Molten Salt Reactors: Overview and Comparison of Uranium and Thorium Fuel Cycles

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1 Molten Salt Reactor Design Principles

A Molten Salt Reactor (MSR) is class of nuclear fission reactors that contain either a liquid salt coolant, a liquid salt coolant-fuel mixture, or a two-fluid blanket and fuel arrangement. The two-fluid breeder designs include a liquid fissile fuel like uranium-235 as well as a liquid blanket of fertile material like thorium-232. The fuel and blanket compounds are dissolved in liquid salts such as FLiBe (Li_2BeF_4). MSRs can operate in the fast, thermal, or epithermal neutron spectra, and can be setup to breed or simply burn fuel. Thermal reactor designs are typically moderated using graphite. A variety of fuels can be used including low-enriched uranium-235, thorium-232 with uranium-233, and waste products from light-water reactors (LWRs). Some proposed commercial designs so far include Seaborg Technologies' uranium-based Compact Molten Salt Reactor (CMSR) and Flibe Energy's Liquid Fluoride Thorium Reactor (LFTR), a two-fluid thermal spectrum breeder reactor.

The Liquid Metal-cooled Fast Reactor (LMFR), while using solid fuel, is related to the MSR. The LMFR is much more extensively tested and has been deployed for commercial power production. The Russian BN-800 reactor, a liquid sodium-cooled fast breeder reactor that uses solid uranium- and plutonium-nitride fuel, went into service in 2016. [9] LMFRs are not discussed here.

1.1 Uranium and Thorium Fuel Cycles

MSRs can be run with various types of fuel, but the most common fuel types are low-enriched uranium and thorium-232 with uranium-233. Uranium-235 is already fissile, whereas thorium-232 is bred into uranium-233 which can then fission to produce energy. This breeding process is described in Figure 2. Uranium-235 and uranium-233 both produce about 2.4 neutrons per fission. However, the thorium fuel cycle requires breeding and fissioning, halving the maximum neutron multiplication factor to

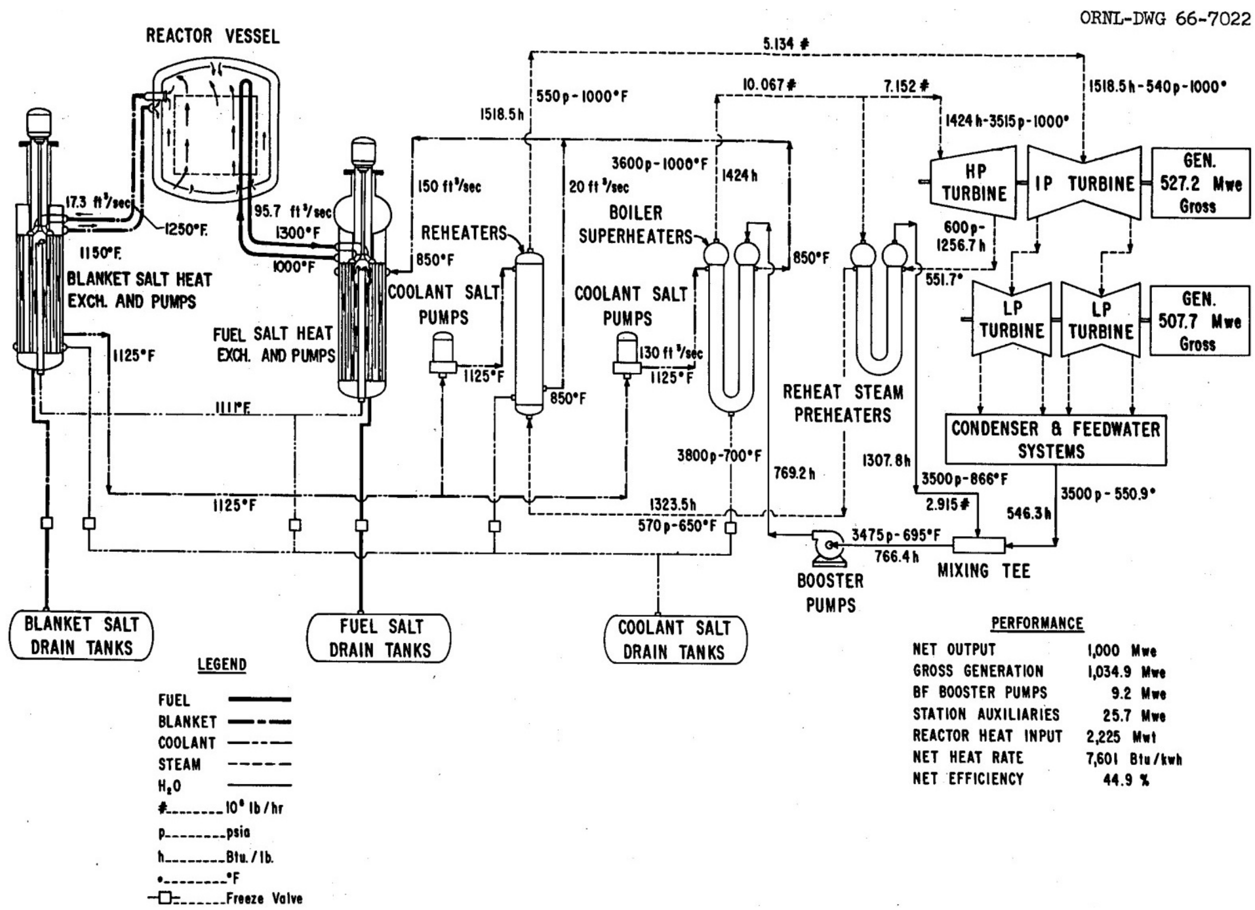

Figure 1: A 1GWe two-fluid molten salt breeder reactor design developed by Oak Ridge National Laboratories in the 1960s. [6]

approximately 1.2. This makes criticality harder to maintain with the thorium fuel cycle. Additionally, the thorium needs neutrons to start the reaction and reach criticality. Having a liquid fuel makes this relatively easy, as the reactor doesn't need to be stopped for refueling: include a small amount of uranium-235, a startup source like californium-241, or both in the initial fuel mixture, then once the reactor has started, replace it with more ^{232}Th or other fuel.

The uranium-235 and uranium-233 both produce about the same amount of usable power upon fissioning (Table 1), so overall reactor efficiency will be determined by how much can be burned and how well the thermal energy can be extracted.

Plutonium fuels can be used in MSRs, as they are often used and bred in fast breeder reactors (FBRs). Most uranium-plutonium MSRs are typically designed to

$$^{232}_{90}\mathrm{Th} + \mathrm{n} \xrightarrow{\text{capture}} {}^{233}_{90}\mathrm{Th} \xrightarrow{\beta^-} {}^{233}_{91}\mathrm{Pa} \xrightarrow{\beta^-} {}^{233}_{92}\mathrm{U}$$

Figure 2: Uranium-233 breeding process.

Component	Energy (MeV)	
	^{233}U	^{235}U
E_{kin} of fission products	168	167
E_{kin} of prompt neutrons	5	5
E of prompt photons	8	6
E_{kin} of beta decays of fission products	5	8
E of photons following beta decay	5	7
E of delayed neutrons	≈ 0	≈ 0
Total capturable	200	193
Neutrinos	7	12
Total	207	205

Table 1: Uranium-233 and 235 induced thermal fission energy balance.

burn plutonium completely rather than breeding it in a blanket to be made fuel later. Plutonium-239 is a serious proliferation risk, and molten salt reactors are typically marketed towards developing nations.

1.2 Liquid Salts

The liquid salts must be able to dissolve the fuel and blanket and allow for easy chemical separation of fission products after irradiation. They must also be chosen to maximize performance and safety. Typical salts can be made of fluorine, chlorine, lithium, sodium, potassium, beryllium, rubidium, and zirconium compounds. Fluoride-based salts are a typical choice for thermal spectrum reactor designs, as they absorb fewer neutrons and are better moderators than other halides. However, penta- and hexavalent fluorides can boil at relatively low temperatures, making its use with uranium somewhat challenging.

Containing the liquid salt is not trivial. In the 1960s when MSR technology was first being tested in the Molten Salt Reactor Experiment (MSRE) at Oak Ridge, a special nickel-chromium-iron-molybdenum alloy was made called Hastelloy-N. It was compatible with the FLiBe (Li_2BeF_4) and FLiNaK salts that were tested. All of the metal parts that contacted the salts were made of Hastelloy-N. Nickel-based alloys are still considered for use today in prototype reactors, but they are known to become brittle when interacting with certain molten salts for long periods of time. Modifications to the alloy have been proposed to solve the problem, but the feasibility of the new materials are not proven for commercial power plants.

2 MSR Advantages over LWRs

2.1 Inherent Safety

Because conventional pressurized water reactors are held at extremely high pressures, often near around 155 bar, one of the biggest safety concerns is rapid depressurization of the reactor vessel. This drives the design of PWRs and necessitates a large containment building. Another safety concern is hydrogen gas production. If cooling fails, when the core reaches about 700°C, the coolant water starts to boil. The steam can react with the zirconium fuel rod cladding to produce hydrogen gas. This is what occurred at Fukushima Daiichi in 2011, destroying several reactor buildings and leading to the evacuation of at least 83,000 residents.

Liquid-fueled MSRs, by virtue of the fuel already being a liquid, cannot melt down. Due to the salts' high boiling and melting points, it is more difficult for an unintended release of radioactive material to occur, as any accidental fuel/salt discharge would solidify. Because the fuel is liquid and kept at regular atmospheric pressure, it can be quickly drained from the reactor vessel in the event of unexpected power loss. This is most often done with what's called a freeze plug, a material that is kept frozen with cool gas blown over it. If power to the reactor is lost or the core heats excessively, the gas blower will shut off, the plug will melt, and the contents of the core would flow into a drain tank. The drain tank can be designed to maximize the rejection of thermal energy from the decay heat to the environment. This is in stark contrast to conventional light-water reactors, as the fuel must remain in the core (where thermal energy loss is minimized) at all times, even in the event of an emergency.

2.2 Economic Advantages

The inherent safety of MSRs saves money by not requiring intricate safety systems. A notable and widely applicable example is the engineering of the containment building. Because MSRs do not need to be able to withstand a steam explosion in the event of pressure loss, their containment buildings can be made much smaller and cheaper. The decreased size also lends the reactors to a modular approach, something that is becoming more common, as the high construction costs of nuclear reactors compared to solar and wind power plants necessitate smaller installations. Several companies have been founded promising modular reactor designs like NuScale Power, Terrestrial Energy, and Seaborg Technologies.

2.3 Efficiency and Sustainability

Many types of reactors can burn nuclear waste from conventional reactors, but MSRs are particularly favorable for this. Because liquid-fueled reactors can be constantly refueled, variable amounts of nuclear waste products can be mixed with the fuel and burned without having to create a new solid fuel element. The liquid fuel also gives another perk: fission products can be removed while the reactor is operating. This means that fuel can be almost entirely used, in contrast with LWRs that burn a small fraction of the fuel element. In a similar fashion, transuranic irradiation products can be fed back into the core to be irradiated further and potentially to fission, resulting in waste that consists only of fission products, not transuranic elements.

Molten salt can generally be heated to higher temperatures than pressurized water. At Oak Ridge, the MSRE reactor operated at 650°C, whereas PWRs typically operate at around 315°C. This allows the electricity generation process to be more efficient: at 700°C, the efficiency is about 45%, much higher than 32-36% of LWRs. [4]

3 MSR Technical Challenges

The corrosive properties of liquid salts remain a challenge. Recent studies [3, 13] found that FLiBe salt can corrode various common choices for structural materials including Hastelloy-N, the FCC high entropy alloy (HEA). as well as 316 stainless steel. Corrosion problems do not pose a serious risk to prototypical reactor development, but commercial reactors would need to use a combination of materials that minimize corrosion. Salt purification may be required to prevent corrosion. In the MSRE, various materials had to be constantly removed like sulfur, various oxides, and structural metals like chromium, nickel, and iron. [10]

Salt phase changes and viscosity remains another challenge. Liquid salts that are designed to stay a liquid at very high temperatures may tend to freeze when being piped around the reactor and they may become viscous near their melting points. Choosing a coolant that will not freeze in the pipes while staying a liquid at operating temperatures further reduces the search space for suitable materials and could require pipes to be constantly heated during operation, detracting slightly from the safety of MSRs. According to Daniel Cooper, Chief Chemist at Seaborg Technologies, there is still much research to be done, including into light (low Z) molten salts.

Neutron irradiation can damage solid moderators in thermal MSR designs. In the MSRE, the graphite rods were made with high tolerance so they would be able to change size without being damaged. Upon irradiation, graphite initially contracts and then may swell until it loses structural integrity. [2]

Volatile fission products must not be accidentally discharged by a MSR into the environment. Solid-fueled reactors have generally solved this problem using vacuum outgassing and vacuum induction melting (VIM) [1, 7], but liquid-fueled reactors (especially uranium-based MSRs) do not have a clear large-scale solution.

4 Thorium vs Uranium

The MSRE demonstrated that thorium fuel can work in an MSR, so some reactor designs, notably the LFTR, use the thorium fuel process. Despite this, the most successful designs today are based on uranium, including Terrestrial Energy's 195 MWe IMSR. In December 2019, the IMSR was selected by Canadian and U.S. regulators for joint review, the first ever joint technical review of an advanced, non-light water nuclear reactor. [11] This motivates us to investigate the potential benefits and challenges of using the relatively untested thorium fuel cycle in a MSR.

4.1 Differences in Fuel Preparation and Compatible Salts

Perhaps the most potent advantage to using thorium is its abundance on Earth. Thorium is about three times as abundant as natural uranium, with 99.98% of natural thorium being the fertile ^{232}Th . Thorium does not require the enrichment (isotopic separation) that is typical of uranium fuels. This allows it to be used more cheaply and for much longer than uranium. Running thorium breeder reactors could allow us to use the existing supply of uranium for many times longer.

There is another notable benefit to using thorium over uranium: aiding salt creation. Fluoride-based salts are suitable for thermal-spectrum MSRs, as they do not become radioactive under neutron bombardment. The liquid fluoride thorium reactor (LFTR) can use fluoride salts because thorium does not produce a volatile fluoride compound when fluorinated, only ever becoming thorium tetrafluoride (ThF_4). Uranium on the other hand does produce uranium hexafluoride (UF_6) which boils at a relatively low temperature.

4.2 Thorium Proliferation Risks

The proliferation risks of the thorium fuel cycle are contentious. The International Atomic Energy Agency (IAEA) and International Nuclear Fuel Cycle Evaluation (INFCE) working groups found in 1980 that the proliferation resistance of any kind of nuclear reactor with the ability to reprocess spent fuel are similar, including thorium-based reactors. [8] In 2005, the IAEA had a different opinion, stating that the thorium fuel cycle has "intrinsic proliferation resistance." [5]

Uranium-233 can be used to create nuclear weapons, and it is also found in spent thorium fuel. In spent fuel, ^{233}U is mixed with ^{232}U , and because they are difficult to separate, this avenue of bomb-making is expensive. However, the chemistry of protactinium can make isolation much easier. [12] When ^{232}Th is irradiated with neutrons, several isotopes of protactinium are produced: ^{231}Pa , ^{232}Pa , and ^{233}Pa . The nuclear reactions with ^{232}Th in a reactor core can produce ^{231}Pa , ^{232}Pa , and ^{233}Pa . ^{232}Pa and ^{233}Pa both decay directly into ^{232}U (1.31 d) and ^{233}U (27 d) respectively, while ^{231}Pa is relatively stable with a half-life of 32.8 ky. In the absence of neutrons, the protactinium is no longer produced and can be isolated. After leaving this sample of ^{231}Pa , ^{232}Pa , and ^{233}Pa to decay for a few days, much of the ^{232}Pa will decay into ^{232}U . After another chemical separation to remove the uranium, this leaves a sample of ^{231}Pa and ^{233}Pa . Given a few months for the ^{233}Pa to decay into ^{233}U and separated again, a nearly pure sample of ^{233}U can be created suitable for use in nuclear weapons.

$$
\begin{gathered}
{}^{232}\mathrm{Th} + \mathrm{n} - 2\,\mathrm{n} \longrightarrow {}^{231}\mathrm{Th} \xrightarrow{\beta^-} {}^{231}\mathrm{Pa} \\
{}^{231}\mathrm{Pa} + \mathrm{n} \longrightarrow {}^{232}\mathrm{Pa} \xrightarrow{\beta^-} {}^{232}\mathrm{U} \\
{}^{232}\mathrm{Th} + \mathrm{n} \longrightarrow {}^{233}\mathrm{Th} \xrightarrow{\beta^-} {}^{233}\mathrm{Pa} \xrightarrow{\beta^-} {}^{233}\mathrm{U}
\end{gathered}
$$

Figure 3: Th-Pa decay chains that may yield highly concentrated ^{233}U .

There is a caveat to this process. As previous discussed, although uranium-233 produces about 2.4 neutrons per fission, they are used for both breeding and fissioning, leaving a maximum neutron multiplication factor of about 1.2. This is suitable for sustaining a nuclear reaction, but if a significant portion of protactinium or uranium is removed to create weapons, the reactor will not be able to sustain the reaction. Many researchers conclude that the proliferation risks of thorium power exist but depend on the specifics of the reactor design.

Thorium is not necessarily required for proliferation resistance. Seaborg Technologies claims that its uranium-based CMSR reactor is highly proliferation resistant.

5 Why Not MSRs?

If the advantages of molten salt reactor technology are clear and the technology was proven in the MSRE in the 1960s, where are all of the MSRs? In the 1960s and 70s, the United States needed their reactors to produce uranium and plutonium suitable for bomb-making, exactly the opposite of what is desired today. It has hard to blame the nuclear weapons program for uranium-based reactors. It paved the way for naval power experiments which led to successful LWR designs that inspired many commercial reactor designs. Without the weapons program and subsequent naval experiments, the initial research and development of nuclear reactor technology would have been prohibitively expensive. The history of reactor designs has skewed regulation to only support uranium-based light water reactors. After the accidents at Three Mile Island, Chernobyl, and Fukushima, safety regulations have only become more strict, and public opinion has turned negative. With public opinion so negative, it's very difficult to secure funding for nuclear power research, and in the current regulatory climate, it is extremely difficult to license and build new types of reactors, especially in the United States. These two problems, licensing and funding, are perhaps the two largest obstacles to generation-IV reactor technology, including molten salt reactors.

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Additional information for this report was collected from two Seaborg Technologies presentations at the 2018 Thorium Energy Conference given by Daniel Cooper and Troels Schönfeldt.