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Physical Aspects of Molten Salt Reactors

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Abstract:

The Generation IV International Forum (GIF-IV) [GIF-002-00 ,2002] has identified the Molten Salt Reactor (MSR) as one of the six technologies for further developments under generation IV. In a molten salt reactor (MSR), the fuel is dissolved in a fluoride salt coolant. The technology was partly developed in the 1950s and 1960s. With changing goals for advanced reactors and new technologies, there is currently a renewed interest in MSRs. Compared with solid-fueled reactors, MSR systems have different nuclear and physical properties. In this paper, the physical and nuclear aspects and properties of molten salt reactors are reviewed and discussed with comparison with solid –fueled reactors.

Key words: nuclear reactors, molten salt reactors, physical characteristics

1. INTRODUCTION

The Generation IV International Forum (GIF-IV) [GIF-002-00, 2002] has identified the Molten Salt Reactor (MSR) as one of the six technologies for further developments under generation IV. The MSR employs a circulating liquid fuel which is a mixture of molten salts. This fuel can be readily transported by pumps and pipes between a simple core (typically containing graphite moderator) and external heat exchangers. The produced heat is removed from the core by the salt itself. The reactor vessel and the piping is designed so that criticality can be achieved only in the core. Heat from the radioactive primary salt is transferred to a clean intermediate salt that then transfers heat to either a steam or gas cycle [1] as shown in Fig. 1.

The MSR is most commonly associated with the U-233/thorium fuel cycle, as the nuclear properties of U-233 combined with the online removal of parasitic absorbers enable the design of a thermal-spectrum breeder reactor. However, MSR concepts have been developed using all neutron energy spectra (thermal, intermediate, fast, and mixed-spectrum zoned concepts) and with a variety of fuels including uranium, thorium, plutonium, and minor actinides **[2].**

Molten Salt Reactors were original developed as a potential military aircraft reactor with a successful test reactor built in 1954 which ran at up to 860 C. Past MSR research was undertaken at Oak Ridge National Lab. (ORNL) during the 1950s/60s/70s; namely the Aircraft Reactor Experiment (ARE), Molten Salt Reactor Experiment (MSRE) and the Molten Salt Breeder Reactor (MSBR) projects **[3] (Samuel, 2009)** and a denatured molten salt reactor (DMSR) concept with enhanced proliferation resistance.

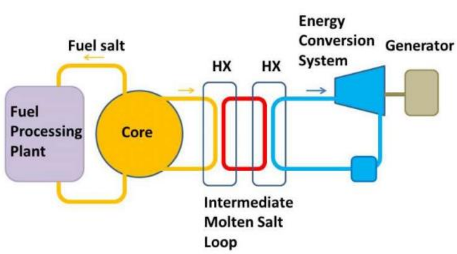

Figure 1. Simplified view of a liquid fuel MSR power plant

An objective review shows MSRs have unique attributes that lead to clear potential advantages ranging from overall costs, safety, resource sustainability and long lived waste issues [4]. In recent years, growing interest in this technology has led to renewed development activities. Much of this revival of interest has continued to focus on breeder options and while fluid fuel does simplify fuel processing technology, the degree of difficulty and costs can be underestimated by many, especially in terms of needed R&D [5].

More recent MSR concepts and fuel-coolant salt related technologies have been researched worldwide. These studies have led to the design of two fast spectrum concepts: the Molten Salt Fast Reactor (MSFR) **[6] [7]** and the Molten Salt Actinide Recycler & Transmuter (MOSART) [8][9]. The latter concept is designed to efficiently burn the transuranic waste from spent Light Water Reactor (LWR) fuel without any uranium or thorium support. Theses past and recent options and their different fuel cycles are shown in Table 1.

Table (1) MSR systems and key features of each system as developed during past and recent research programs. [10]

Reactor type	Mission	Neutron Spectrum	Reference Salt	Secondary Coolant
MSRE	power (breeder)[a]	Thermal	7LiF-BeF2-ZrF4-UF4	7LiF-BeFe2
MSBR	power (breeder)[b]	Thermal	7LiF-BeF2-ThF4-UF4	NaF-NaBF4
TMSR	power(breeder)[b]	Thermal	7LiF-BeF2-ThF4-UF4	7LiFeBeF2
MSFR(Th)	power (breeder)[b]	Fast	7LiF-ThF4-UF4	7LiF-(HN)F4
MSFR(Th)	Actinide burner[c]	Fast	7LiF-ThF4-UF4-(Pu-MA)F3	7LiF-(HN)F4
MOSART	Actinide burner[c]	Fast	7LiF-NaF-BeF2-(Pu-MA)F3	NaF-15^{7}LiF-BeF$_2$

[a] *feed by ^{238}U, ^{235}U (^{233}U) and after six months, the reactor was fuelled with ^{233}U (Capelli , 2015).*
[B] *feed by ^{232}Th, ^{233}U., "HN" refers to the heavy nuclides—thorium, uranium, and actinides.*
[c] *feed by TRU, Transuranic mixture: 87.5% Pu, 6.3% Np, 5.3% Am and 0.9% Cm in the form of fluorides. It corresponds to the composition of a conventional LWR fuel after discharge and 5 years storage (Capelli, 2015).*

In this paper, the physical and nuclear aspects and properties of molten salt reactors are reviewed and discussed with comparison with solid –fueled reactors.

2-PHYSICAL ASPECTS OF MSRs

2.1 Nuclear characteristics and design

Generally, the use of liquid rather than solid fuel in MSRs allows many reactor design features that are not possible with solid fuel. These include circulation of the fuel-containing liquid to act as a coolant and heat transfer mechanism, online chemical processing to remove parasitic absorbers and optimize the breeding and burning of materials, and different means of passive safety, such as draining the fuel from the core. [2]. A comparison in reactor design specially between Light Water Reactors (LWRs) and the molten salt reactors(MSBR) reveals the following differences [11]:

- The nuclear characteristics of as in case MSBR remains unchanged throughout the entire reactor life and hence only the initial nuclear design fulfills all the design requirements. On the other hand, for a LWR, the nuclear characteristics change during each fuel cycle as fuel burns. Hence the evaluation of nuclear parameters is necessary at certain steps during the fuel cycle. In addition, fuel inspection, shuffling, loading and discharging of fuel assemblies at each of fuel cycle is required.
- For a LWR, heat flux at hot channel factors is limited to conserve fuel rod integrity. In other words, strict power distribution monitoring is required for insuring the safe operations. For the MSBR, no such local monitoring is required for reactor safely, only the macroscopic power level monitoring is required. Problems such as densification of fuel pellets, pellet-clad interaction and fuel rods bow observed in LWRs do not exist in the MSBR.
- Also for the MSBR, consideration on the neutron absorption by xenon is not a serious problem as in a LWR because the inventory of xenon in the reactor is reduced to a very low level by fuel processing system, which is incorporated in the reactor system. As we can see above, we can conclude that nuclear design of the MSBR is considered simpler when compared with that of LWR.

2.2 Reactor physics characteristics

Such reactors have several reactor physics characteristics that are different from those of solid-fuel reactors. Some of these physical characteristics:

- **Circulating fuel and Delayed neutron fraction**

The physics of fluid fuel systems is characterized by some peculiar phenomena, connected to the movement of the multiplying material through the core and in the primary circuit [12].

It is known that in a reactor with stationary fuel the spatial delayed-neutron density distribution is the same as the distribution of prompt neutrons. In circulating fuel reactors, however, these distributions are different since the motion of the fuel alters the distribution of delayed neutron sources. This phenomenon generally leads to a change in the spatial distribution of prompt neutrons, but this change is insignificant if the contribution of delayed neutrons to the multiplication coefficient (β_{eff})is small compared to the neutron leakage (K_∞-1).Moreover, motion of the fuel causes a decrease in the number of delayed neutrons taking part in the fission reaction. There are two reasons for this. First, the motion of the fuel leads to a rise in the concentration of delayed neutrons at the boundary of the active zone, there is a consequent decrease in leakage, and this causes a decrease in the value of the delayed neutrons. Secondly, in circulating fuel devices some of the delayed neutrons escape from the active zone and do not take part in the fusion reaction [13].

The reduction of importance of delayed emissions associated to the motion of the fluid fuel affects both the static and dynamic behavior of the reactor. The critical condition depends on the velocity regime and the effective fraction of delayed neutrons is reduced, with obvious effects on the time-dependent response of the system.[12].

- **Nuclear Reactivity**:

Negligible xenon effect occurs because xenon continuously escapes from the fuel salt into the off-gas system. There is no change in core reactivity with time because fuel is continuously added as required. The fuel inventory in the reactor core is coupled to the reactor temperature. An increase in reactor temperature reduces the fuel inventory by expansion of the fuel salt with less mass of fuel salt in the reactor core. For fast-spectrum MSRs, this results in the unusual characteristic for a large fast reactor of having a strong negative temperature and void coefficient—a major potential safety advantage **[14]**.

A key characteristic of MSRs is the possibility to reprocess the fuel during reactor operation. This allows on one hand limiting parasitic captures due to FPs and on the other hand avoiding having a reactivity reserve to ensure criticality over time **[15]**

- **Fissile Inventory:**

MSRs have lower fissile inventories (Table. 2) than other reactor systems. This is a consequence of several factors: (1) no large out-of-core spent nuclear fuel SNF inventories that must be cooled before transport to reprocessing plants; (2) high power densities, which are a consequence of no power-density limits imposed by solid-fuel peak temperature constraints; (3) online addition or subtraction of fissile materials for reactivity adjustments; and (4) removal of high-cross-section fission products (such as xenon) from the reactor core. In solid-fuel reactors, the fuel must have excess fissile material to overcome the effects of fuel burn up and the buildup of neutron-absorbing fission products between refuelings. Minimizing the actinide inventories minimizes many of the risks and costs associated with actinide burning.**[16]**

Table.1 Fissile inventories [Mg/Gwe in Reactor] of different Reactor System"[16]

	Reactor	Reactor and Fuel Cycle	Commentary
MSR (thermal/epithermal)	1.45	1.45	Thorium Molten Salt Reactor (EU)
MSR (Fast)	5.5	5.5	Thorium Molten Salt Reactor (EU)
Pb FBR	6.7	20.1	BREST (Russia)
Na FBR	4.1	12.3	European Fast Reactor
He GFR	5.7	17.1	Gas-Cooled Fast Reactor (France)

[a]For all fast reactors, the ^{239}Pu equivalent mass is given (multiply it by 1.5 to find the total plutonium inventory). Thermal, epithermal, and fast refer to the neutron spectrum. FBR = fast breeder reactor; GFR = gas-cooled fast reactor; EU = European Union.

• **Burn up and Plutonium Isotopics:**

Relative to solid-fuel reactors, MSR fuel cycles have very high equivalent fuel burn ups and unusual plutonium isotopics with high concentrations of 242Pu.

- In solid-fuel reactors, SNF burn up is limited by fuel-clad lifetime that, in turn, limits fuel burn up and the burnout of plutonium. In non-breeder reactors, SNF burn up is also economically limited, independent of the technology. Excess fissile material is in fresh fuel when it is initially placed in the reactor core. This is required to compensate for fuel burn up over time. To assure reactor control, burnable neutron absorbers are then added to the fresh fuel to avoid excess nuclear reactivity in new fuel assemblies. There is an economic cost (extra enrichment) in "storing" excess fissile fuel in the new fuel assembly until it is needed toward the end of the fuel assembly lifetime. These factors fundamentally limit solid-fuel burn up**[14].**

- In an MSR, fuel is added incrementally to the liquid as required. No excess fuel and associated burnable absorbers are required. Selected fission products are removed from the molten salt and solidified as a waste form. Consequently, the normal burnup limits that define solid fuels do not apply to a liquid-fuel reactor. The plutonium remains in the salt, with the lighter plutonium fissile isotopes burned out faster than 242Pu. This has major implications in terms of proliferation resistance. The high 242Pu content makes the plutonium from an MSR much less desirable than plutonium from any other reactor type for use in weapons because of its very high critical mass.

• **Impact of the Moderation Ratio:**

The neutron spectrum of a nuclear reactor depends on its moderation ratio, i.e., the ratio of the amount of fuel to that of the moderating matter. In the thorium MSR, the neutron spectrum can be controlled by changing the size of the salt channels, extending from an extremely thermalized to a relatively fast spectrum. It appears from these studies that the TMSR can be operated with those three neutron spectrum types.

In the thermal spectrum, the safety issues are alleviated by reducing the distance between the salt channels in the graphite matrix. It is then possible to obtain a reactor with satisfactory properties although its feedback coefficient is only slightly negative and the irradiated graphite has to be reprocessed every few years. In the epithermal spectrum, the safety is improved thanks to a very negative feedback coefficient, but the drawback is more frequent processing of the irradiated graphite and a larger fissile matter inventory. The fast spectrum configuration with a single salt channel and without moderating graphite is the most promising. It has very negative feedback coefficients. This is true not only for the global temperature coefficient but also for the partial coefficients that characterize the dilatation or the heating of the salt, and the void effect **[15].**

- **Operating characteristics of MSRs under conditions of variable loads**.

Some problems arise in nuclear reactors with solid fuel elements with the periodic variation of output and reductions power plant peak loads can be avoided or at least significantly eased if one uses a reactor with a liquid nuclear fuel, e.g., a liquid-salt reactor (LSR) or MSRs: problem of cyclic thermal stresses which can result in destruction of the fuel elements with the periodic variation of output and problem of xenon poisoning of the reactor and arises in reactors with solid fuel elements upon a reduction of its output.

The investigation of the operating characteristics of an LSR with circulating fuel under variable-load conditions has shown the following **[17]:**

i. A change of the reactor output both in the case of its daily and weekly shutdowns and in a regulating range can be provided for by means of a change in the flow rate of the circulating fuel with simultaneous regulation by absorbing rods in such a way that the temperature field in the fuel salt remains constant during the regulating process. Such a regulating method reduces to a minimum the distortions of the energy distribution in the active zone of the reactor and excludes cyclic thermal loads on the reactor radius and the structural elements of the coolant loop.

ii. Regulation of an LSR by alteration of the flow rate of circulating fuel provides for a change in its output within the limits ± 10 % in a time not exceeding 6 sec.

iii. The continuous extraction of^{135}Xe from the fuel salt of an LSR excludes any appreciable poisoning upon a shutdown of the reactor.

iv. The maximum thermal stresses in the graphite moderator rods are less by an order of magnitude than the tensile strength of graphite. Such a strength reserve permits one to assume that the danger of cyclic thermal fractures of the graphite rods in an LSR is minimal.

v. The possibility of the operation of an LSR with a coolant temperature of 700-900^{0}C permits using a gas turbine electrical energy converter, for which the conditions of output variation are realized significantly more easily than for a steam turbogenerator.

vi.

Thus the physical characteristics of an LSR appreciably lighten the problems which arise in contemporary solid-fuel reactors upon their operation with variable output.

4. Neutron kinetics calculation and modeling

Because the fuel state of this reactor is liquid, and the reactor system based on the circulation of the molten salt in the core reactor and the heat exchanger. On the molten salt reactor, delayed neutron precursors drift with the fuel salt flow. This conditions make the neutron kinetics characteristics of the molten salt reactor difference than others solid-fuel reactors. In consequence, the neutron kinetics calculation analysis of the MSR is difference with the other type of react **[18]:**

Also according to the heat transfer phenomena are different as well. Most of the heat is directly deposited in the salt which acts as coolant of the reactor. Furthermore, in a moderated system a part of the fission heat is deposited in the moderator by gamma and neutron heating. This heat is removed from the core by the salt as well. Therefore, the moderator is at a higher temperature than the salt during operation. For these reasons, the modeling of the MSR differs from other reactors and the development of dedicated tools is important and as reviewed by [1]:

First, the basic reactor physics problems were introduced [19] and later the neutronics of fluid fuel systems were investigated using a point-kinetics model [20] or the quasi-static method [21][22]. It is common in these studies that the calculations used a prescribed one dimensional velocity field. The first coupled neutronics and thermo-hydraulic calculations were performed on one channel (that is the fuel channel and the surrounding graphite) of the moderated MSRs. This approach was employed for the AMSTER reactor [23] and for the MSBR [24][25]. Full core coupled calculations, which incorporate 2-D or 3-D reactor physics and 3-D heat transfer calculations, were reported as well, applied to the MSRE [26] and to the MSBR [27][28]. Even in these calculations, the flow field is parallel with the channels in the entire primary loop which includes the plena below and above the core and the loop outside the reactor vessel. Thus, the mixing of the precursors in the plena was not incorporated in the calculations although it was demonstrated to have an impact on the kinetic behavior of the reactor [29].

5. MATERIALS PHYSICAL PROPERTIES OF MOLTEN SALT REACTORS FUEL

Fuel Salts Must Integrate Reactor Physics, Heat Transfer, and Material Compatibility [30]:
・Reactor physics requirements
– Low neutron absorption
・ Thermal neutron absorption is of lower importance for fast spectrum reactors
– Radiolytic stability under in-core conditions
– Dissolve fissile materials
・ Both chloride and fluoride salts are industrially used as heat transfer fluids
– High heat capacity, high boiling point, low thermal conductivity fluids
– Melting point must be below ~525°C
– Relatively insensitive to fission products
・ Both fluoride and chloride salts, under mildly reducing conditions, are reasonably compatible with high temperature structural alloys and graphite
This led to the current options [31]:
- a 7LiF-BeF2 mixture as in MSBR for the thermal breeder to have optimum breeding conditions.
- a 7LiF-NaF mixture for the fast breeder to have the optimum spectrum.
- a 7LiF-NaF-BeF2 for the fast burner to optimize both the spectrum and the AnF3 solubility.

・**MSR coolants**

The options for the coolant salt has not changed since the MSBR design in the 1970s, see Table 3. Especially the NaF-NaBF4 mixture is currently considered as a good candidate. Table 3 compares the molten salt coolant to other conventional

Gen IV coolants. LiF-BeF2 (FLiBe) is considered as a primary in-core coolant, whilst NaF-NaBF4 is suitable as a secondary coolant in a separate loop. The data are after Forsberg [32]. Because of its excellent coolant properties, it is not unlikely that the near future use of molten salt will not be as fuel or coolant in a Molten Salt Reactor, but as the coolant of the Advanced High Temperature Reactor (AHTR) [31].

Table 3. Several coolants proposed in GenIV compared [31]

	unit	helium 60 bar 773 K	CO_2 60 bar 773 K	water 150 bar 573 K	sodium 1 bar 773 K	$LiF\text{-}BeF_2$ 1 bar $T > 723$ K	$NaF\text{-}NaBF_4$ 1 bar $T > 723$ K
ρ	$kg \cdot m^{-2}$	3.7	40.9	726	865	1940	1938
C_p	$kJ \cdot kg^{-1} \cdot K^{-1}$	5.2	1.2	5.6	1.3	2.3	1.5
$\rho\, C_p$	$kJ \cdot m^{-2} \cdot K^{-1}$	19.4	48.6	4066	1125	4540	2907
k	$W \cdot m^{-1} \cdot K^{-1}$	0.29	0.06	0.56	80	1.0	0.5
η	10^{-5} Pa·s	3.8	3.3	9	23.3	563	250

6-CONCLUSIONS

In this paper, the various physical aspects of molten salt reactors and nuclear properties were reviewed and discussed in comparison with other solid fuel reactors. It is important to identify these characteristics of liquid fuel reactors that need to be considered when determining the different requirements for the design, operation and selection of materials as well as the design requirements for simulation models for these reactors.

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