MSR lattice optimization for economic salts with LEU fuel
Dr. Ondřej Chvála <ochvala@utk.edu>

Presentation overview

- Historical context and lessons
- Contemporary R&D issues
- MSR salt & lattice choices
- LEU fueled MSRs economic viability
- NB: http://web.utk.edu/~ochvala/MSR/
- Thanks to Terrestrial Energy Inc.
ORNL Aircraft Nuclear Reactor Progress (1949-1960)

1949 – Nuclear Aircraft Concept formulated

1951 – R.C. Briant proposed Liquid-Fluoride Reactor

1952, 1953 – Early designs for aircraft fluoride reactor

1954 – Aircraft Reactor Experiment (ARE) built and operated successfully (2500 kWt2, 1150K)

1955 – 60 MWt Aircraft Reactor Test (ART, “Fireball”) proposed for aircraft reactor

1960 – Nuclear Aircraft Program canceled in favor of ICBMs
The Aircraft Reactor Experiment (1954)

In order to test the liquid-fluoride reactor concept, a non-circulating core, sodium-cooled reactor was hastily converted into a proof-of-concept liquid-fluoride reactor.

The Aircraft Reactor Experiment ran for 1000 hours at the highest temperatures ever achieved by a nuclear reactor (1150 K).

- Operated from 10/30/1954 to 11/12/1954
- Liquid-fluoride salt circulated through beryllium reflector in Inconel tubes.
- $^{235}$UF$_4$ dissolved in NaF-ZrF$_4$
- Produced 2.5 MW of thermal power.
- Gaseous fission products were removed naturally through pumping action.
- Very stable operation due to a large negative temperature-reactivity coefficient.
- Demonstrated load-following operation without control rods.
It wasn’t that I had suddenly become converted to a belief in nuclear airplanes. It was rather that this was the only avenue open to ORNL for continuing in reactor development. That the purpose was unattainable, if not foolish, was not so important:

A high-temperature reactor could be useful for other purposes even if it never propelled an airplane...

—Alvin Weinberg
Molten Salt Reactor Experiment (1965-1969)

ORNLs' MSRE: 8 MW(th)
Designed 1960 – 1964
Started in 1965, 5 years of successful operation

Developed and demonstrated on-line refueling, fluorination to remove uranium UF4+F2→UF6,
Vacuum distillation to clean the salt

Operated on all 3 fissile fuels U233, U235, Pu239

Some issues with Hastelloy-N found and solved

Further designs suggested (MSBE, MSBR, DMRS), none built

After Alvin Weinberg was removed from ORNL directorate, very little work done, almost no funding

Current Predicament

- ORNL's program in the 1960s was predicated on many historical circumstances, which are **not valid any more**.
- Current political priorities: inherent “walkaway” safety, proliferation resistance, TRU actinide minimization and spent nuclear fuel inventory management, among others.
- Economic necessity: **minimization of upfront costs**, maximization of resource utilization (see later), and exploring new markets.
- Any futuristic R&D program needs to get actually funded.
- Any new reactor R&D and deployment (R&D&D) needs:
  - to get regulated using the standard rules tailored to LWR → significant but not insurmountable challenge,
  - necessitates new generation of experts in related areas.
Glenn Seaborg: “Status in 1969”

LMFBR success dependent on simultaneous fulfillment of assumptions:

1) Electric demand doubles every decade

2) Nuclear will capture more electricity generation market share

3) Uranium will remain scarce

4) LMFBR R&D will be easy

5) Public and private funding will be available

“The non-fulfillment of any one, or at most two, of these assumptions might be sufficient to bring the whole edifice tumbling to the ground. In the actual event, none of the assumptions proved correct.”

Ideas for Partial Solutions

• Acknowledge differences between near-term and ultimate solutions, and realize the consequences to plant engineering.

• Use LEU as fuel – no Thorium means lower enrichment and less challenges from the regulatory authority.

• Simplify on-site reprocessing to basic fuel reconditioning
  • Gaseous FP sparging with He or other noble gas.
  • Extraction of refractory metals by Ni sponge in the primary circuit cold leg.
  • Move actual fuel reprocessing to a central location (i.e. later).

• Carrier salt selection is crucial to performance and cost.

• Low fuel enrichment and decent conversion ratios are attractive, while maintaining MS fluid fuel advantages.
Salt Selection – Considering Fluorides

- Alkali(ne)-halide carrier salts: fluorides minimize corrosion issues of all halides due to its extreme electro-negativity.

- $^{19}$F is rather light, which limits fast-spectrum reactors feasibility with UF$_4$ as a fuel.
  - Though the French/GenIV salt-only MSFR is close.

- Thermal reactors have many advantages:
  - Maximum reactivity configuration (safety).
  - Longer reactivity period (safety).
  - Much less fissile feed need for criticality (fixed costs).

- Several fluoride salts are commonly considered.

- Issues: salt up-front cost ($^7$Li), small scale lab work impediments (Be), tritium production (Li, Be), and industry start-up expenses (Rb).
Some Other Ignored Engineering Design Issues

- Salt THD performance, but all likely good enough.
- Vessel material – SS316, HastalloyN, Inconel, ...
  - Temperature dependent
  - Important fixed expense
- HX configuration and design
  - SmAHTR shows possible avenues of exploration, but is wrong.
  - DR(A)CS outside of scope of this presentation.
- ...
- New and innovative ideas of trading fixed costs for variable ones much encouraged. Email me!
# Salt Selection

- Salts composition differences impacts salt cost, tritium production, and neutronic performance.
- THD differences neglected.

<table>
<thead>
<tr>
<th>Salt composition</th>
<th>Melting point [°C]</th>
<th>Density [g/cm³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. 72%⁷LiF-16%BeF₂-12%UF₄</td>
<td>480</td>
<td>3.353</td>
</tr>
<tr>
<td>2. 73%⁷LiF-27%UF₄</td>
<td>490</td>
<td>4.340</td>
</tr>
<tr>
<td>3. 78%NaF-22%UF₄</td>
<td>618</td>
<td>4.056</td>
</tr>
<tr>
<td>4. 49%NaF-38%ZrF₄-13%UF₄</td>
<td>540</td>
<td>3.757</td>
</tr>
<tr>
<td>5. 58%NaF-30%BeF₂-12%UF₄</td>
<td>525</td>
<td>3.208</td>
</tr>
<tr>
<td>6. 74%NaF-12%BeF₂-14%UF₄</td>
<td>500</td>
<td>3.437</td>
</tr>
<tr>
<td>7. 46%NaF-33%RbF-21%UF₄</td>
<td>470</td>
<td>4.026</td>
</tr>
<tr>
<td>8. 50.5%NaF-21.5%KF-28%UF₄</td>
<td>490</td>
<td>4.326</td>
</tr>
</tbody>
</table>
Salt/Graphite Lattice

- Parametric scan of neutronics with different salts.
- Infinite hexagonal lattice of graphite and salt fuel.
- Lattice parameters are channel pitch $p$ and salt fraction $f$.

- Salt channel radius: $r^2 = \frac{p^2 \sqrt{3}}{2\pi} f$

- Reflective unit cell:
Other Material Parameters

- $^7\text{Li}$ is 99.995% $^7\text{Li}$
- All salts at 650°C using 900K ENDF/B-VII.0 libraries
- Graphite density is assumed as 1.8 g/cm$^3$, temperature 700°C, using 900K ENDF/B-VII.0 libraries. Impurities modeled as 2ppm of boron. Thermal scattering library is ENDF/B-VII.0sab at 1000K.
- One case uses Zr$_5$H$_8$ moderator, density 5.61 g/cm$^3$, same libraries and temperatures. $S(\alpha\beta)$ treatment applied for both Zr and H in Zr$_5$H$_8$. 
Criticality Searches by Iterative Secant Method

- Parameters are channel pitch (1 to 60 cm) and fuel salt fraction (0.5% to 55%). ~900 points sampled/salt.

- For each point in the parameter space, criticality search is performed to find BoC criticality (1<k<1.001) using the iterative Secant method.

\[
e^{(n+1)} = e^{(n)} - \rho(e^{(n)}) \frac{e^{(n)} - e^{(n-1)}}{\rho(e^{(n)}) - \rho(e^{(n-1)})}
\]

- NB: This is a finite difference version of the Newton's method.

\[
e = \text{enrichment (x axis)}
\]
\[
\rho(e) = \text{reactivity (y axis)}
\]
\[
\text{upper index = iteration step}
\]

- NB: This is a finite difference version of the Newton's method.
Interesting Quantities

- Enrichment for the critical lattice
- Conversion ratio (CR):
  \[ CR = \frac{^{238}U\ captures}{^{235}U\ captures + ^{235}U\ fissions}. \]
- Reproduction factor:
  \[ \eta = \frac{\nu}{1+\alpha} = \frac{2.44}{1 + \frac{^{235}U\ captures}{^{235}U\ fissions}}. \]
- Fast fission bonus = $^{238}U$ fission fraction.
- Figure of Merit: CR/enrichment
49%NaF-38%ZrF₄-13%UF₄.

58%NaF-30%BeF₂-12%UF₄.

74%NaF-12%BeF₂-14%UF₄.
46%NaF-33%KBF-21%UF₄

50.5%NaF-21.5%KF-28%UF₄

73%LiF-27%UF₄ with Zr5H8 moderator
Summary of the critical lattices properties: the minimum enrichment of $^{235}\text{U}$ needed for criticality and its location in the parameter space of pitch versus salt fraction; the maximum CR and its position in the parameter space. Table below: maximum FoM.

<table>
<thead>
<tr>
<th>Salt composition</th>
<th>$^{235}\text{U}[$%$]$</th>
<th>$p$ [cm]</th>
<th>$f$ [%]</th>
<th>CR</th>
<th>$p$ [cm]</th>
<th>$f$ [%]</th>
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</thead>
<tbody>
<tr>
<td>$72%^7\text{LiF}-16%\text{BeF}_2-12%\text{UF}_4$</td>
<td>0.944</td>
<td>30.0</td>
<td>10.0</td>
<td>0.937</td>
<td>50.0</td>
<td>37.5</td>
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<tr>
<td>$73%^7\text{LiF}-27%\text{UF}_4$</td>
<td>0.813</td>
<td>28.0</td>
<td>8.0</td>
<td>0.983</td>
<td>50.0</td>
<td>32.5</td>
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<tr>
<td>$78%\text{NaF}-22%\text{UF}_4$</td>
<td>1.181</td>
<td>24.0</td>
<td>6.0</td>
<td>0.877</td>
<td>60.0</td>
<td>40.0</td>
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<td>1.458</td>
<td>28.0</td>
<td>10.0</td>
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<td>1.0</td>
<td>18.0</td>
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<td>$58%\text{NaF}-30%\text{BeF}_2-12%\text{UF}_4$</td>
<td>1.416</td>
<td>28.0</td>
<td>8.0</td>
<td>0.852</td>
<td>1.0</td>
<td>14.0</td>
</tr>
<tr>
<td>$74%\text{NaF}-12%\text{BeF}_2-14%\text{UF}_4$</td>
<td>1.417</td>
<td>26.0</td>
<td>8.0</td>
<td>0.850</td>
<td>1.0</td>
<td>14.0</td>
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<tr>
<td>$46%\text{NaF}-33%\text{RbF}-21%\text{UF}_4$</td>
<td>1.241</td>
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<td>8.0</td>
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<td>1.0</td>
<td>10.0</td>
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<td>$50.5%\text{NaF}-21.5%\text{KF}-28%\text{UF}_4$</td>
<td>1.305</td>
<td>24.0</td>
<td>6.0</td>
<td>0.852</td>
<td>1.0</td>
<td>10.0</td>
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<table>
<thead>
<tr>
<th>Salt composition</th>
<th>FoM</th>
<th>$p$ [cm]</th>
<th>$f$ [%]</th>
<th>$^{235}\text{U}[$%$]$</th>
<th>CR</th>
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<tbody>
<tr>
<td>$72%^7\text{LiF}-16%\text{BeF}_2-12%\text{UF}_4$</td>
<td>0.827</td>
<td>28.0</td>
<td>14.0</td>
<td>0.987</td>
<td>0.815</td>
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<td>$73%^7\text{LiF}-27%\text{UF}_4$</td>
<td>1.036</td>
<td>28.0</td>
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<td>0.840</td>
<td>0.870</td>
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<tr>
<td>$78%\text{NaF}-22%\text{UF}_4$</td>
<td>0.559</td>
<td>30.0</td>
<td>12.0</td>
<td>1.259</td>
<td>0.703</td>
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<tr>
<td>$49%\text{NaF}-38%\text{ZrF}_4-13%\text{UF}_4$</td>
<td>0.400</td>
<td>30.0</td>
<td>16.0</td>
<td>1.534</td>
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<td>0.423</td>
<td>24.0</td>
<td>14.0</td>
<td>1.538</td>
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<td>$74%\text{NaF}-12%\text{BeF}_2-14%\text{UF}_4$</td>
<td>0.424</td>
<td>26.0</td>
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<td>$46%\text{NaF}-33%\text{RbF}-21%\text{UF}_4$</td>
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<td>1.281</td>
<td>0.615</td>
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<td>$50.5%\text{NaF}-21.5%\text{KF}-28%\text{UF}_4$</td>
<td>0.467</td>
<td>28.0</td>
<td>12.0</td>
<td>1.443</td>
<td>0.673</td>
</tr>
</tbody>
</table>
Note on Lattice Heterogeneities

• Molten Salt Reactor Experiment used the lattice pitch of 5.1 cm (2”), and the salt fraction of 22.5%
• Molten Salt Breeder Reactor used a similar pitch of ~5 cm and salt fractions of 13.2% and 37% in its two zones.
• Denatured Molten Salt Reactor's pitch ~15 cm, and the salt fraction of 13% in 95% of the core.
• These seem smaller pitches than optimal.
Comparison of Enrichments for $k=1$

- The dependence of $^{235}\text{U}$ enrichment on the lattice pitch for the salt fraction value which maximizes FoM.
How Do Enrichment Needs Rise With Leakage?

- Example: at 2% leakage ($k=1.02$), we need 5% higher enrichment relative to $k=1 \rightarrow$ Instead of 1% LEU we need 1.05% LEU
Impact of Conversion Ratios on Power Cost

- Conversion ratios (CR) in the lattice configuration investigated range between 0.6 – 0.9
- Say 800kg needs to fission for 1GWe.year → annual fresh fuel requirements range from 320 to 80 kg of fissile.
- Current cost of fresh U235 is below $40/g for LEU fuel.
- The total makeup fuel cost is thus $12.8M to $3.2M
- Which can be expressed as a part of the electricity cost in the range of 0.15 to 0.037 c/kWh.
Plutonium Solubility and LWR SNF Re-Use

- All the salts discussed are expected to dissolve 1-2mol% of trifluoride actinides, in particular PuF₃.
- Low fissile loadings, see below, seem to allow direct re-use of LWR SNF actinides in such hypothetical reactor.

Fraction of the fissile atoms for FoM maximizing lattices

<table>
<thead>
<tr>
<th>Salt composition</th>
<th>Fissile loading [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>72%⁷LiF-16%BeF₂-12%UF₄</td>
<td>0.118</td>
</tr>
<tr>
<td>73%⁷LiF-27%UF₄</td>
<td>0.227</td>
</tr>
<tr>
<td>78%NaF-22%UF₄</td>
<td>0.277</td>
</tr>
<tr>
<td>49%NaF-38%ZrF₄-13%UF₄</td>
<td>0.199</td>
</tr>
<tr>
<td>58%NaF-30%BeF₂-12%UF₄</td>
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</tr>
<tr>
<td>74%NaF-12%BeF₂-14%UF₄</td>
<td>0.214</td>
</tr>
<tr>
<td>46%NaF-33%RbF-21%UF₄</td>
<td>0.269</td>
</tr>
<tr>
<td>50.5%NaF-21.5%KF-28%UF₄</td>
<td>0.404</td>
</tr>
</tbody>
</table>
Future work

- Look at Zr5H8 moderator in inverted configuration.
- Investigate other moderator geometries, in particular moderating by inert graphite pebbles.
- Temperature-reactivity feed-backs for finite cores.
- Depletion studies for more realistic fuel cycle assessment.
- Reactor kinetics and dynamics.
- ...
- Build a reactor!
Conclusions

- LEU fueled MSRs seem as an attractive proposition!
- Large selection of fluoride salts seem all feasible with surprisingly low uranium enrichments.
- The data have been posted: http://web.utk.edu/~ochvala/MSR/
- Stay tuned for more to come. The website will be updated with new results.
- Thank you for your attention!

- Thanks also to Terrestrial Energy Inc. who made my presentation at ICAPP 2014 possible.