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Re-evaluating the Thermal-Spectrum Molten-Salt Breeder Reactor Fuel Cycle in a Modern Context

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Changing the World's Energy Future

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Introduction

The current need to rapidly and substantially expand clean power production combined with limited capability for domestic uranium enrichment results in a U.S. nuclear power production planning environment similar to that of the first nuclear era. The U.S. thermal-spectrum molten-salt breeder reactor (TS-MSBR) program emerged during the first nuclear era as means to support rapid nuclear power expansion while minimizing the demand on fissile resources. The primary reasons that the TS-MSBR program was discontinued in the 1970s were the comparatively large amount of required effort to mature the technology sufficiently for commercial deployment and the access to separated fissile material provided by the integrated fuel cycle design. The slowdown in U.S. electrical load growth after 1980 and the abundance of inexpensive fossil fuels inhibited the reconsideration of TS-MSBR development for decades. The purpose of this discussion is to reexamine the value and technical challenges of TS-MSBRs with integrated fuel-salt chemical processing considering the renewed need for the rapid expansion of clean energy production worldwide, the current state of technology, the challenge of actinide wastes, and the continuing need to maintain a high degree of proliferation resistance and integrate safeguards into the design.

Costs, Benefits, and Risks

Costs, benefits, and risks all need to be considered when evaluating future activities in nuclear technology development. The current, pressing need for large quantities of clean power is the primary driver behind advanced reactor technology development. One differentiating benefit of TS-MSBRs from other advanced reactors is that they will not require high-assay, low-enrichment uranium (HALEU) or uranium enrichment at all once criticality has been established. Additionally, molten-salt reactors have the highest exergy (or the ability to do work) of any reactor class enabling them to efficiently support transitioning a broader set of industrial processes to clean energy. TS-MSBRs have advantageous safety attributes such as low-pressure, a chemically non-reactive fuel/coolant, effective passive decay heat removal, and the tolerance of large reactivity excursions without damage, which can be leveraged to decrease their cost. Moreover, TS-MSBRs with integrated fuel-salt chemical processing would not produce an actinide waste stream due to their indefinite fuel-salt lifetime.

TS-MSBRs have multiple, high-value potential benefits. Hence, the primary hurdles to investment in TS-MSBRs are cost and risks. The decision to terminate the historic U.S. TS-MSBR program was primarily based upon the predicted government costs to mature the technology. Today, reactor technology development would be a private-public partnership with focused government resources used primarily to spur private sector investment. Moreover, significant technology development has occurred in related fields over the intervening decades necessitating the reevaluation of technology maturity and development costs. However, TS-MSBRs rely upon sufficiently distinctive technologies (e.g., integrated fuel-salt chemical processing) to require focused development, which has not yet occurred. The lack of focused, government-sponsored TS-MSBR development, with its implied endorsement of the reactor class, combined with the abrupt cancellation of the historic program remains a key impediment to private sector investment.

A baseline requirement for any nuclear fuel cycle today is that it should not increase the potential for non-peaceful uses of fissile materials. However, the fuel cycle design decisions of the historic U.S. TS-MSBR program largely predated the Non-Proliferation Treaty of 1968. The then planned fuel cycle included several steps with direct access to fissile materials [1]. The modern redesign of the TS-MBSR fuel cycle would avoid generating separated fissile material that would not meet the International Atomic Energy Agency (IAEA) requirements for the longest conversion time to non-peaceful uses [2]. Additionally, all steps within the fuel processing cycle require the timely ability to monitor that their fissile and fertile materials have not been diverted from peaceful uses.

Modern Fuel Cycle Technologies

Integrating fuel-salt processing with reactor operations is necessary to achieve significant breeding gain with thermal-spectrum neutrons. Hence, reactor and fuel-salt processing technology development requirements overlap. Only ^{233}U has sufficient fission neutron yield at thermal energies to result in significant breeding gain. Thus, the primary fissile material in a TS-MSBR must be ^{233}U. The ^{233}U would be produced via neutron transmutation of ^{232}Th. However, pure ^{233}U is an unacceptably attractive material. Uranium can be denatured (meet the IAEA requirements for the longest conversion time to non-peaceful uses) by mixing with at least 80% of a non-fissile isotope (e.g., ^{238}U). As ^{233}U is not available in nature, initial criticality would be achieved by initially including 5 w% ^{235}U (while always maintaining > 80 w% ^{238}U).

The central challenge to achieve breeding gain is that ^{232}Th requires exposure to neutrons to initiate the transmutation chain into ^{233}U, yet the intermediate product, ^{233}Pa, has a roughly 27-day half-life and a sufficiently high neutron absorption cross section that it cannot be exposed to a significant neutron fluence. Consequently, the ^{233}Pa needs to be separated from the ^{232}Th, so it can be selectively removed from the neutron flux.

Integrated Separations

One method to separate ^{232}Th from ^{233}Pa, without producing separated ^{233}U, is to co-separate the trivalent actinides from the remainder of the fuel salt; ^{232}Th does not have a trivalent state. The understanding of the process thermodynamics for separations within fluoride salts has advanced markedly since the historic TS-MSBR program. In particular, the French fuel

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[1] McWherter, J. H. 1970. *Molten Salt Breeder Experiment Design Bases*. ORNL-TM-3177. Oak Ridge, TN: Oak Ridge National Laboratory. doi.org/10.2172/4104902.

[2] International Atomic Energy Agency (IAEA). 2022. *IAEA Safeguards Glossary*, 2022 Ed., International Nuclear Verification Series No. 3 (Rev 1), Table 2 – Estimated Material Conversion Times for Finished Pu or U Metal Components. Vienna: IAEA. https://www-pub.iaea.org/MTCD/publications/PDF/PUB2003_web.pdf.

processing program demonstrated an aluminum-based process to selectively strip trivalent actinides from fluoride salts [3][4]. While still at an early development stage, the aluminum process is more thermodynamically favorable than the bismuth-based reductive extraction process planned by the historic TS-MSBR program and much more compatible with engineering alloys facilitating the integration of the separation process with reactor operation. Maturation of actinide separation processes needs to occur in a secure (likely government) environment.

The ^{135}Xe, including its ^{135m}Xe metastable state, is also important to separate from the active fuel salt to maximize the reactor's breeding ratio. Xenon has very low solubility in fluoride salts, so physical separation methods can effectively separate gaseous xenon from molten fuel salt. The historic TS-MSBR program included methods to strip both ^{135}Xe and its ^{135}I precursor from the fuel salt because significant quantities of ^{135}Xe would have been trapped in graphite pores [5] in historic designs. Designs in which the moderation is external to the fuel-salt boundary significantly reduce the in-core trapping potential, which can obviate the need for chemical iodine stripping.

Container Materials

The structural alloy developed within the historic TS-MSBR program was vulnerable to corrosion and neutron embrittlement. The planned bismuth-based reductive extraction technology for ^{233}Pa would have required an entirely separate material system for its container as bismuth rapidly dissolves nickel out of engineering alloys. Developing and validating bismuth resistant materials and non-dispersive contactors for the reductive extraction system were uncompleted technology elements of the historic TS-MSBR program.

The historic TS-MSBR program developed chemical redox control methods, which significantly reduced fuel-salt corrosion, and was in the process of validating the performance of a container alloy which would have been much less susceptible to radiation embrittlement. Additionally, one of the lessons learned through the historic experimental program was to include substantial shielding between the fission process and the reactor vessel to reduce radiation damage and in-containment doses. One possible shielding approach (employed by nearly all other liquid-cooled reactors) would be to include a substantial thickness of unfueled liquid-coolant between the core and the reactor vessel. In this configuration, the fuel-salt container material would have similar performance requirements as the cladding employed for solid fuel.

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[3] Conocar, O., N. Douyere, J.-P. Glatz, J. Lacquement, R. Malmbeck, and J. Serp. 2006. "Promising Pyrochemical Actinide/Lanthanide Separation Processes Using Aluminum." *Nuclear Science and Engineering* 153(3): 253-261. doi.org/10.13182/NSE06-A2611.

[4] Conocar, O., N. Douyere, and J. Lacquerment. 2005. "Extraction Behavior of Actinides and Lanthanides in a Molten Fluoride/Liquid Aluminum System." *Journal of Nuclear Materials* 344: 136-141. doi.org/10.1016/j.jnucmat.2005.04.031.

[5] Engel, J. R. and R. C. Steffy. 1971. *Xenon Behavior in the Molten Salt Reactor Experiment*. ORNL-TM-3464. Oak Ridge, TN: Oak Ridge National Laboratory. doi.org/10.2172/4731186.

The loop plant configuration (e.g., with the primary heat exchanger and pump located outside of the reactor vessel) planned by the historic TS-MSBR program was an artifact of the available materials and components. The major issues driving in-core material development are fuel-salt corrosion, radiation damage, and parasitic neutron absorption. The recent confirmation of the usefulness of cathodic protection in fluoride salts indicates that sufficiently thin metallic tubing, which would avoid unacceptably large neutron absorption, may be possible [6]. Also, radiation damage tolerant ceramic composite tubing (e.g., SiC-SiC composite cladding [7]) had yet to be developed at the time of the historic TS-MSBR program.

Wastes

Fuel salt is indefinitely reusable as its chemical composition can be maintained through refueling and separations processes. U_{nat} and Th are the equilibrium actinide feedstocks, while the heavier actinides build up to equilibrium concentrations. Once the trivalent actinides have been separated (via reductive extraction into an aluminum alloy), a wide potential set of processes is available to strip fission and corrosion products as well as contaminants from barren fuel salt. Melt recrystallization and vacuum distillation are leading candidate processes that exploit the comparatively low melting point of FLiBe ($2LiF-BeF_2$) carrier salt.

The noble fission gases have very low solubility in the fuel salt. Consequently, they along with volatile compounds, aerosols, and a mist of insoluble fission products will inherently separate from the fuel salt. The off-gas waste stream will contain a substantial amount of decay energy for the first hour or two (1–2% of full power). Consequently, the off-gas stream is likely to be kept within the reactor vessel for the first couple of hours to minimize the requirements for separate safety-grade decay heat removal. Solid decay products as well as mists and aerosols are likely to deposit on the first solid surface that they impact. Electrostatic precipitation would remove suspended materials from the noble gases [8]. Both non-radioactive krypton and xenon as well as longer-lived ^{85}Kr and ^{133}Xe are likely to be the primary constituents of the gas stream emerging from the reactor vessel. A zeolite bed is likely to be employed as the next stage in the off-gas system to permit somewhat longer-lived radionuclides to decay prior to bottling the remaining mixture of ^{85}Kr and stable noble gases. The limited amount of ex-vessel off-gas decay heat rejection needed can be provided by a variety of passive systems.

The fission and activation products separated from the fuel salt and/or deposited within the off-gas system will constitute a highly radioactive byproduct stream. The most likely stabilization method for the byproduct materials is mixing into grout to convert them to a

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6 Sankar, K. M. and P. M. Singh. 2023. "Viability of Cathodic Protection for Preventing Corrosion of Stainless Steel 316H in Molten LiF-NaF-KF." *Corrosion* 79, no. 7: 809-814. doi.org/10.5006/4265.

7 Braun, J., C. Sauder, J. Lamon, and F. Balbaud-Célérier. 2019. "Influence of an original manufacturing process on the properties and microstructure of SiC/SiC tubular composites." *Composites Part A: Applied Science and Manufacturing* 123: 170-179. doi.org/10.1016/j.compositesa.2019.04.031.

8 Mizuno, A. 2000. "Electrostatic precipitation," *IEEE Transactions on Dielectrics and Electrical Insulation* 7, no. 5: 615-624. doi.org/10.1109/94.879357.

robust cementitious form [9]. The resulting grout could either be allowed to set into blocks for near-surface storage or injected into deep fractured rocks [10].

Future Activities Recommendation

TS-MSBRs have multiple areas in which their chemical science and engineering require maturation. Recommended near-term development tasks include:

- Integrate TS-MSBR fuel cycle chemistry and reactor operational modeling planning and development. TS-MSBRs and their fuel cycles are much more deeply intertwined than other reactors and should not be evaluated in isolation. For example, a tubed liquid-salt core could be segmented to enable load following and optimize fuel-salt chemical processing system sizing and scheduling.
- Perform an integrated demonstration of fuel-salt separation technology. The leading recommendation from the historic independent review of the TS-MSBR program was to perform “proof testing of an integrated reprocessing system” [11]. The recommendation remains valid more than 50 years later.
- Develop and validate the performance of fuel-salt compatible structural ceramic composites. The structural ceramic composites developed for other reactor classes provide a starting point for TS-MBSRs. However, the uranium within the fuel salt has a strong affinity for carbon while fluorides dissolve both oxides and many common ceramic binder materials (e.g., silicon).
- Develop and demonstrate processing methods for converting TS-MSBR byproduct materials into cementitious waste.
- Integrate proliferation resistance into the chemical fuel processing steps of the TS-MSBR fuel cycle. For example, the ^{238}Pu content of the fuel salt would be increased by co-separating the ^{237}U bred from the initial fuel-salt loading of ^{235}U (^{235}U^{236}U$\rightarrow$^{237}U) from the high neutron flux along with the ^{233}Pa; ^{237}U beta decays to ^{237}Np with a 6.7 d half-life and a large thermal neutron absorption cross section. Neutron capture in ^{237}Np yields ^{238}Pu.

Summary

TS-MSBRs have extremely high potential to safely and cost effectively provide large quantities of energy without generating actinide wastes, increasing the potential to misuse fissile materials, requiring access to HALEU, or requiring continuing uranium enrichment. The lack of any governmental investment in TS-MSBR specialized fuel cycle technologies for nearly half a century remains their most significant impediment to maturation. A sustained, focused development program to mature their fuel cycle technologies is needed to trigger sufficient

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[9] Batchelor, B. 2006. "Overview of waste stabilization with cement." *Waste Management* 26, no. 7: 689-698. doi.org/10.1016/j.wasman.2006.01.020.

[10] Weeren, H. O. 1976. *Evaluation of Waste Disposal by Shale Fracturing*. ORNL-TM-5209, Oak Ridge, TN: Oak Ridge National Laboratory. doi.org/10.2172/4078703.

[11] U.S. Atomic Energy Commission. 1972. “An Evaluation of The Molten Salt Breeder Reactor.” WASH-1222. Germantown, MD: U.S. Atomic Energy Commission. doi.org/10.2172/4372873.

private sector investment to commercialize the technology.