

Molten-Salt-Reactor Technology Gaps

Charles W. Forsberg
Oak Ridge National Laboratory*
P.O. Box 2008; Oak Ridge, TN 37831-6165
Tel: (865) 574-6783; Fax: (865) 574-0382
E-mail: forsbergcw@ornl.gov

File Name: MSR Publish: MSRGap.ICAPP06.Paper
Final Paper Due Date: March 15, 2006
Manuscript Date: February 10, 2006

Manuscript Number: 6295

Session: 3.09 Liquid-Salt-Cooled High-Temperature Reactors-III
2006 International Congress on the Advances in Nuclear Power Plants (ICAPP '06)
Embedded Topical in the 2006 American Nuclear Society Annual Meeting
American Nuclear Society
June 4–8, 2006
Reno, Nevada

The submitted manuscript has been authored by a contractor of the U.S. Government under contract DE-AC05-00OR22725. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

*Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract DE-AC05-00OR22725.

Molten-Salt-Reactor Technology Gaps

Charles W. Forsberg
Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee
Tel: (865) 574-6783; Email: forsbergcw@ornl.gov

Abstract — Molten salt reactors (MSRs) are liquid-fuel reactors that can be used for producing electricity or hydrogen as well as burning actinides and producing fissile fuels (breeding). Fissile, fertile, and fission products are dissolved in a high-temperature molten fluoride salt with a very high boiling temperature (~1400°C). The fuel salt flows through a reactor core, where fission occurs within the flowing salt; through an intermediate heat exchanger; and back to the reactor core. An intermediate heat-transfer loop transports the heat to a turbine hall or to a hydrogen production facility. Two experimental reactors were successfully built in the 1950s and 1960s. MSRs are being reexamined today because of their unique fuel cycle capabilities and safety characteristics. A technology gap analysis has been initiated to understand technological challenges for development and deployment. Some technology challenges have been resolved by new technologies that did not exist in the early 1970s when the program was shut down. Other technological challenges remain. Six areas (power cycles, fuel inventories, noble metal plate-out, fuel storage, high-level waste forms, and peak reactor temperatures) were examined. The results of the analysis are summarized.

I. INTRODUCTION

Between 1950 and 1976^{1,2} a large Molten Salt Reactor (MSR) development program was conducted in the United States, two test reactors were successfully operated, a design of a 1000-MW(e) reactor was completed, and plans were developed to construct a demonstration reactor. Since then, little research and development (R&D) has been done on MSRs. This paper summarizes recent work³ that identified technology gaps for deployment of MSRs and general technical advances that have either eliminated specific technology gaps or shown pathways to potentially more economically viable solutions.

The MSR was originally developed for the aircraft nuclear propulsion program, where a very high power density was required to minimize the reactor size and hence the weight of the reactor shielding. It was then developed as a breeder reactor and was the backup option to the sodium-cooled fast reactor. Ultimately, it was decided to concentrate efforts on the development of a single breeder reactor concept—the sodium-cooled fast reactor. These billion-dollar programs created the base MSR technology. The relatively trouble-free 8-MW(t)

Molten Salt Reactor Experiment (MSRE) provided an effective demonstration of many aspects of the reactor technology.

The MSR was chosen as one of the six Generation IV reactor concepts because it uses liquid fuel; that is, the fuel is dissolved in the coolant. All of the other reactors use solid fuel. The liquid fuel creates major advantages and some unique challenges. In the three decades since the large-scale MSR development program was undertaken, major changes have occurred.

- *Technology.* Advances in technology have partly or fully addressed several of the technical challenges that would be associated with full-scale development of an MSR. Technological advances have created the potential more capable MSRs, such as a very-high-temperature MSR for hydrogen production and other new missions.
- *Goals.* The original goal of the MSR was to serve as a breeder reactor that produces electricity. Current goals include hydrogen production and a variety of different fuel cycle missions including burning of actinides from other reactors.

- *Requirements.* The safety, environmental, and nonproliferation requirements for all power reactors have changed.

These changes imply that an MSR developed for today's market may have significantly different features than would have been required 30 years ago. This paper reports on studies of six technological challenges for a commercial MSR. The six areas were chosen because they have significant impact on commercial viability (economics and developmental cost) and are generally applicable to all MSRs. For specific missions, such as burning actinides from other reactors, other mission-specific technical challenges may exist. This paper reports on work completed to date and is not a comprehensive examination of all technical issues.

II. GENERAL DESCRIPTION OF THE MSR

In an MSR (Fig. 1), the molten fluoride salt with dissolved fissile, fertile, and fission isotopes flows

through a reactor core moderated by unclad graphite. In the core, fission occurs within the flowing fuel salt, which then flows into a primary heat exchanger, where the heat is transferred to a secondary molten-salt coolant. The fuel salt then flows back to the reactor core. The graphite-to-fuel ratio is adjusted to provide the optimal neutron balance, an epithermal neutron spectrum. In the preconceptual 1000-MW(e) designs developed in the early 1970s, the liquid fuel salt typically enters the reactor vessel at 565°C and exits at 705°C and ~1 atmosphere (coolant boiling point: ~1400°C). Volatile fission products (e.g., krypton and xenon) are continuously removed from the fuel salt. The secondary coolant loop with a liquid salt transfers the heat to the Brayton power cycle to produce electricity or a hydrogen production facility. The term *liquid salt* denotes a clean fluoride salt that does not contain fissile materials, fertile materials, or fission products.

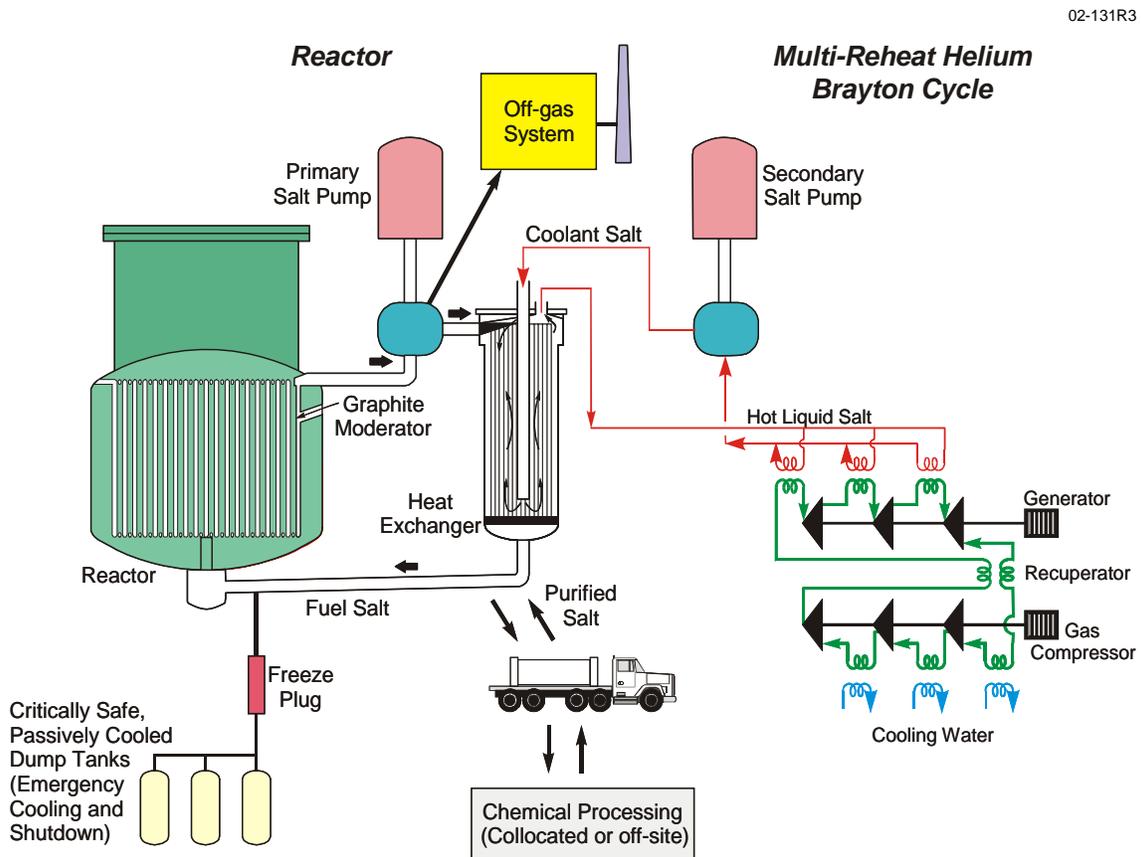


Fig. 1. MSR with multi-reheat helium Brayton cycle.

As a liquid-fuel reactor, the MSR has two sets of unique characteristics relative to solid-fuel reactors.

- *Safety.* Under emergency conditions, the liquid fuel is drained to passively cooled critically safe dump tanks. By the use of freeze valves (cooled sections of piping) and other techniques, this safety system can be passively initiated upon overheating of the coolant salt. MSRs operate at steady-state conditions, with no change in the nuclear reactivity of the fuel as a function of time. Last, the option exists to remove fission products online and then solidify those radionuclides into a stable waste form. This minimizes the radioactive inventory (accident source term) in the reactor core and potential accident consequences.
- *Fuel cycles.* The liquid fuel allows online refueling and a wide choice of fuel cycle options: burning of actinides from other reactors, a once-through fuel cycle, a thorium-²³³U breeder cycle, and a denatured thorium-²³³U breeder cycle. Some of the options, such as a thermal-neutron-spectrum thorium-²³³U breeder cycle require online refueling and thus can not be practically achieved using solid fuels. The use of a liquid fuel also avoids the need to develop fuel or fabricate fuel.

III. ANALYSIS OF TECHNOLOGY GAPS

A series of issues in the 1970s designs of MSRs were identified, analyzed, and evaluated in terms of technical solutions available today. Many² but not all of these issues were identified in evaluation and closeout reports of the MSR projects in the 1970s.

III.A. Power Cycles

When MSRs were being developed in the 1960s, the only demonstrated power cycle for the large-scale conversion of heat to electricity was the steam (Rankine) cycle; thus, early MSRs (and liquid metal fast reactors) were designed with steam power cycles. The coupling of an MSR with a steam cycle resulted in a series of technical challenges. Since that time, gas Brayton power cycles have been developed by the aircraft industry and are now widely used in the utility industry, with natural gas as the preferred fuel. Direct and indirect Brayton cycles are also being developed for various high-temperature reactors. Indirect multi-reheat nitrogen or helium Brayton^{4, 5}

cycles offer major economic and technical advantages relative to steam cycles for electricity production using MSRs.

- *Efficiency.* MSRs are naturally high-temperature reactors. Depending upon the choice of salt, the freezing points are between 320 and 500°C. The heat transfer properties (viscosity, thermal conductivity, etc.) improve rapidly with increasing temperature. Consequently, the detailed 1000-MW(e) conceptual design of the MSR had a reactor-core fuel-coolant exit temperature of 705°C. However, because of corrosion and other constraints in steam cycles, peak steam cycle temperatures are between 500 and 550°C. In the 1960s designs, high-temperature heat was inefficiently dumped to lower temperatures to match what the steam cycle could tolerate. This process reduces heat exchanger sizes but has a large penalty in terms of efficiency. In contrast, many Brayton cycles operate above 1000°C. The adoption of closed helium or nitrogen Brayton power cycles enables the power cycle to efficiently use the high-temperature heat generated by the MSR. *This capability allows a 15% improvement in electrical power output without changing the temperatures of the fuel salt exiting the reactor core.*
- *Freeze protection.* Salt coolants must be kept sufficiently hot to ensure good heat transport and avoid freezing of the molten fuel salt and the liquid salt in the intermediate heat-transport loop. With a steam cycle and the lower temperatures, special design features must be used so that feed water does not freeze the salt. With the higher-temperature Brayton cycle, freeze protection is greatly simplified.
- *Tritium control.* In an MSR, tritium is generated as a fission product and may be generated by coolant activation. Unlike solid-fuel reactors, the tritium in an MSR is highly mobile in the salt and tends to diffuse through the high-temperature heat exchangers into the working fluid of the power cycle. At the time the MSR program was cancelled, tritium control was considered the largest remaining engineering development challenge, because any tritium that entered the steam cycle resulted in tritiated water. Isotopically separating tritiated water from nontritiated water in the steam cycle is difficult and expensive. The MSR program partly developed the use of a fluoroborate

coolant salts in the secondary heat transfer system to trap the tritium. While technically workable, such systems are potentially complex and expensive for high levels of tritium trapping. Adoption of a Brayton cycle provides an alternative tritium trapping option where the tritium is removed from the helium in the Brayton power cycle. This is potentially a high-performance low-cost option based on demonstrated inexpensive methods to remove tritium gas or tritiated water from helium. Helium-cooled high-temperature reactors produce tritium from nuclear reactions with ^3He and from leaking fuel; consequently, these reactors are equipped with systems to remove the tritium from the helium.

- *Chemical reactions.* Molten salts do not chemically react with nitrogen or helium. However, these molten salts will slowly react with steam over time. The reaction rate is many orders of magnitude slower than for sodium and water. Changing from a steam cycle to a gas Brayton cycle eliminates this class of challenges.

Closed Brayton power cycles using helium are being developed for the modular high-temperature gas-cooled reactors (MHTGRs) for which helium is the coolant gas. A prototype helium-cooled MHTGR is being constructed in South Africa with a helium Brayton power cycle. Additional technology development would be required for an MSR; however, the closed Brayton cycle technology is transitioning to a commercial technology.

III.B. Fuel Inventory

In an MSR, the fuel salt circulates from the reactor core to the intermediate-loop heat exchangers and back to the reactor core. Heat is produced in the core and subsequently dumped in the heat exchangers. Thus, a significant fraction of the fuel salt is outside the reactor core in the heat exchangers. Historically, MSRs have been designed with various tube-and-shell heat exchangers. In the last decade, compact plate-fin and printed circuit high-temperature heat exchangers have been developed for the aircraft, chemical, and offshore-oil industries. The adoption of compact heat exchangers drastically reduces the molten fuel salt inventory in the heat exchangers and may reduce the inventory of fuel salt in the reactor by up to 50%. There are major benefits in using such heat exchangers.

- *Fuel salt inventory.* Reducing the fuel inventory reduces both fuel salt costs and nonproliferation risks, because the total fissile inventory in the nuclear system is decreased.
- *Fuel salt processing.* In an MSR, volatile fission products (including xenon) are removed continuously, which creates a large parasitic neutron sink in solid-fuel reactors. For nonvolatile fission products, the fuel salt is processed online or off-line, depending upon design goals. Reducing the salt inventory reduces the quantities of salt to be processed.
- *Heat exchanger size.* The size of the heat exchangers is reduced by a factor of 3 or more. This reduction has major economic implications because the primary heat exchangers have fuel salt flowing through them on one side and clean salt flowing through on the second side. The fuel salt, which contains the fission products and actinides, is highly radioactive. In an MSR, the reactor vessel and primary heat exchangers are located in a hot cell. Reducing the size of the heat exchanger significantly reduces the size of the hot cell, its support equipment, and the reactor building.
- *Tritium control.* The aircraft and other industries have developed compact heat exchangers with buffer gas zones to separate different fluids that may react explosively—such as hot gases vaporizing fuels in aircraft. The same technologies enable trapping of tritium from the primary system in the heat exchanger. While this may not be important for electricity production when using Brayton cycles that allow trapping of tritium, it is another option for tritium trapping if the MSR is used for hydrogen production where high temperature heat is required for the thermochemical hydrogen production cycles.

The advanced heat exchangers are commercial products used in industry. These heat exchangers are being considered for use in high-temperature helium-cooled reactors⁶ and in the transport of heat⁷ from high-temperature gas-cooled reactors to hydrogen production plants using liquid-fluoride-salt heat-transport systems. Additional work is required to fully evaluate their use in MSRs.

III.C. Noble Metal Plate-Out

In an MSR, fission products are generated in the molten salt. Most of the fission products form stable fluorides that dissolve in the salt. Noble and seminoble metals (e.g., Nb, Mo, Tc, Ru, Rh, Pd, Ag, etc.)³ form multi-atom clusters in the molten salt and ultimately plate out on metal surfaces such as those of heat exchangers or are vented to the off-gas system. The noble metals produce significant decay heat. If the plate-out is excessive, the decay heat from the noble metals may damage the heat exchangers via overheating should a loss of cooling occur. At the end of the MSR projects in the United States in the early 1970s, the assessments indicated that plate-out was not likely to be a major safety issue (potential for noble metals to escape to the environment). However, it had the potential to be a significant design and operational issue. In the last decade there have been major advances in this area.

- *Better understanding of the physical processes.* The R&D challenges are to understand the plate-out mechanisms and to test methods for removal of the metal atoms. A major problem has been the difficulty in generating a molten salt with noble metal atoms that can be used to study plate-out and removal mechanisms. Generating molten salts with noble metal atoms using a molten salt test reactor or an irradiation loop is extremely expensive and involves highly radioactive systems. Recently the French have successfully developed laboratory methods to generate molten salts with noble metal atoms in nonradioactive systems. This development should enable more rapid progress in understanding and development of technologies to remove noble metal atoms from molten salts.
- *New materials.* Plate-out depend upon the surface characteristics. Earlier work showed that noble metals preferentially plated out on metal surfaces relative to carbon surfaces. The potential use of carbon-carbon composite heat exchangers, rather than metal heat exchangers, may significantly reduce noble-metal plate-out on the heat exchangers—the thin-walled reactor component with most of the reactor surface area and most sensitive to decay heat when cooling systems are not operating. Slowing noble-metal plate-out in the primary system provides the time for the salt cleanup systems to remove a larger fraction of the noble metals from the salt.

- *Improved separation methods.* There are several potential methods to remove noble metal from molten salts. The molten salts can be purged with inert gases, with the noble metals preferentially forming aerosols that can be filtered from the gas stream. Newer options include ultra-high-surface-area metal or coated carbon-foam matrixes designed to preferentially encourage plate-out of noble metals in the salt cleanup system.

These developments may allow successful resolution of issues regarding noble metal plate-out issues within several years and help determine (1) whether a significant challenge exists and (2) what the preferred options are for control of noble metal fission products.

III.D. Fuel Storage

The use of MSRs will require the storage and transport of MSR fuel salts with and without uranium. The MSR projects in the 1960s did not identify any issues with long-term storage of the fuel salts; however, no long-term tests were conducted. Since that time, events⁸ have revealed challenges in the long-term storage of highly radioactive fuel salts in solid form. The MSRE, an 8-MW(t) test reactor, was shut down and placed in storage in 1969, with the fuel salt (including its uranium) dumped to drain tanks. The fuel salt was stored as a solid at ambient temperatures. In 1994, a gas sample taken from the MSRE off-gas system (which remained connected to the fuel and flush-salt drain tanks) showed the generation of fluorine from the fuel salt and the partial transport of uranium (in the form of UF₆) from the salt into the off-gas system.

Molten fuel salts in high radiation fields do not release fluorine, because the fuel salt is an ionic solution with very rapid recombination rates. However, it is now known⁹ that if the fluoride fuel salt is a *solid at a temperature significantly below its melting point*, radiation can cause the partial decomposition of the salt, release free fluorine, and result in the formation of UF₆. The 30-year storage of MSR fuel salt (an unintended large-scale long-term experiment in fuel-salt storage) and the subsequent remediation program now provide the basis to understand (1) what happens when frozen fuel salts are stored for multi-decade periods of time, (2) the requirements for safe long-term storage, and (3) alternative methods to ensure safe storage.

III.E. High-Level Waste (HLW) Form

In an MSR, the fuel is a fluoride salt. Ultimately, the fission products must be removed and disposed of as HLW, while the fissile materials are recycled. This process requires the chemical conversion of the fission products from a fluoride chemical form chosen for in-reactor operations to a repository-acceptable waste form. In the 1960s, no significant work was done to develop such an HLW form.

Since the 1960s, however, a variety of other nuclear processing facilities have generated fluoride waste forms. Laboratory studies have been conducted on how to produce high-quality waste forms from many of these fluoride waste streams. Radioactive fluoride waste streams have been generated from (1) processing of spent nuclear fuel (SNF) in the Idaho Chemical Processing Plant, (2) plutonium processing in the weapons complex, (3) development and use of fluoride volatility processing to recover uranium from SNF, and (4) molten salt processing.

Two approaches have been partly developed to convert fluoride waste forms to an acceptable form for repository disposal. Either approach potentially provides a basis for development of an MSR waste conversion and solidification process.

- *Conversion to nonfluoride waste forms.* Several processes have been partially developed to convert fluoride waste forms to traditional nonfluoride waste forms. The glass material oxidation and dissolution system (GMODS)^{3, 10} converts fluoride wastes into non-fluoride-containing borosilicate glass—the traditional HLW glass. Other processes produce phosphate waste forms.
- *Fluoride HLW form.* Several potential waste forms contain significant fluorides and may meet the requirements for a repository-acceptable waste form. Borosilicate glasses containing fluorides are a leading candidate for processing the fluoride HLW at the Idaho site¹¹; however, the viability of a fluoride-containing borosilicate glass is strongly dependent upon the chemical composition of the initial HLW form. Other researchers¹² are examining fluorapatites as a waste form. Examples of fluorapatites include $\text{Sr}_{10}(\text{PO}_4)_6\text{F}_2$ and $\text{Sr}_8\text{CsNd}(\text{PO}_4)_6\text{F}_{2.3}$.

A fully developed process to convert fluoride HLWs into repository-acceptable waste forms does not currently exist. However, several candidate processes and waste forms have been partly developed in the last 30 years.

III.F. Peak Reactor Temperature

The peak temperature of an MSR is limited by the materials of construction. The developmental work on MSRs resulted in the development of a modified Hastelloy-N, a high-nickel code-qualified alloy suitable for MSR service that allows peak temperatures to $\sim 750^\circ\text{C}$. In the longer term, higher temperatures are highly desirable to (1) improve efficiency in the production of electricity, (2) provide the high-temperature heat required for hydrogen production, and (3) allow the use of higher-melting-point fuel salts that may provide major fuel cycle advantages. While there are many higher-temperature alloy options for systems with clean fluoride salts, an MSR with dissolved uranium and other species presents special challenges. It is the uranium and certain fission products (not the fluoride salt itself) that primarily determine corrosion rates. In these systems, the corrosion rates are very low with the use of high nickel alloys; however, such alloys lose strength at higher temperatures.

Long-term experience shows carbon-based materials to be compatible with molten salts at temperatures of 1000°C . Short-term tests have shown graphite to be compatible with molten salts at temperatures to 1400°C . Carbon-carbon composites are presently being developed for many industrial applications (pumps, heat exchangers, etc.) and have already been developed for use in high-temperature reactors, particularly for in-core high-temperature applications (control rods, core support structures, etc.). Carbon-carbon composites¹³ are potentially an enabling technology for very high temperature MSRs. However, there are major technical uncertainties including joining technologies. If these uncertainties can be overcome, large-scale development work and demonstrations would be required before these materials can be considered for major safety-related components such as reactor vessels. This is a new long-term materials option that did not exist 30 years ago.

IV. CONCLUSIONS

MSRs were developed in the 1950s and 1960s. The large-scale R&D efforts yielded a workable reactor concept but a reactor with significant

operational and other challenges. In the last three decades, there have been major advances in technology. A technology gap analysis has identified potential solutions for many of the technological challenges that were identified in the 1970s and that may significantly lower the capital cost of the MSR. The commercial viability of the MSR has improved both in absolute terms and in comparison with other reactor concepts. However, significant work is required before definitive conclusions can be made about the economics, advantages, and disadvantages of the MSR relative to those of other advanced reactor concepts.

REFERENCES

1. *Nuclear Applications and Technology*, **8**, 2 (Entire issue), (February 1970).
2. U.S. ATOMIC ENERGY COMMISSION, *An Evaluation of the Molten Salt Breeder Reactor*, Wash-1222, Washington, D.C. (September 1972).
3. C. W. FORSBERG, *Molten Salt Reactor Critical Systems Evaluations: Control of Noble Metals, Acceptable Waste Forms, and Fuel-Salt Storage*, ORNL/GEN4/LTR-05-005, Oak Ridge National Laboratory, Oak Ridge, Tennessee (2006).
4. H. ZHAO and P. F. Peterson, *A Reference 2400 MW(t) Power Conversion System Point Design for Molten-Salt-Cooled Fission and Fusion Energy Systems*, UCBTH-03-002, University of California at Berkeley (2004).
5. P. F. PETERSON, "Multiple-Reheat Brayton Cycles for Nuclear Power Conversion with Molten Coolants," *Nuclear Technology*, **144**, 3, 279–288 (2003).
6. C. F. McDONALD, "Compact Buffer Zone Plate-Fin IHX—The Key Component for High-Temperature Nuclear Process Heat Realization with Advanced MHR," *Applied Thermal Engineering*, **16**, 1, 3–32 (1996).
7. W. E. KIRST, W. M. NAGLE, and J. B. CASTNER, "A New Heat Transfer Medium for High Temperatures," *Transactions of the American Institute of Chemical Engineers* **36**, 371–394 (1940).
8. G. D. DEL CUL, A. S. Icenhour, D. W. Simmons, L. D. Trowbridge, D. F. Williams, L. M. Toth, and S. Dai, "Overview of the Recovery and Processing of ^{233}U from the Oak Ridge Molten Salt Reactor Experiment (MSRE) Remediation Activities," *Proc. of Global 2001, Paris, France, September 9–13, 2001*.
9. L. M. TOTH and L. K. Felker, "Fluorine Generation by Gamma Radiolysis of a Fluoride Salt Mixture," *Radiation Effects and Defects in Solids*, **112**, 201–210 (1990).
10. C. W. FORSBERG, E. C. Beahm, and J. C. Rudolph, "Direct Conversion of Halogen-Containing Wastes to Borosilicate Glass," *Symp. Proc. Scientific Basis for Nuclear Waste Management XX, December 2–6, 1996, Boston, Massachusetts*, **465**, 131–137, Materials Research Society (1997).
11. K. VINJAMURI, *Candidate Glass-Ceramic Waste Forms for Immobilization of the Calcines Stored at the Idaho Chemical Processing Plants*, INEL-94/00026, Idaho National Engineering Laboratory, Idaho Falls, Idaho (1994).
12. O. HEMMERS and B. E. Burakov, Transmutation Research Program, Task 16: "Evaluation of Fluorapatite as a Waste-Form Material," <http://aaa.nevada.edu/task16.html>, (2005).
13. P. F. PETERSON, C. W. Forsberg, and P. Pickard, "Advanced CSiC Composites for High-Temperature Nuclear Heat Transport with Helium, Molten Salts, and Sulfur-Iodine Thermochemical Hydrogen Process Fluids," *Proc. of Second Information Exchange Meeting on Nuclear Production of Hydrogen, Argonne National Laboratory, Illinois, USA, October 2–3, 2003*.