Development Principles for Thermal-Spectrum Molten-Salt Breeder Reactors

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ABSTRACT

Thermal-spectrum molten-salt breeder reactors (MSBRs) have potentially highly advantageous characteristics. This paper provides guiding principles to increase the probability of realizing their potential in future deployments. The guiding principles range from specific technology recommendations, to means to minimize the impact of current nuclear supply chain issues, to approaches to decreasing the difficulty of safety adequacy assessment, to methods to minimize proliferation vulnerability. The most substantial overall recommendation, however, is to initiate a modern, thermal-spectrum MSBR development program.

KEYWORDS
molten-salt reactor, thermal-spectrum, breeder

1 BACKGROUND

Thermal-spectrum, molten-salt breeder reactors (MSBRs) have the potential for advantageous safety, fissile resource utilization, proliferation resistance, and exergy characteristics. They were one of the first reactor classes proposed due to their high potential. The United States pursued a thermal-spectrum MSBR technology development program from the late 1940s through the mid-1970s. While this historic program was remarkably successful at advancing MSBR science and technology, the program was much smaller than those for other reactor classes, even at its peak. Consequently, MSBR technological readiness remained significantly lower than other reactor classes in the 1970s when the United States consolidated its breeder reactor efforts to a single concept, the sodium-cooled fast reactor (SFR).

While the United States has not had a thermal-spectrum MSBR development program in nearly half a century, high-temperature technology has advanced markedly over the intervening decades. Moreover, the continuing use of nuclear power has provided important insights to guide future reactor deployment. In addition, minimizing the potential to misuse fissile materials and nuclear facilities has become a key consideration for all nuclear power plants.

The world currently needs enormous and ever-increasing quantities of energy as more of its population adopts an energy-intensive lifestyle. Without sufficient research and development, only today’s technologies can be deployed. Moreover, substantial nuclear power technology development is now going forward outside of the United States. Without sufficient, consistent technology development support from the U.S. government, the United States will lose its leadership in nuclear power.
2 INTRODUCTION

Historically, advanced nuclear power has not appeared sufficiently advantageous for the technology to break into the commercial sector from state-sponsored demonstrations. Reasonable follow-up questions are “Why not?” and “What has changed sufficiently that might result in widespread commercial deployment?” The dominant reason for the lack of progress has been the lack of need. Simply put, it has proven cheaper, safer, and faster to meet energy demands with incremental growth in established (largely fossil) technologies. Advanced reactor commercial deployments have had an overall poor performance (e.g., Fermi-1, Fort Saint Vrain, Superphénix), and even recent advanced light-water reactor (LWR) deployments have cost substantially more and taken much longer than originally planned.

Advanced nuclear power has enormous potential and can be implemented in extremely beneficial or harmful ways. The potential for great harm makes its public perception vulnerable to specious, apocryphal attacks. Thermal-spectrum MSBRs are especially vulnerable to these attacks as the historic record largely consists of MSBRs intended to produce as much separated fissile material as possible and whose safety characteristics represent a larger departure from well-known LWRs than other solid-fueled, advanced reactors.

Thermal-spectrum MSBRs should be included within the national reactor technology development portfolio as they have distinctive advantageous characteristics compared to other reactor types. Thermal-spectrum MSBRs with desirable safety, nonproliferation, and economic characteristics are not yet ready for deployment but could rapidly (within a few years) become so with consistent support. Advocacy for thermal-spectrum MSBRs should, however, not be interpreted as disparaging the pursuit of other advanced reactor concepts—in particular other molten-salt reactors (MSRs). Broad spectrum advanced reactor technology development provides the highest probability of achieving national energy goals.

This paper outlines the guiding development principles for successful, future thermal-spectrum MSBRs but does not describe any specific concept. The paper is organized as a structured description of recommended principles to be considered in the development and deployment of thermal-spectrum MSBRs guided by the technology, regulations, and nuclear supply chain statuses.

3 GUIDING PRINCIPLES

3.1 Minimize the Potential to Misuse Fissile Materials and the Facility

3.1.1 Minimize the potential to misuse uranium

Fissile materials are required to achieve and maintain criticality. However, some forms require less effort to misuse than others. The International Atomic Energy Agency (IAEA) provides guidance on how difficult various materials are to divert for nonpeaceful uses. The IAEA Safeguards Glossary [1] indicates that isotopic mixtures of uranium containing less than 20% of $^{233}$U and $^{235}$U have the highest conversion time. The United States does not currently have a definition of low-enrichment uranium (LEU) that includes any $^{233}$U, and 10 CFR 110 Appendix M (same as IAEA INFCIRC/225/Revision 5) currently only acknowledges the potential to decrease the material category of materials containing $^{235}$U and $^{239}$Pu through isotopic dilution (e.g., by including greater than 80% $^{238}$U or $^{238}$Pu, respectively). Nevertheless, a key concept to decrease the potential to misuse fissile materials in thermal-spectrum MSBRs is to keep all uranium from initial creation through consumption within a low-enrichment environment. To achieve the fissile isotope concentration objective, $^{233}$Pa, the precursor to $^{233}$U, can never be separated from uranium and the plant facilities should not include technologies that can readily separate uranium from the other trivalent actinides (e.g., no fluorination processing).
3.1.2 Minimize the potential to misuse plutonium

Exposing $^{238}\text{U}$ to neutrons will generate fissile $^{239}\text{Pu}$ via absorption. Thermal-spectrum MSBRs that employ $^{238}\text{U}$ to denature their fuel salt have three primary means to minimize the potential to divert $^{239}\text{Pu}$:

- Minimize production;
- Maximize consumption;
- Keep all plutonium mixed with other highly radioactive, nonfissile materials (do not have onsite technology capable of separating the plutonium).

One means to minimize $^{239}\text{Pu}$ production is to minimize the $^{238}\text{U}$ precursor content in the fuel salt. However, nonfissile $^{238}\text{U}$ is necessary to maintain the uranium in the fuel as LEU. The overall uranium content requirement for the fuel salt is driven by the need to maintain criticality. Thermal-spectrum MSBRs will employ an optimized core configuration with the minimum necessary fissile material content and thereby the minimum necessary $^{238}\text{U}$ content.

Another means to minimize $^{239}\text{Pu}$ production is to maximize the concentration of isotopes (in this case, $^{232}\text{Th}$) with competing neutron absorption cross sections. In the energy range of interest (a few hundred meV), the absorption cross section of $^{232}\text{Th}$ is roughly twice that of $^{238}\text{U}$. Fuel salt can maintain a low melting point with a substantial ThF$_4$ content. An optimally moderated thermal-spectrum MSBR would only require roughly 1 mole % LEU in its fuel salt to maintain criticality. The large MSBR concept in the historic program intended to employ a 71.7-16-12-0.3 mole% 7LiF-BeF$_2$-ThF$_4$-UF$_4$ fuel salt composition [2]. However, nearly all its uranium would have been $^{233}\text{U}$. Neutron absorption in $^{238}\text{U}$ is somewhat parasitic at MSR-temperature thermal energies as the $^{239}\text{Pu}$ neutron yield per absorption is less than two for thermalized, high-temperature neutrons.

One method to maximize $^{239}\text{Pu}$ consumption is to tailor the reactor’s thermal neutron spectrum peak to match $^{239}\text{Pu}$’s thermal fission cross-section peak near 300 meV (see Figure 1). Fully thermalized neutrons at MSBR operating temperature would be near 90 meV. Slightly undermoderating the core (e.g., via decreasing the moderator to fuel salt ratio) would maximize the $^{239}\text{Pu}$ consumption.
The plutonium needs to be kept with other highly radioactive materials to maximize the difficulty for its misuse. The relatively large $^{233}$Pa neutron capture cross-section results in a necessary decay in a low-neutron flux environment to achieve breeding gain. The plutonium would be co-removed from the reactor’s high-flux environment along with $^{233}$Pa and other actinides to avoid generating separated $^{233}$U or $^{239}$Pu. Once the $^{233}$Pa has decayed into $^{233}$U (over a few months; 27-day half-life), the mixed actinides would be reintroduced to the core.

3.2 Avoid Dependency on High-Assay, Low-Enrichment Uranium

High-assay, low-enriched uranium (HALEU) is currently in extremely limited supply in the United States. Not requiring HALEU feedstock is, consequently, necessary for practical near-term deployment. Thermal-spectrum MSBRs can start-up effectively on LEU and only require natural enrichment uranium and thorium as equilibrium actinide feedstocks. Note that, without employing HALEU for initial start-up, a combination of larger amounts of LEU and smaller amounts of thorium will be necessary in the start-up fuel salt to achieve the initial criticality. As the $^{233}$U in the fuel salt progressively builds in, larger amounts of thorium can be introduced into the fuel salt and the uranium loading can decrease.

3.3 Minimize Fuel Qualification Time and Effort

Fuel qualification is a key element of overall reactor safety and is based on developing an adequate understanding of the fuel characteristics under both normal and accident conditions to enable conservatively modeling the role of the fuel in overall facility safety. Liquids cannot be mechanically damaged, do not have a long-range structure, and do not exhibit mechanical history. Consequently, modeling the fuel salt properties under both normal and accident conditions derives from understanding
the thermochemical and thermophysical properties of the salt as a function of temperature and composition. The Department of Energy Office of Nuclear Energy MSR campaign is currently engaged in populating and validating molten halide salt thermophysical and thermochemical property databases. Validated fuel salt property data can then be employed to assess the achievement of the fundamental safety functions under design basis and beyond design basis accident conditions as well as the potential to damage safety-related systems, structures, or components (SSCs) during normal operations or anticipated operational occurrences. The Nuclear Regulatory Commission has recently accepted a fuel salt property and fundamental safety function based fuel salt qualification methodology [3].

3.4 Avoid Bismuth in Systems that are Hydraulically Connected to Engineering Alloys.

The historic MSBR development program planned to employ bismuth as its reducing agent to separate trivalent actinides from fuel salt. However, bismuth is incompatible with MSR engineering alloys. Specifically, bismuth dissolves nickel at temperatures where the salt is liquid. Employing a bismuth-based reductive separation system would substantially increase the cost and technical difficulty of separating actinides from the fuel salt. The historic MSBR program expended significant resources developing nondispersive contactors and structural materials compatible with both liquid bismuth and fluoride salts at elevated temperatures [4]. Aluminum is a promising reducing agent within fluoride salt that can separate all the trivalent actinides (e.g., largely leaving thorium) from the fuel salt [5]. The protactinium, uranium, plutonium, americium, and curium all thermodynamically separate into the metallic phase (without needing an electrical bias). The protactinium within the actinide aluminum alloy would then be allowed to decay into uranium away from the core prior to the actinides being converted into fluorides and reintroduced to the active fuel salt.

3.5 Minimize Radiation Damage to the Reactor Vessel and Containment Radiation Levels

Radiation damage to the reactor vessel has the potential to significantly decrease its useful lifetime, resulting in substantial replacement downtime, maintenance expense, and increased radioactive waste generation. One of the lessons learned from the historic MSBR program is to include substantial amounts of neutron reflectors and shielding between the active core and its container [6,7]. The shielding can be provided by solid material akin to high-temperature gas-cooled reactors or the liquid downcomers in LWRs or SFRs. Keeping the fuel salt within externally cooled tubes has been repeatedly proposed [8] (e.g., in the MOSEL reactor [9] and the Aircraft Reactor Experiment with stagnant fuel [10]). An additional advantage of providing substantial shielding within the reactor vessel is sufficiently decreasing the containment radiation levels to be able to employ more conventional cabling and instrumentation. Decreasing the radiation dose within containment would also substantially simplify maintenance.

3.6 Simplify the Plant Configuration

3.6.1 Employ an integral primary system layout

The general configuration of the Molten Salt Reactor Experiment and planned large MSBR was derived from the technology available at the time and their functional objectives. The design choice of employing an ex-vessel pumped fuel salt loop was derived from the technology available at the time. The containment radiation levels and fuel salt spill potential that result from the external pumped loop configuration are significantly disadvantageous. Integral primary systems have been selected for other modern reactor concepts (e.g., pool-type SFRs and integral pressurized-water reactors) for similar
reasons. Modern designs will keep the fuel salt within the shielding, within the reactor vessel. Integral
designs also simplify implementing natural-circulation-based decay heat removal.

3.6.2 Avoid introducing additional materials into the primary system

Introducing another fluid into the fuel salt to disengage the fission gases substantially increases the gas
volume, effectively precluding retaining the fission gases in the reactor vessel for the first couple of days
(to decay sufficiently to avoid needing safety-related cooling). The spray ring and helium sparging $^{135}$Xe
disengagement systems employed at the Molten Salt Reactor Experiment and planned for future
deployment were technology-availability-based choices. An ultrasonic gas disengagement system, a spray
ring system, or tray-type gas disengagement system into a vacuum environment would avoid introducing
additional material into the primary circuit. Retaining the fission gases within the reactor vessel for the
first few days would substantially simplify the off-gas system design and its safety requirements.

3.6.3 Avoid freeze valves

The ability to rapidly remove fuel from the primary (critical) circuit is an advantageous characteristic of
liquid fuel. A bottom-located freeze valve would enable liquid fuel salt to be passively drained by gravity
in the event of a loss-of-power accident. Molten-salt freeze valves, however, tend to be slow to operate
and have proven to be technically difficult. A simpler, passive fuel salt removal technique would be to
employ a gooseneck-type pipe connection below the reactor vessel and differential gas pressure above
and below the fuel salt as a control mechanism. The gas pressure necessary to blow out the fuel salt could
be retained in an accumulator with an electrically closed valve. The accumulator and valve would be
positioned in inert gas outside of the high-flux region. Similar fuel removal methods have previously been
employed for aqueous homogeneous reactors. Also, gas accumulators are currently employed to drive
control blades into boiling-water reactors and so have well-known safety-performance characteristics.

3.7 Avoid Generating Actinide Wastes

Nuclear fuel becomes waste when it can no longer adequately perform its functions. Liquids cannot be
mechanically damaged. The ability of fuel salt to perform its nuclear fuel and heat transfer functions
derives from its chemical and isotopic composition. MSBR fuel salt would be chemically processed as
part of normal operations. Fissile and fertile materials would be added or removed as needed, whereas
(nonactinide) parasitic neutron absorbers and elements that adversely impact its heat transfer properties
would be removed from the fuel salt. Once the actinides have been co-separated from the fuel salt (to
allow for $^{233}$Pa decay), multiple different neutron absorber separation technologies become possible such
as vacuum distillation, oxidative precipitation, or zone refining. Actinide neutron absorbers would
gradually build up to equilibrium concentrations (well below solubility limits). No materials bearing
significant quantities of actinides would become waste until the end of the reactor class.

3.8 Plan to Control Tritium

Thermal-spectrum MSBRs will produce substantial quantities of tritium due to neutron interactions with
the lithium and beryllium (~1 Ci/day-MWth) [11]. Fast-spectrum and thermal-spectrum MSRs without
lithium or beryllium near their critical region will produce substantially less tritium. At an operating
temperature, tritium readily permeates through structural materials [12]. The largest pathway for tritium
escape is through the thin walls of the primary heat exchanger. Blocking, trapping, and stripping are all
potentially useful mechanisms to limit tritium escape. The selected tritium management mechanisms need
to be integrated into the plant design from the outset as they can have a significant impact on the overall layout.

3.9 Improved Materials can Increase Performance

The fuel salt boundary material needs to withstand high temperatures and substantial neutron fluxes as well as being chemically compatible with the fuel salt. While proven engineering alloys such as 316SS can provide an acceptable performance, the combination of its limited strength at high temperatures and parasitic neutron absorption provides the incentive to develop improved materials. Additionally, the radiation damage vulnerability of graphite combined with the higher power density made possible by liquid cooling provides the incentive to develop improved moderator materials. For designs with fuel salt in tubes, the moderator would only need to be chemically compatible with the coolant, increasing the set of potential moderator materials. Beryllium carbide has the potential to be an advantageous moderator within unfueled FLiBe as it will preferentially react with oxygen contamination within the salt, forming beryllium oxide at its surface. Beryllium carbide is vulnerable to hydrolysis ($\text{Be}_2\text{C} + 4\text{H}_2\text{O} \rightarrow 2\text{Be(OH)}_2 + \text{CH}_4$) [13]. While the hydrogen (tritium production) and oxygen (contamination) sources are distinct, the beryllium carbide has the significant potential to serve as a tritium trap within FLiBe.

3.10 Minimize Safety Significance of Specialized SSCs

Much of the cost of any nuclear plant’s SSCs is in their quality assurance. The need for nuclear reactor level quality assurance derives from the SSC safety significance. SSCs that are not credited to achieve safety functions under accident conditions do not require nuclear-grade quality assurance. For example, if the results of a brittle failure of the normally salt-wetted container can be accommodated by exterior containment layers (i.e., a non-normally wetted guard vessel), the normally salt-wetted layer would not require nuclear-grade quality assurance. A non-normally salt-wetted vessel would only need to perform its safety functions for the duration of an accident and would likely be much less costly (e.g., a 304SS guard vessel appropriate to contain low-pressure molten salt for a limited period would be commercially available).

3.11 Leverage Related Technology Developments

The Department of Energy Office of Science has been sponsoring the development of lithium isotope separation technologies as part of its isotope program. Electrochemical deposition employing the electrochemical isotope effect can achieve a substantially higher separation coefficient than previous technologies; a separation coefficient of 1.2 was achieved with a nickel-polypropylene carbonate system [14]. The innovative technology appears to be readily scalable. Consequently, the cost of isotopically separated lithium is anticipated to be substantially reduced as increased demand drives supply chain development.

3.12 Leverage Safety Characteristics to Minimize Regulatory Costs

Reasonably designed, constructed, operated, and maintained MSRs lack the credible potential for accidents that breach containment. Adequate external shielding (such as that provided by shallow below grade deployment or an external berm) can provide reasonable protection from external hazards. A containment-focused safety adequacy evaluation while subject to a maximum hypothetical accident was
the key element of a safety adequacy assessment for both early commercial reactors and current nonpower reactors. The prescriptive requirements of 10 CFR 50 Appendix A were developed as it became apparent that credible accidents at sufficiently large, water-cooled reactors could not be adequately contained. Employing a modernized containment-focused safety adequacy evaluation methodology akin to the lines-of-defense methodology currently employed in France [15] offers the potential for a substantial simplification of the safety adequacy evaluation.

4 CONCLUSIONS AND RECOMMENDATIONS

Two primary perceptions have prevented U.S. government investment in thermal-spectrum MSBRs for the past several decades. First, thermal-spectrum MSBRs are sufficiently technologically immature and difficult as to require extensive, long-term development to realize their benefits, and second, thermal-spectrum MSBRs increase the potential to misuse fissile materials relative to other nuclear power options. Both these perceptions are incorrect. High-temperature technologies have advanced markedly over the intervening decades, and reasonable development pathways for all remaining technology hurdles have now been identified. Moreover, the combination of the lack of dependence on the availability of HALEU and lack of need for solid fuel qualification and fabrication technology development is anticipated to substantially shorten the time to deployment. Moreover, a blended Th-U and U-Pu fuel cycle that enables thermal-spectrum breeding without creating unacceptably attractive material while also avoiding creating an actinide waste stream has also been proposed. Simply put, the historic rationale for not pursuing thermal-spectrum MSBR development is no longer valid. Creating a modern, thermal-spectrum MSBR development program is recommended.

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REFERENCES


