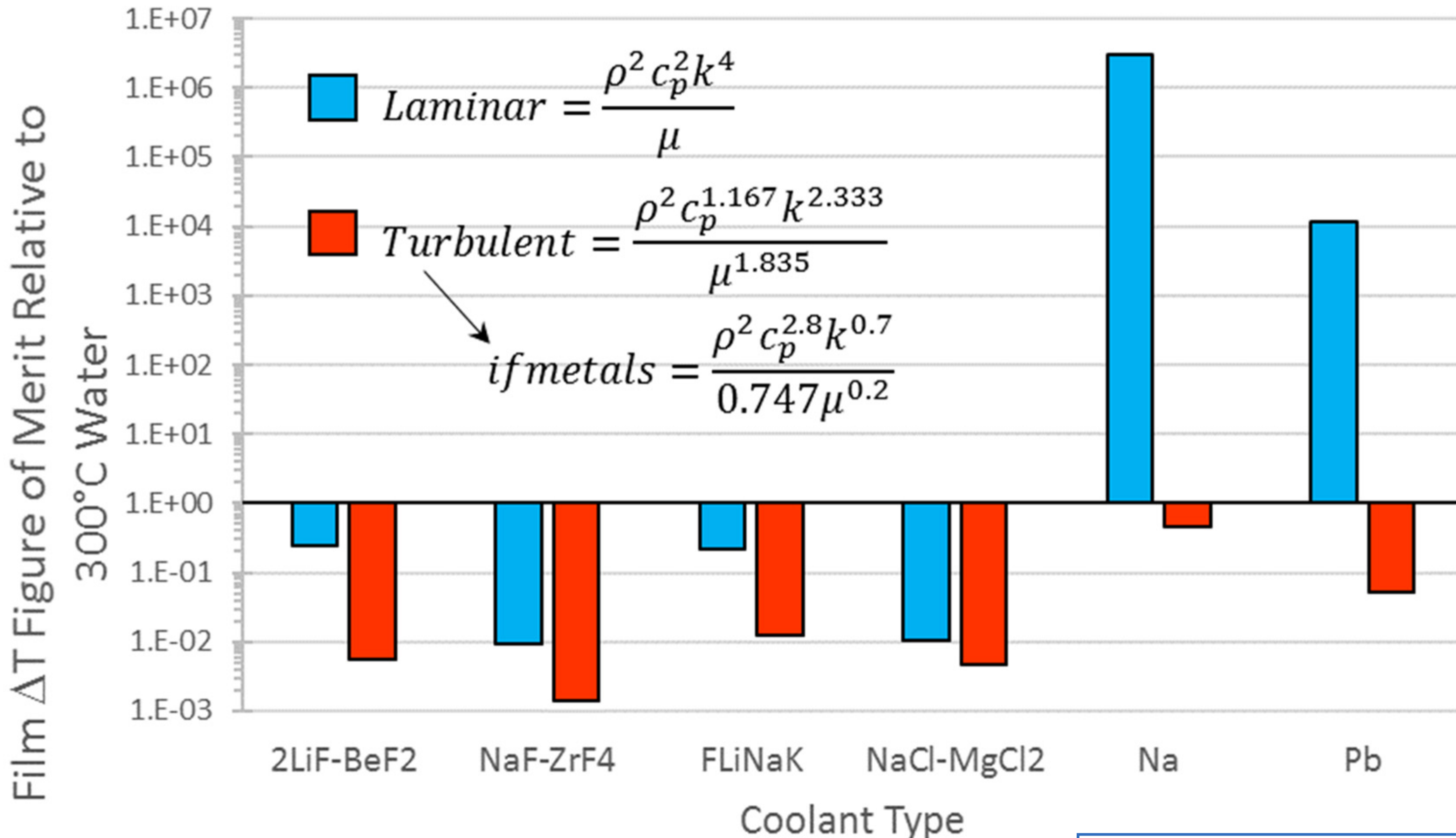
Salts Have Sharp Boundary Layer (High Prandtl Number)



ρ = density
c_p= heat capacity
μ = dynamic viscosity
β = volumetric expansion coefficient

- Turbulence is required for effective heat transfer (or tritium stripping)

Source:
Nuclear Engineering Handbook 9-90,
D. F. Williams et al., ORNL/TM-2006/12

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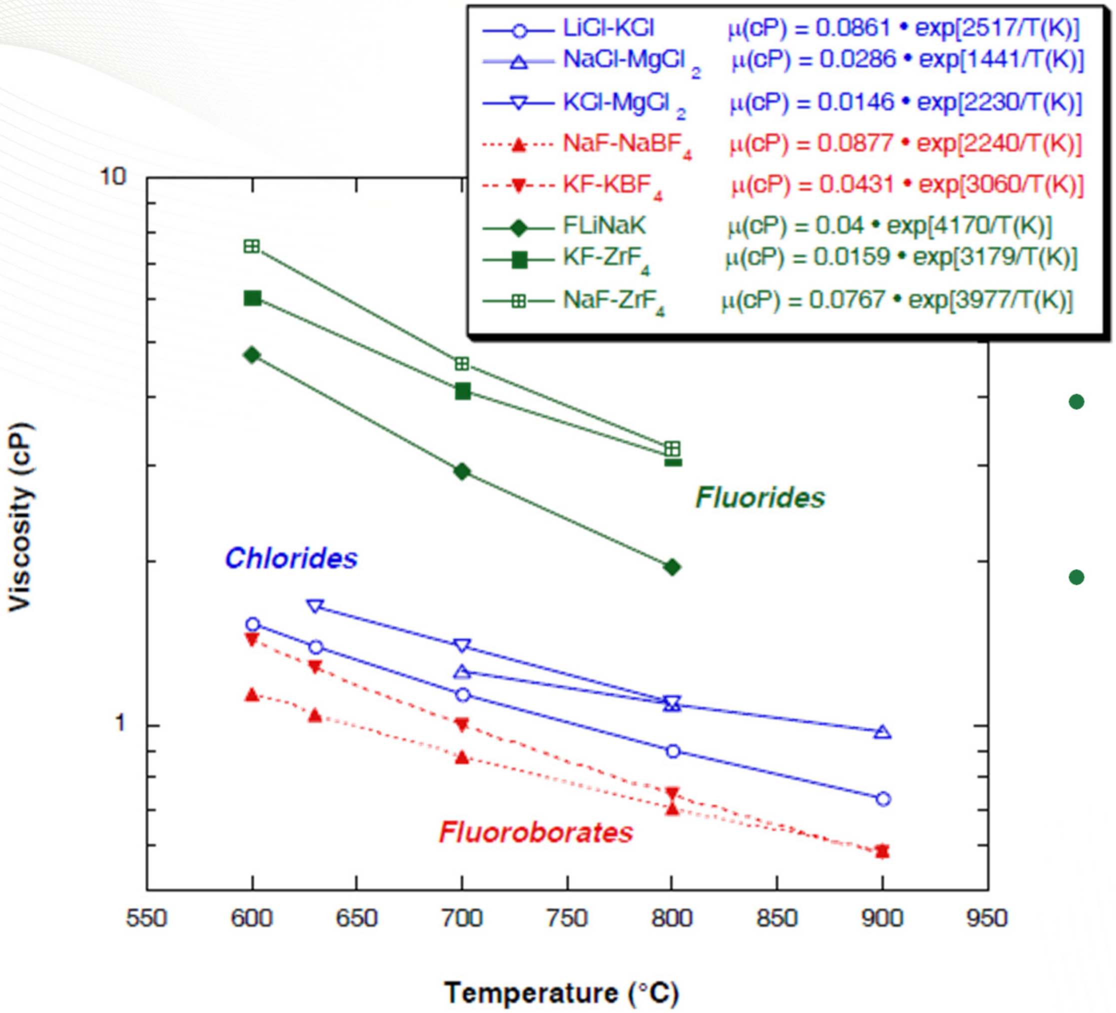
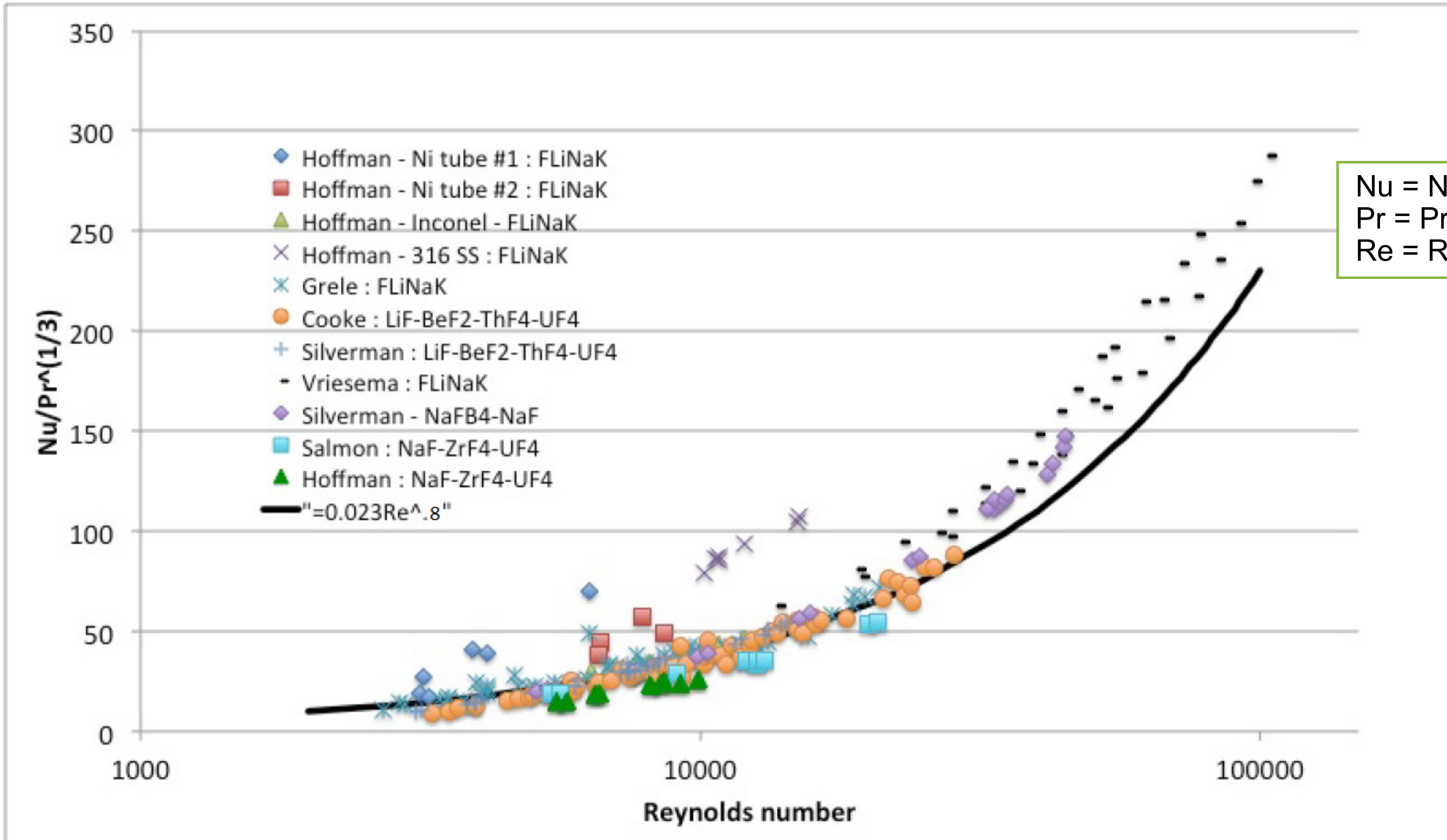
Salt Viscosity Decreases with Temperature



- Flow increases to hotter regions
- Improves temperature uniformity

Source:
D. F. Williams et al., ORNL/TM-2006/12
D. F. Williams, ORNL/TM-2006/69

Significant Uncertainty Remains in Fluoride Salt Turbulent Heat Transfer



Nu = Nusselt number
Pr = Prandtl number
Re = Reynolds number

Source: Yoder, ICAPP 14332, 2014

- Little experimental data with few material combinations and geometries
- Y-axis is a common heat transfer correlation for fully developed turbulent flow in tubes

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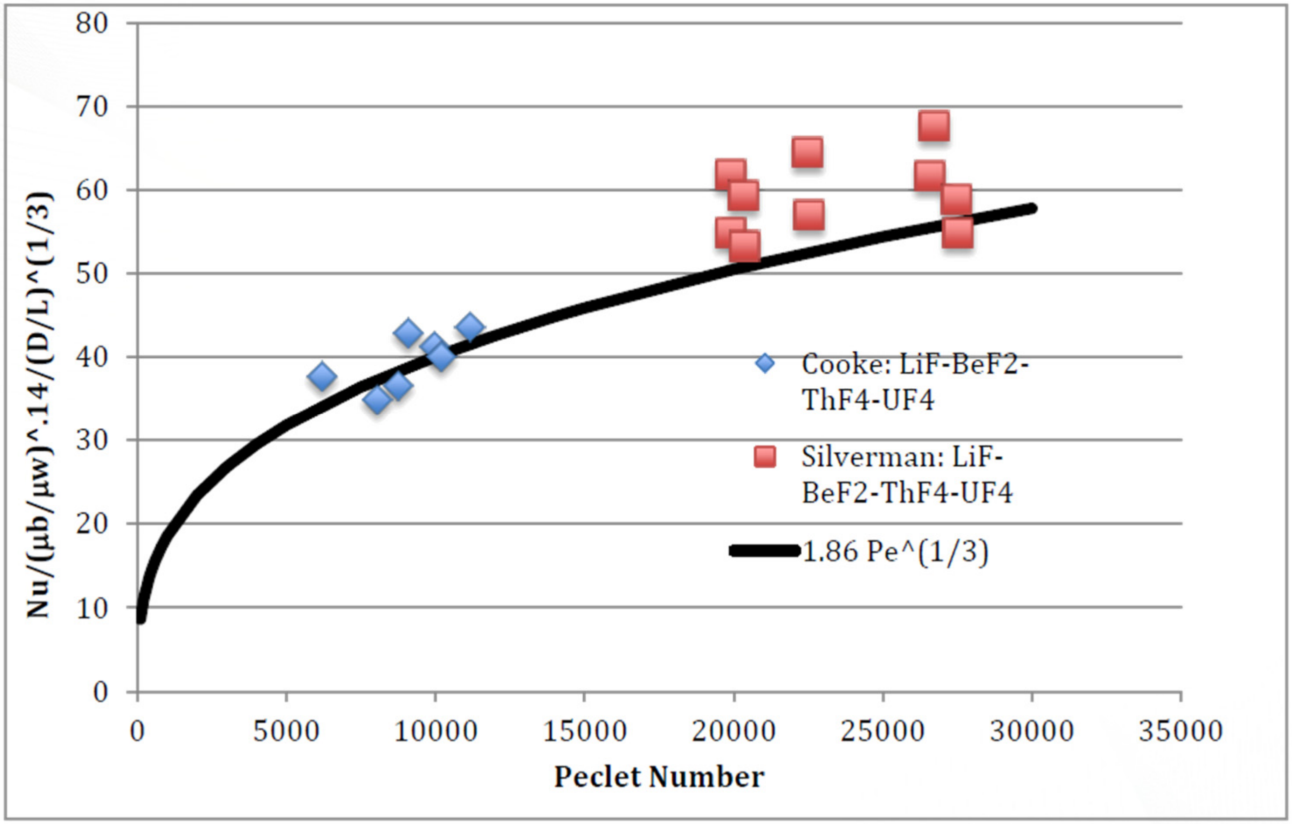
Laminar Flow Heat Transfer Also Has Significant Remaining Uncertainty



Source:
Yoder,
ICAPP 14332, 2014

- Axes selected to enable comparison with prior laminar flow correlations (Seider and Tate)
- Peclet number is a dimensionless ratio of the thermal energy convected to the fluid to the thermal energy conducted within the fluid

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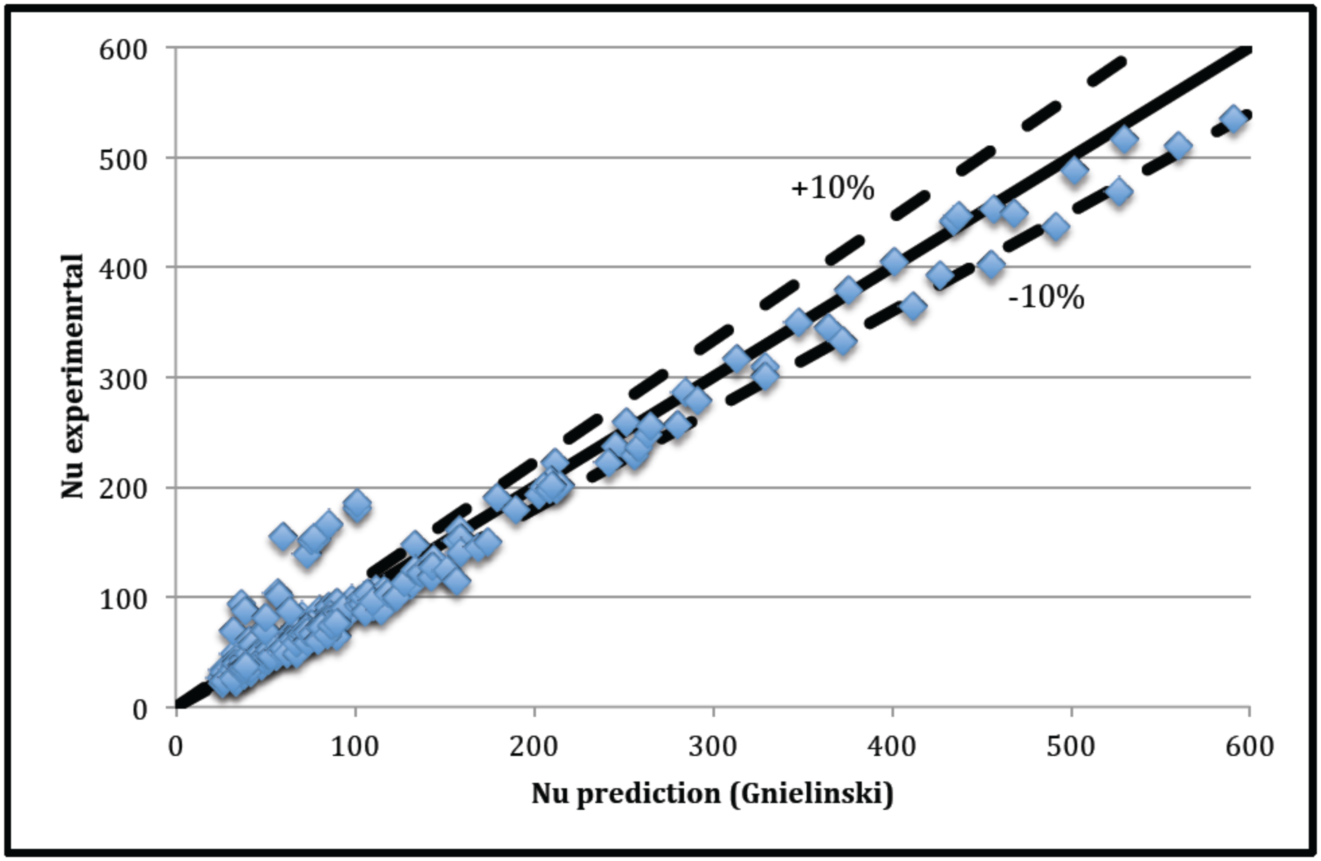
Significant Remaining Uncertainty in Prediction of Conductive / Convective Heat Transfer Ratio



Source:
Yoder,
ICAPP 14332, 2014

- Plot compares experimental and predicted conductive/convective heat transfer ratios
  - Prediction based upon reference Gnielinski correlation - commonly used for heat transfer comparisons

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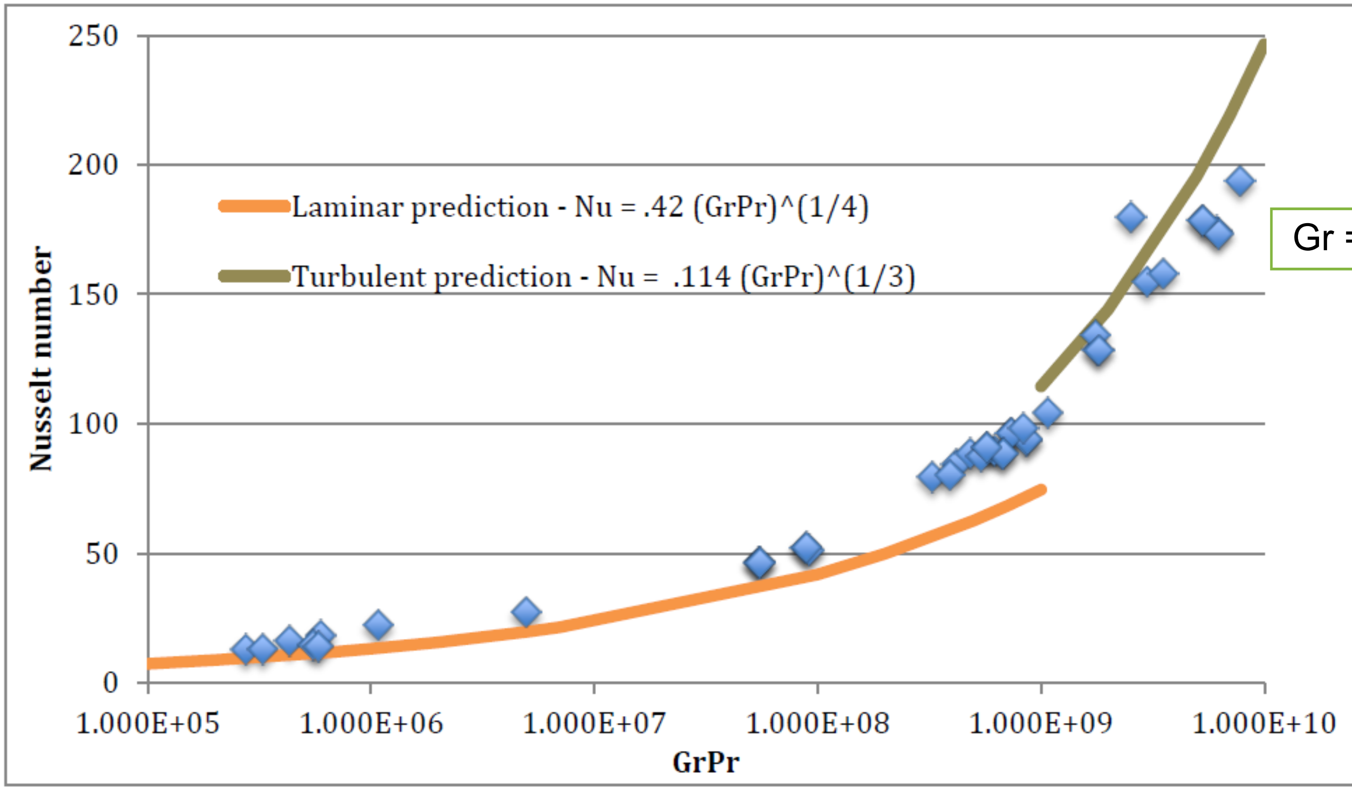
Natural Circulation Heat Transfer Has Significant Remaining Uncertainty



Gr = Grashof number

Source:
Yoder,
ICAPP 14332, 2014

- Product of Grashof and Prandtl number (X-axis) is the Rayleigh number associated with buoyancy-driven flow
  - Above critical Rayleigh number heat transfer is primarily convection below primarily conduction
  - Y-axis is ratio of convective to conductive heat transfer

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Heat Transfer Uncertainties Affect Operating Margin Calculations

- Material combinations and geometries of interest to MSRs have not been thoroughly characterized in past experiments
- Sources of experimental uncertainty include:
  - Salt purity and purification during the experiment
  - Film layers/deposits on heated surfaces
  - Temperature
- More targeted, controlled experimental data is required to improve the confidence in thermophysical property correlations

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Molten Fluorides Are Highly Thermodynamically and Radiolytically Stable

- Salts are combinations of strongly electronegative elements with strongly electropositive metals
  - Very high bond energies
  - Negative change in Gibbs free energy ($-\Delta G_f$) > 100 kcal/mol-F
  - Structural metal fluorides have Gibbs free energies at least 20 kcal/mol-F less negative
    - MSRE graphite and Hastelloy N exposed to coolant salt was untouched after ~3 years of operation
  - Salt radiolysis is overwhelmed by recombination at operating temperatures

Source: ORNL/TM-4174

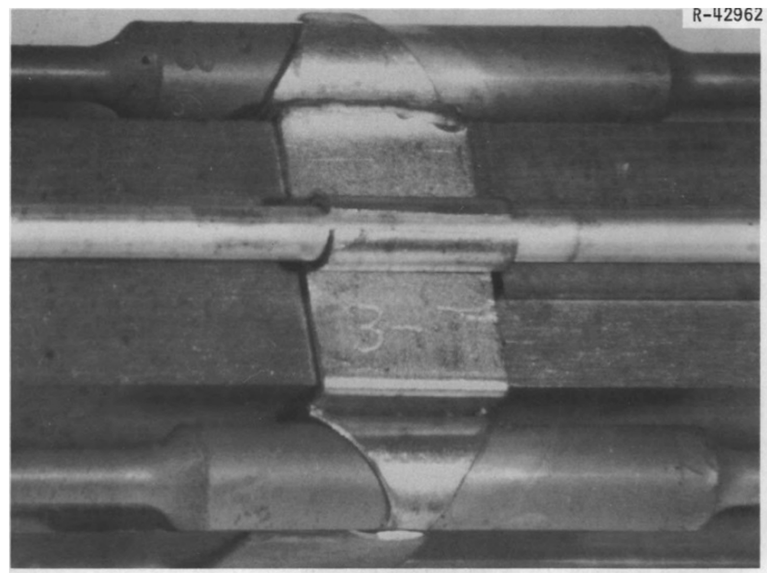

Fig. 1. Graphite and Hastelloy N Surveillance Assembly Removed from the Core of the MSRE After 72,400 Mwhr of Operation. Exposed to flowing salt for 15,300 hr at 650°C.

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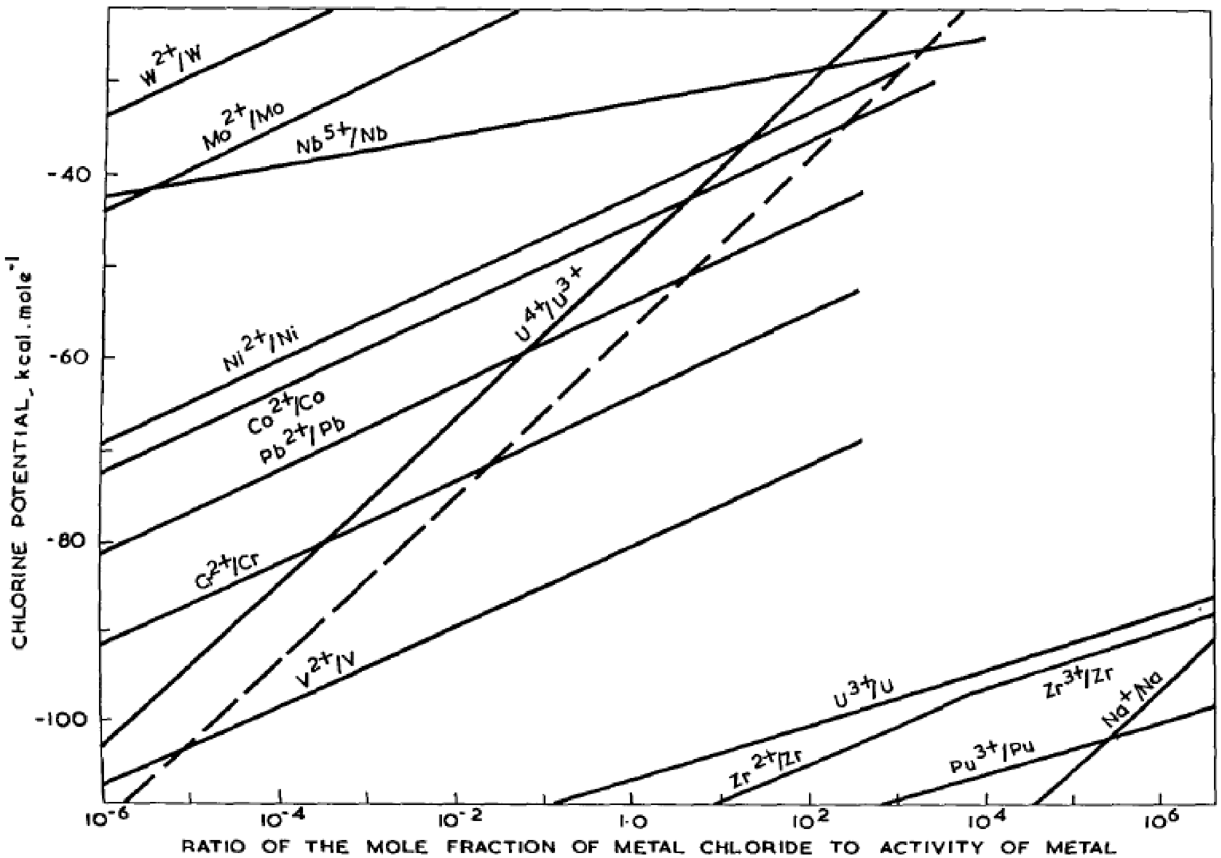
Thermochemical Stability Drives Both Corrosion and Fissile Solubility

- Increased free chlorine results in larger amounts of dissolved structural alloy chlorides
- Increasing ratios of UCl_4/UCl_3 restrict acceptable choice of structural alloys
- Use of nickel-based structural alloys restricted to UCl_4/UCl_3 ratios of roughly 0.003 to 5%
  - Smaller amounts of UCl_4 results in disproportionation of UCl_3
    $4UCl_3 \rightleftarrows UCl_4 + U^0$
- Refractory coatings would enable higher UCl_4/UCl_3 ratios



1. Chlorine Potential Diagram at 1000°K.

Source:
Harder, Long, and Stanaway, *Nuclear Metallurgy* 15:405-432, 1969.

- $PuCl_3$ disproportionation is less favorable than that of UCl_3

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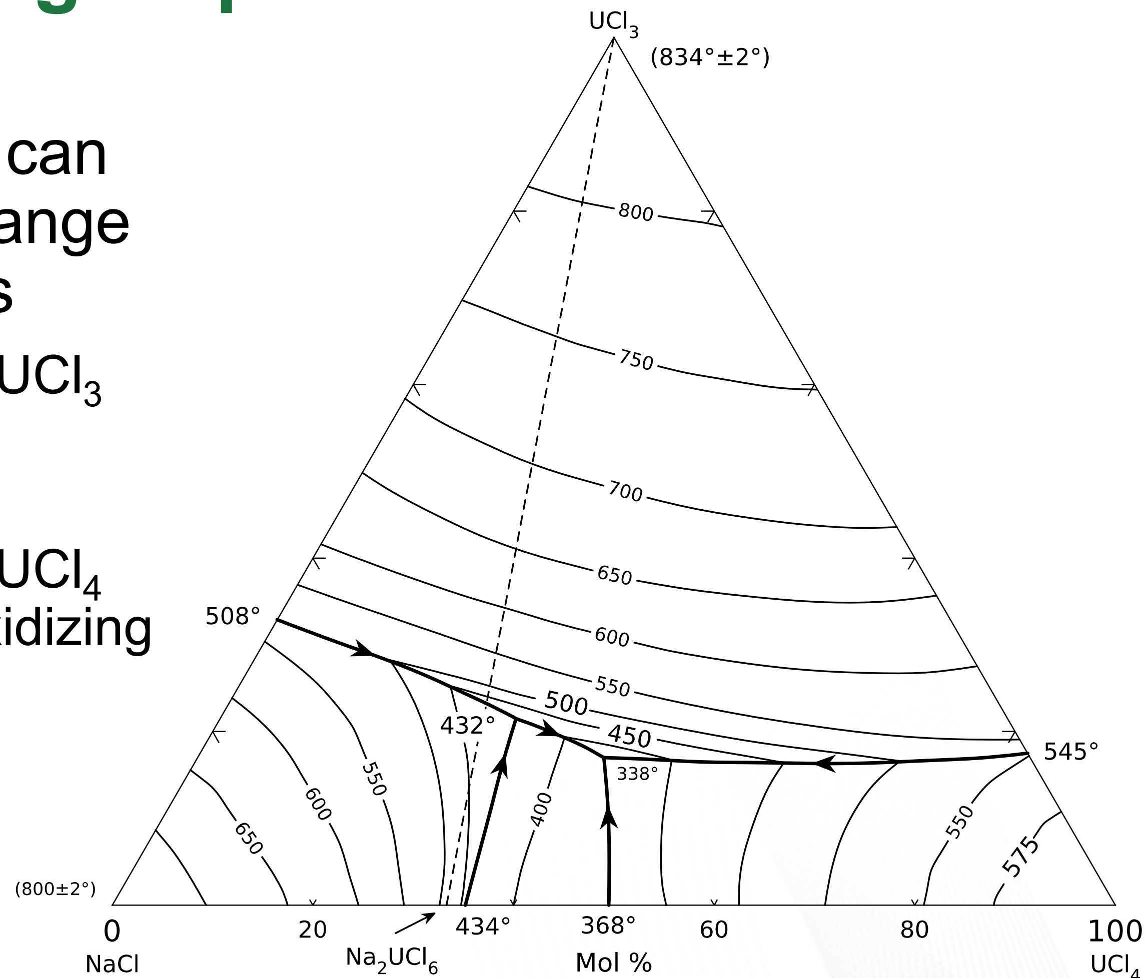
Phase Diagrams of Chloride Fuel Salts Show Fuel System Design Options

- A ~500°C melt point can be achieved with a range of UCl_3 to UCl_4 ratios
  - Systems with higher UCl_3 fractions have lower uranium loading
  - Systems with higher UCl_4 fractions are more oxidizing (corrosive)



Source: V. N. Desyatnik et al. *At. Energ.*, 31 [6] 631-633 (1971); *Sov. At. Energy* (Engl. Transl.), 31 [6] 1423-1424 (1971). Fig. 05953—System NaCl-UCl4-UCl3

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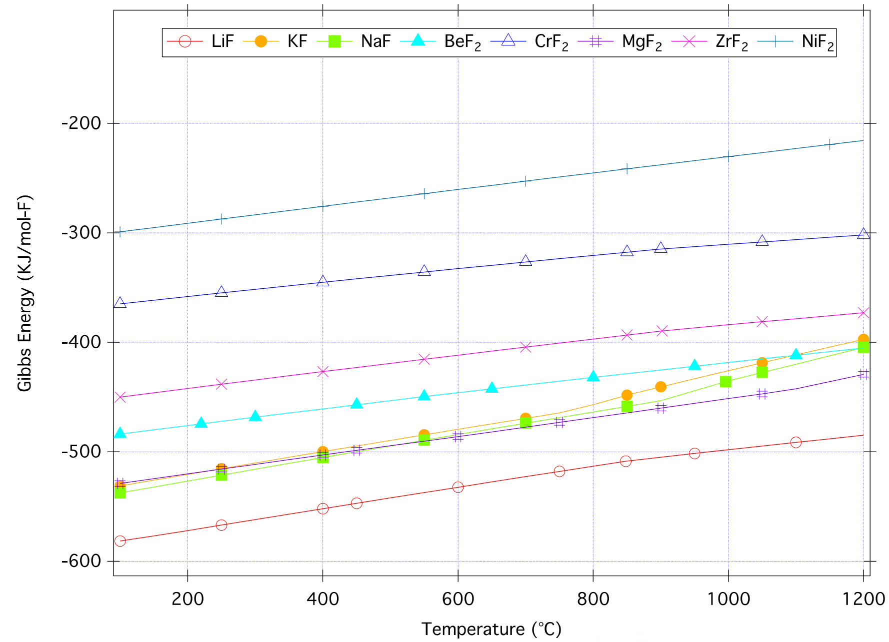
Maintaining Mildly Reducing Redox Conditions Key to Enabling Use of Engineering Alloys

- Use of a circulating redox buffer provides means to maintain redox condition
  - Fission changes oxidation state of salts
- Ratio of U^{4+}/U^{3+} serves as a measure of the redox potential of the salt
  - Applicable to both fluoride and chloride salts
  - Adding beryllium to FLiBe
- Fluoride salts will likely have an ideal ratio of ~10–100

Source:
Baes,
Keiser, ORNL/TM-6002



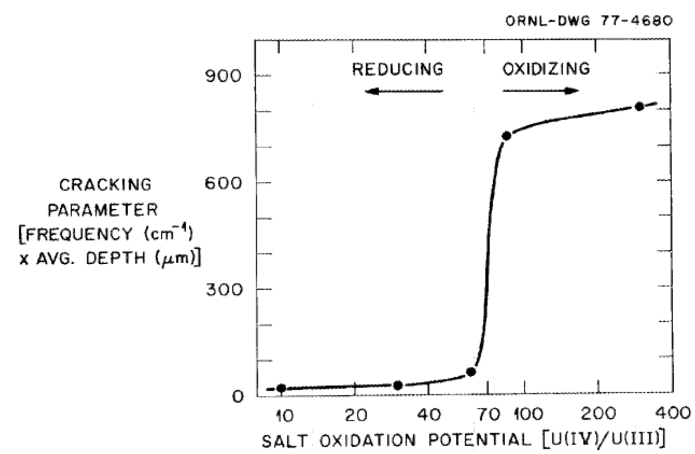


Fig. 12. Cracking Behavior of Hastelloy N Exposed 260 hr at 700°C to MSBR Fuel Salt Containing $CrTe_{1.266}$.

Fission Process Continuously Alters the Fuel Salt Redox Conditions

- When a U or Pu ion fissions, the available electrons will rearrange on each fission product to satisfy its valence requirements and produce either net oxidizing or reducing conditions in the melt
  - For ^{235}U (as UF_4) four F ions are released. The fission products require less than four 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. The fission products require more than three and thus there will be a F ion deficit with net reducing conditions
- MSRE periodically added metallic beryllium (strong reducing agent) to maintain UF_4/UF_3 ratio

Salt Type	Fission Product	Oxidation State (Z)	Yield (Y) [atoms]	Cl atoms reacted (Y*Z)
Chloride Salt (UCl_3)	Kr, Xe	0	25	0
	Rb, Cs	1	19	19
	Sr, Ba	2	10	20
	Rare Earths	3	46	138
	Zr	3	22	66
	Nb, Mo	0	2	0
	Te, I	0	6	0
	Pd, Re, Rh Ag, Cd	0	61	0
	Total Cl atoms reacted out of 300 available			243
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

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

Because Chemical Activity in Molten Salts Is Controlled by Melt Composition…

- Monovalent salts are “basic” in that they supply fluoride ions (F^-)
- Polyvalent salts are “acidic” in that they form complexes with F^-
- Lewis acid/base coordination equilibria are established
  - $ZrF_4 + 3F^- \leftrightarrow ZrF_7^{3-}$
  - $BeF_2 + 2F^- \leftrightarrow BeF_4^{2-}$
- The chemical reactivities of these and other metal ions are higher when they are not sufficiently coordinated with fluoride ions
- In the absence of the extra fluoride ions supplied by LiF component, for example, ZrF_4 and BeF_2 would be volatile and distill from the system

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Fission Products and Contaminants Would Alter Fuel Salt and Cover Gas Properties

- Oil leak along MSRE pump shaft resulted in foaming in pump bowl
  - Foam overflowed into gaseous waste handling system
- Noble fission products do not dissolve into salt and consequently lack a surface tension inhibition for entering cover gas (i.e., they readily enter the cover gas)
- Contamination particles, solid oxide precipitate, etc., may form a scum layer on the salt surface

Source:
Yoder et al., ORNL/TM-2014/499

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Fluoride Salts Are Vulnerable to Radiolytic Decomposition at Low Temperatures

- Intense radiation creates more free fluorine than is recombined below ~200°C
- Experience with chloride salts is almost nonexistent
  - Likely has similar vulnerability as fluoride salts
  - Pyroprocessing salts and conditions are different from fuel salts
- Free fluorine can react with structural materials resulting in dramatically increased corrosion or converting solid UF_4 to gaseous UF_6
  - Origin of the issue with the stored MSRE fuel salt in the 1990s

Source:
Haubenreich, ORNL-TM-3144, 1970

Long Term Waste Forms from MSRs Remain Unproven

- Primary US work remains "Applied Technology"
- Offgas sorbent could serve as ultimate fission gas disposal medium
  - Charcoal beds were employed for MSRE
- May be possible to make fluorides more stable by conversion to a fluorophosphate
- Chlorides are currently converted to a *salt-cake* waste form as part of the ongoing EBR-II processing campaign
- Synthetic rock process developed by ANSTO appears applicable
- Dutch SALIENT project has primary objective to develop final waste form for their test salts

Characteristics of MSRs Derive from the Chemistry and Physics of Halide Salts

- Low pressure, high temperature operation
- Dissolve useful amounts of fissile material
- Chemically compatible with engineering alloys in mildly reducing environments
- Strong passive safety features
  - Negative reactivity feedback
  - Natural circulation-based decay heat removal
  - Reduced potential for radionuclide release
- Fluoride salts have substantially more experimental data than chloride salts for reactor operations
- Tritium production from lithium-bearing salts can be mitigated by stripping, blocking, and trapping