Review of R&D of thorium molten-salt reactor

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Abstract: Nuclear power will be used continuously as low-carbon energy for a sustainable society. On the other hand, prime attention is inclined to thorium molten-salt reactor (MSR) in recent years due to necessity for radioactive waste disposal, nuclear non-proliferation ("world without nuclear weapon") and higher safety of nuclear power. Thorium MSR is a type of liquid-fueled reactor utilizing thorium as fertile with some fissile materials contained in molten-salt. Research and development (R&D) of MSR started in the 1950s at Oak-Ridge National Laboratory (ORNL) for aircraft reactors. R&D activities were later extended to develop civilian power reactors using thorium, a concept which was completed in the 1970s. Nonetheless, technical problems still remained, such as temperature reactivity coefficient, material corrosion, and tritium permeation at heat exchanger. Since the 1980s several R&D activities to circumvent these problems have been done inside and outside ORNL. The problems relating to thorium MSR and recent R&D activities will be introduced in this paper.

Keyword: thorium; molten-salt reactor; technical problems

1 Introduction

There is hardly any change in the environment of humans before and after FUKUSHIMA accident. World population has exceeded 7 billion persons and energy consumption continues to escalate with economic growth in developing countries [1]. Most of the primary energy is supplied from fossil fuels such as coal, oil and natural gas. These fossil fuels emit not only greenhouse effect gases, but also other anthropogenic air pollutants (such as SO_x, NO_x and microscopic particles of PM2.5) [2]. In addition, it is reported that the production of conventional oil had already peaked in 2006 [3], thus it will be impossible to share the large portion of the primary energy supply considering projected future demand increase. Most countries such as China, India and USA continue to use nuclear power, while only a few countries, such as Switzerland and Germany, have adhered to a non-nuclear power policy.

Nuclear power, however, is plagued with several problems. There is no fixed final disposal area of radioactive waste except Finland, and its disposal difficulty mainly depends on long-lived radioactive materials. Another concern pertains to nuclear proliferation. The last – and the most important – problem is safety. FUKUSHIMA accident accentuates the need to strongly enhance the safety of nuclear reactors. FUKUSHIMA accident was initiated by earthquake and the consequent aftermath of the tsunami; therefore, it can be said that the possibility of occurrence of related accident of nuclear power plants is low at places which do not experience earthquakes and tsunamis. However, the likelihood of artificial destruction of nuclear power plant was made apparent by the 9.11 incident [4], leading to many researches being conducted regarding safety analysis against airplane crash after 9.11. The ultimate target of a nuclear power plant, from a viewpoint of safety, is not to release radioactive materials to the environment. Therefore, release and diffusion of radioactive materials in the case of destruction of a nuclear reactor should be evaluated if nuclear power plants are to be implemented in the future.

In these contexts, thorium molten-salt reactor (MSR) has attracted paramount attention. Generally, MSR entails two different concepts of nuclear reactors using molten-salt. One utilizes molten-salt only as coolant for primary circuit, while the other uses molten-salt as liquid fuel. In this paper, liquid-fueled molten-salt reactor will be discussed, and thus the word "MSR" represents liquid fuel MSR. Merits of using thorium from radioactive waste and nuclear non-proliferation points of view are described in the references [5, 6], and therefore are not elaborated in this paper. However, an aspect of thorium as resource will be described in Section 2, since usually the merit of thorium as resource is only enhanced as such

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abundance or widely available in the world, and such a simple description consequently causes misunderstanding. Thorium can be used in any kind of nuclear reactor. Nonetheless, the use of thorium does not guarantee enhancement of safety. Considering this, MSR is reconsidered owing to its high safety features. It should however be noted that technical problems of MSR especially using thorium are not recognized adequately. Thus, an outline of the concept of MSR will be explained in Section 3, and previous works at ORNL will be introduced in Section 4. Pending problems of R&D of MSR at ORNL are discussed in Section 5. Finally, recent R&D activities of MSR after ORNL will be introduced in Section 6.

2 Thorium and rare-earth

The relation between thorium and rare-earth cannot be disregarded when we discuss thorium as a resource. This is because: ① thorium exists in the same minerals which are mined to obtain rare-earth [8], ② there is no commercial value of thorium nowadays and thorium utilization is not independent, ③ rare-earth, on the other hand, has its own commercial value and thorium occurs as a byproduct of rare-earth refining, ④ rare-earth contains 17 different chemical elements and it was difficult to separate them and finally, ⑤ thorium is a radioactive material.

Regarding reason②, it is true that thorium was commercially used since its discovery in 1928 for crucible, mantle of lantern and so on. However, its consumption amount was less than 100 tonnes per year and recently thorium is not used for these purposes due to reason⑤. Thorium had been studied as a nuclear fuel since 1940. Uranium is another nuclear fuel and, currently, only uranium is used as commercial nuclear fuel. This is because only uranium contains fissile isotopes. On the contrary, thorium does not have fissile isotopes and requires other fissile materials such as plutonium, HEU or artificially-produced uranium-233 to initiate fission chain reaction. The use of HEU has to balance with the uranium fuel cycle owing to its limited supply capacity. In the case of using plutonium, its supply amount is mainly constrained by the capacity of uranium fuel cycle. These are the reasons that reinforce that thorium utilization is not independent. Moreover, there is no commercial production facility of uranium-233. Therefore, opinions that thorium is alternative to uranium and that thorium use guarantees energy security [9] due to its larger reserve than uranium, are essentially not valid.

In spite of the aforementioned dependency of using thorium to other fissile supply, thorium was stored by the US government in the 1950s, due to its feasibility as nuclear fuel [10] and the lack of enough investigation regarding uranium resource. As a result of thorium storage, huge amounts of rare-earth occurred as byproduct due to the previously mentioned reason①. Thus, application of rare-earth was developed. Since it was difficult to separate each respective element from rare-earth (reason④), its application was limited to membranes for oil extraction by Mischmetall (mixed metal). It then became possible to separate each element by solvent-extraction method and thus extending their realm to various applications, amongst the important ones being optical and magnetic applications. For example, europium and terbium were used to obtain clear red luminosity. Hitachi's "Kido color" (commercialized in 1968) is a popular color TV. The word "Kido" has two meanings of rare-earth and luminosity in Japanese. Neodymium magnet, which was developed by Masato Sagawa in the early 1980s, has 10 times stronger magnetic induction than ferrite magnet. Strong neodymium magnet contributed to minimization of the electric motor size. Small and strong electric motors have facilitated the miniaturization of many machines. Thus, rare-earth use will continue in the future. Contrary to the development of rare-earth application, thorium utilization stagnated due to its requirement of other fissile supply and adequate reserves of uranium, therefore rendering the termination of USA's national storage of thorium in 1958.

However, as noted in reason①, expansion of rare-earth production caused accumulation of thorium as byproduct. In addition, since thorium is radioactive material (reason⑤), it is necessary to spend money to manage the tailing of rare-earth refining containing thorium for environmental protection. This means that if some country does not carter the necessary cost for managing rare-earth tailing and increases in the commercial competitiveness of rare-earth supply, it becomes possible to monopolize rare-earth supply. This is the policy which China did since the 1990s and currently China has 97% of rare-earth supply share. Although there are many developments in rare-earth mining after rare-earth crisis initiated by China in 2010, it is difficult to create a healthy rare-earth market. This is due to the fact that no essential countermeasures have been enacted to take care of thorium (such as inspection of environmental protection) except what the author proposes [11 -14]. An IAEA's meeting on thorium resource will be held at Wien in September 2013 based on the background information mentioned here [15].

The reason why thorium is utilized currently is not from a viewpoint of enhancing safety of nuclear power, but from reduction of radioactive waste, possibility of nuclear proliferation and solving rare-earth problem accompanying thorium use. It can be said that accumulated amount of plutonium in the past usage of uranium, which relates both to problems of radioactive waste and nuclear proliferation, reaches an amount adequate enough to consider thorium utilization [16]. The author's evaluation is based on using a molten-salt reactor, thus the available capacity of thorium utilization is somewhat large. In the case of thorium utilization with ordinary heavy water reactor, the corresponding capacity becomes smaller [17]. In any case, it is necessary to utilize thorium for internalizing thorium in economic activity and related technological development is required.

3 Outline of a molten-salt reactor

MSR is included as one of the generation IV reactors [18]. Only MSR explicitly indicates the use of thorium among 6 candidates of generation IV reactors. General concept of MSR will be described in this section.

A nuclear reactor is an equipment which induces nuclear fission reaction to generate thermal energy which is converted to electricity. It mainly has two functions: to control nuclear fission reaction and to remove heat. Therefore, the surrounding instruments have to satisfy engineering requirements (such as mechanical properties or operability) so as to meet the above two functions. Nuclear fission reaction is an interaction between fissile and neutron having different combination of fissile materials and neutron energy. Fissile materials are, for example, uranium-235, plutonium-239 and uranium-233. In the case of using thorium as fertile, uranium-233 is produced from this thorium. Uranium-235 and plutonium can also be used with thorium. Difference in neutron energy (such as fast or thermal) affects the use and materials of moderator.

Since thorium does not contain fissile isotope (even uranium contains merely 0.72% of fissile isotope), a breeder reactor was targeted historically to use thorium. The fact that uranium-233 has a large η–value in the thermal energy spectrum, thermal neutron was mainly considered to use thorium. Conversely, plutonium has a large η–value in the fast energy spectrum, and thus a fast breeder reactor is usually considered.

A system consisting of fissile, fertile and moderator (sometimes it is not used) can be operated at critical conditions. In the case of a light-water reactor (LWR), low-enriched uranium is used as fissile (uranium-235) and fertile (uranium-238), and water is used as moderator. A typical design of thermal spectrum MSR uses graphite as moderator. Fluoride uranium and thorium are mixed with fluoride lithium – fluoride beryllium (Flibe). Flibe can also serve as a moderator. This mixed salt is heated at temperatures higher than its melting point, so that it readily flows in the gap of solid moderator. In short, MSR is a kind of liquid-fueled reactor.

Concentration of fissile material in the fuel salt is less than a few %, while concentration of fertile is only about 10 %. If the temperature of fuel salt becomes lower than its freezing point, most of the radioactive materials including fission products (FP) are maintained in the bulk of fuel salt. This is slightly distinct from ordinary oxide pellet fuel of uranium, in which all the radioactive materials occupy the surface being exposed to atmosphere if the crud of fuel rod disappears. Even in a liquid fueled reactor, radioactive material can easily diffuse into the atmosphere as long as aqua is used owing to its high vapor pressure and low boiling point. On the contrary, molten-salt has a high melting point (450^{o}C for Flibe) and low vapor pressure, thus fuel salt soon freezes once decay heat reduces after reactor shut down. The above discussion serves to distinguish the characteristics of MSR. From a viewpoint of heat removal, water which acts as moderator removes heat from fuel rod and transfers toward the outside of the reactor vessel in LWR. Contrary, fuel salt (which is mainly carrier of fuel materials (fissile and fertile) and fission products) transfers heat by increasing its own temperature. Difference in the mechanism of nuclear fission reaction and heat removal between LWR and MSR is shown in Fig. 1.

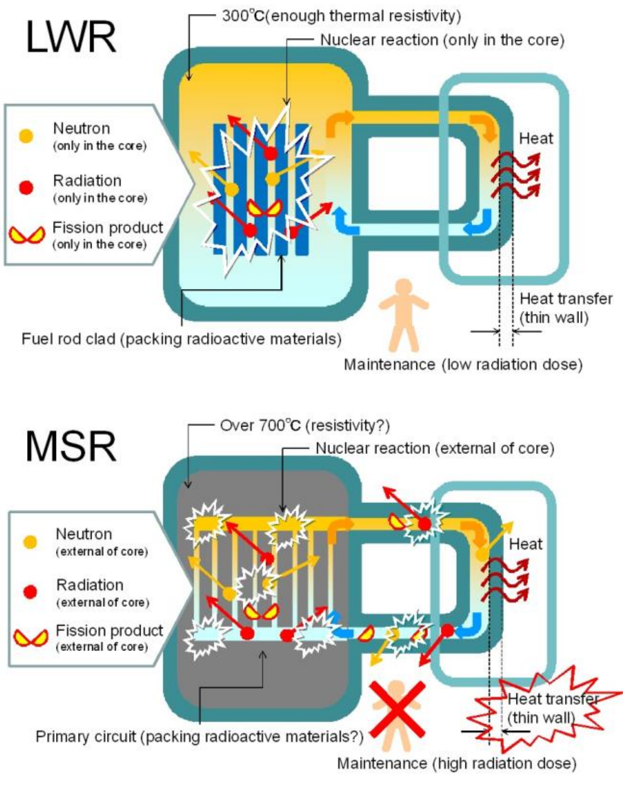


Fig. 1 Mechanism of nuclear fission reaction and heat removal of LWR and MSR.

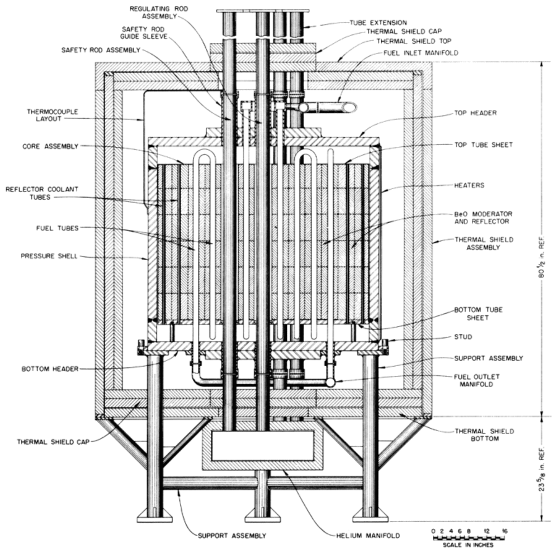
In LWR, so long as operation is normal, radioactive materials exist only inside the core and radiation occurs only here except for a few cases. On another hand, all radioactive materials circulate in the primary circuit in MSR with molten-salt as coolant. During this circulation, therefore, radioactive materials deposit or permeate in structural materials such as graphite moderator. Moreover, a reactor whose thermal output exceeds 10 MW has to use external heat exchanger incorporating many narrow tubes to confer a large surface area for heat transfer. As a result of circulation of radioactive materials to these pipes, not only γ–rays from FP, but also delayed neutrons are emitted outside the reactor core. This external emission of radioactivity is the most significant difference from LWR, and causes problems in design and operation of MSR. The design of a nuclear reactor tends to focus on neutronics. However, the heat removal system of both the fission energy and decay heat often became a cause of engineering accident; thus a suitable design of heat removal is key to realizing safe and stable operation of nuclear reactors. This is particularly important for MSR, since fluid fuel affects both fission reaction and heat removal as described in this section.

4 Early R&D of MSR at ORNL

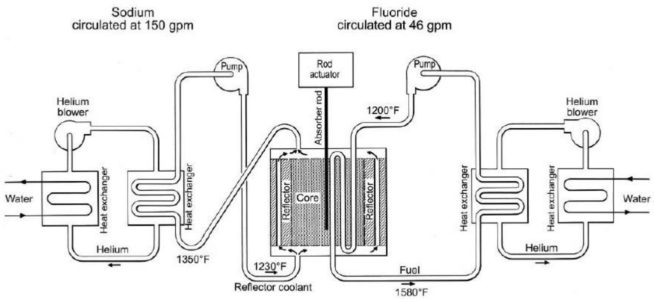
4.1 ARE

It is useful to review ORNL's early works of MSR, to glean aspects for evaluating recent R&D activities in the world. As shown in the previous section, MSR is a liquid-fueled reactor. Other liquid-fueled reactors besides MSR will not be elaborated for brevity purposes, but it should be mentioned here regarding the rationale for considering liquid-fueled reactors. It is because nuclear fission reaction not only generates heat, but also many different chemical elements. Additionally, fission reaction changes along with time, requiring chemical process such as removal of neutron poison or addition of consumed nuclear fuel. Since chemical processing is done with solvent, liquid-fueled reactor utilizes the solvent for chemical processing and heat removal. Molten-salt is used as solvent in MSR.

The first MSR in history is ARE (Aircraft Reactor Experiments), which was developed for jet engine of long range bomber to carry atomic bomb [19]. ARE was not MSR from the first design. At first, ARE was a solid-fueled reactor using uranium oxide fuel pin inserted in a BeO moderator block. Heat was removed by cooling this BeO block with liquid Na. It was later found that the reactivity temperature coefficient became positive under this configuration. Molten-salt fuel (containing uranium in fuel pin instead of solid fuel pin) was expected to exhibit a negative temperature reactivity coefficient due to the characteristic expansion of liquid phase with increasing fuel temperature. However, it was also confirmed that this method was not suitable for heat removal and fuel exchange. Finally, an idea to realize both heat removal and fuel exchange in externally extended pipes outside of reactor core using circulation of molten-salt fuel was proposed. This was the first example to realize the primitive concept of MSR, in which fuel salt has both functions of nuclear fission reaction and heat transferring medium. Reactor core of ARE and its heat removal system is illustrated in Fig. 2.



(a) Core design.



(b) Heat removal system.

Fig. 2 Schematics of ARE.

Thermal output of ARE was 2.5 MW and it was operated for 3 days. ARE was an experimental reactor for jet engine and long lifetime (for several years) was not considered. In the next step of ARE, which was named ART (Aircraft Reactor Test program), only 6 months was the required lifetime of reactor. Since the lifetime of the jet engine's reactor was short, fertile was not contained in fuel salt; only uranium-235 was considered as fissile material.

Experiences of ARE give us many hints of designing MSR. At first, ARE showed that liquid fuel is useful to maintaining a negative value of reactivity temperature coefficient, which is the most important aspect for safety of nuclear reactor. ARE could be operated without using a control rod for load-following mode. This is due to the large negative reactivity temperature coefficient. However, it should be noted here that there are instances in which MSR designs might have positive values of reactivity temperature coefficient as described in Section 5. This becomes apparent for cases of using thorium as fertile or in the event of a large thermal output power. The second is that it was naturally available to separate gaseous FP without using any active operation. Xenon and krypton (which are neutron poisons) are gases, and in principle their solubility to molten-salt is low. Thus, the gas phase separates from the liquid phase. In the case of using uranium oxide fuel rod, it is not possible to operate the reactor at high temperatures due to thermal limitation of fuel crud even though oxide itself has high thermal resistance. On the contrary, ARE could be operated at high temperatures (860°C) because the molten-salt fuel had merely no thermal limitation. In addition, since its vapor pressure is low compared to aqueous liquid fueled reactor, ARE could be operated at ordinary (ambient) pressure.

ARE utilized liquid Na as coolant for BeO moderator, which was equipped in the first design of ARE using solid fuel rod. In order to avoid mixing of liquid Na and fuel salt, fuel salt was packed in pipes. The pipes were inserted in holes which were originally designed to insert fuel rods. Thus, the piping materials were exposed to high neutron flux and high temperature. Although nickel-based Inconel was used as piping material for ARE, Inconel did not have enough corrosion resistivity. A necessity therefore existed to develop new configuration and structural material for the next program of MSRE (Molten-Salt Reactor Experiment). Several reactor designs were proposed for the ART after the success of ARE. Amongst the exemplar designs, one design in ART aided to evaluate latter designs such as MSRE, MSBR and recent R&D. An example is shown in Fig. 3.

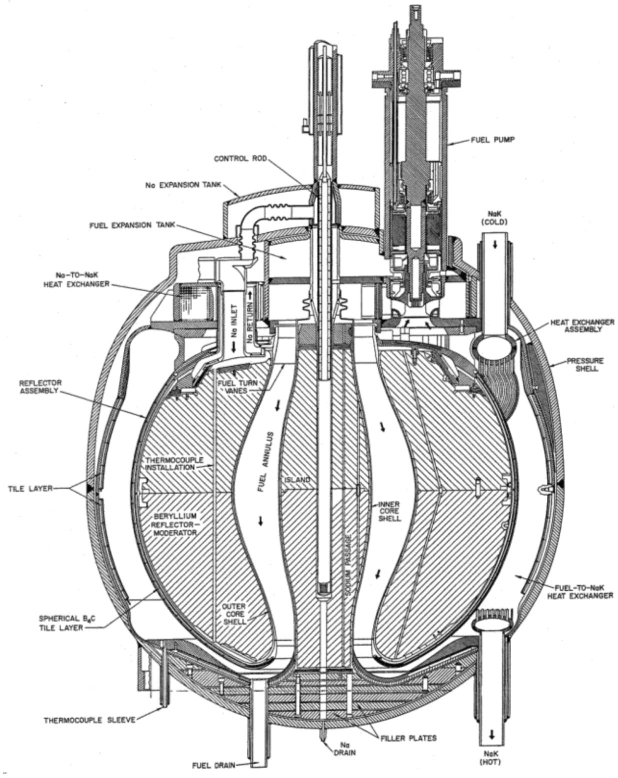


Fig. 3 Reactor core and heat removal system proposed in ART.

The design shown in Fig. 3 is termed as "Fireball", based on its unique shape. As can be seen from the figure, reactor core corresponds to the center part of spherical vessel having a diameter of 1.4 m. Its thermal power is 60 MW. Fuel salt flows into the reactor core from atop, and flows down into the annulus channel formed by moderator made of BeO. The annulus channel is surrounded by a BeO reflector. The hot fuel salt coming out from the core distributes toward the gap formed by the reflector and reactor vessel. Heat exchanger from fuel salt to liquid Na-K metal is located in this gap having a number of narrow pipes, and the Na-K transfers heat to air. It can therefore be said that both reactor core and heat exchanger are located inside the reactor vessel in this design. Such a design was aimed at making the whole system compact, since this reactor was to be used as jet engine for an airplane. This unique design is reconsidered in the R&D works after ORNL activities because it was seen helpful to overcome engineering problems relating to heat exchange of MSR.

4.2 MSRE

Although the ART program was cancelled due to the successful development of ICBM (InterContinental Ballistic Missile), the technology background was transferred to civilian power program of MSRE (Molten-Salt Reactor Experiment) [20]. MSRE has one common feature of using molten-salt fuel with ARE, but the design requirements and structure are quite distinct.

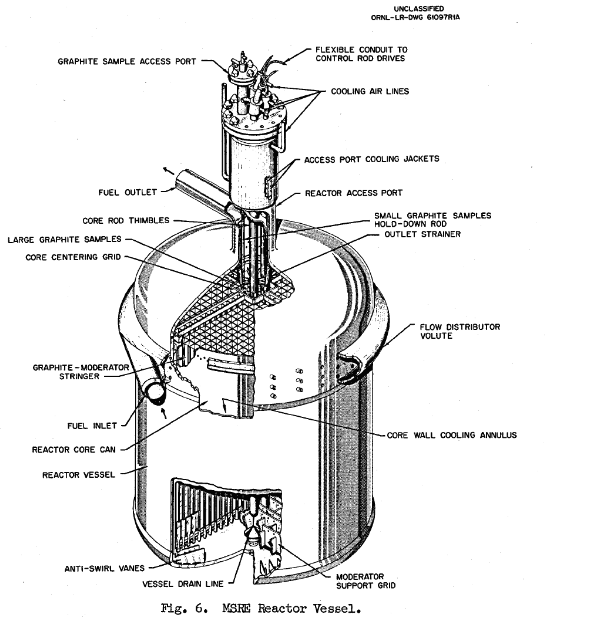
The largest difference is in the design lifetime of the reactor. While ARE's lifetime was shorter than half a year, MSRE was ultimately targeted to have a 30-year operation time. Obviously, MSRE itself was an experimental reactor tested for 4 years, but its fuel salt component, structural material, total system configuration including heat exchanger, and replacement of moderator were determined by considering years-long operation. An important issue to operate nuclear reactor for long years is to convert fertile to fissile by using excess neutrons emanating from nuclear fission reaction. If conversion ratio (CR) is equal to 1, this reactor can be operated by using initially equipped fissile for an entire lifetime. To facilitate the conversion of fertile to fissile, MSRE was designed to use thorium in the next demonstrator named MSBR (Molten-Salt Breeder Reactor), albeit MSRE itself did not utilize thorium.

The second largest difference pertains to output power. Recently USA is promoting SMR (Small Modular Reactor) [21], although a large output power was considered for civilian power station such as 1 GWe in the 1960s. Even though MSRE was designed to have 10 MW of thermal output power, its system configuration was determined to demonstrate MSBR having 3 GWth. This means that MSRE was not optimized for small MSRs. Total output thermal power of a reactor can be determined by a product of the power density and volume, thus it increases by a third power against reactor dimensions. On the other hand, surface area (which is an index of heat transfer) increases only by a second power against reactor size. Therefore, 4 loops were considered in the primary circuit for external heat exchanger to increase the surface area of pipes in MSBR [22] and could not apply the heat removal system of ART's "Fireball". Though output power of MSRE is not so large, this heat removal system of MSBR was demonstrated in MSRE. In short, this configuration is not necessarily an optimized design for small MSR as it produces a few MWs of thermal power.

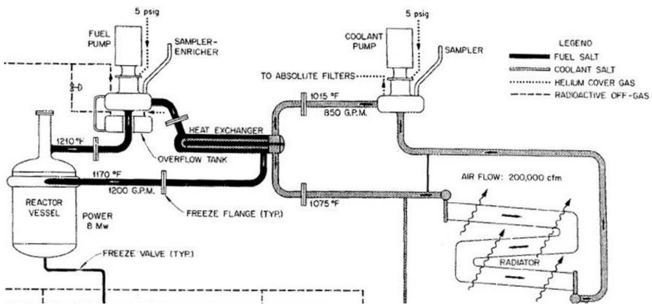
A large reactor which has 3 GW thermal output power adopts a Rankine cycle using steam. As mentioned in Section 4.1, the primary circuit is operated at ordinary pressure because vapor pressure of selected salt is low. Conversely, steam circuit is operated at 25 MPa to enhance thermal efficiency [22]. In order to avoid core destruction caused by a break of pressure boundary at the heat exchanger, a secondary circuit is inserted in MSBR. Even though there was no need to use pressure buffers due to the usage of air cooler having low pressure for heat removal, MSRE also adopted a secondary circuit as MSBR configuration.

Another role of the secondary circuit is to prevent the transfer of tritium toward the steam circuit which occurs at the primary circuit. NaF-ZrF_4 was used as solvent salt in ARE, although LiF-BeF_2 was used in MSRE. Here, enriched ^{7}Li is used because natural Li contains ^{6}Li whose thermal neutron absorption cross section is large. However, small amounts of remaining ^{6}Li absorb thermal neutron and produces tritium. ^{7}Li also produces tritium by absorbing fast neutrons [23]. Tritium easily permeates metallic material. The heat exchanger tube is made thin to enhance heat transfer, thus tritium intrudes the tube and is replaced with hydrogen forming the water in the steam circuit. To prevent the transfer of tritium, a suitable salt composition in the secondary circuit is selected to trap tritium. The problem posed by tritium was not well recognized when MSRE was developed, and thus Flibe was also utilized as secondary salt. In MSBR, NaF-$NaBF_4$ is considered as secondary salt. In this case, about 70 % of tritium is trapped and accumulates in the secondary salt. Therefore, it is crucial to handle tritium after termination of MSBR [24].

In the ARE, it was necessary to insert metal pipe in the holes in which fuel rods were inserted, thereby metal pipe posed a problem of corrosion resistance. For the MSRE, molten-salt fuel directly flows in holes formed in graphite moderator, to remove heat from moderator heated by γ-ray and to remove heat of fuel salt itself. In other words, there is no fragile metallic material in the core. Core and heat removal system of MSRE are shown in Fig. 4.



(a) Core design.



(b) Heat removal system.

Fig. 4 Schematic of MSRE.

At this juncture, it is worthwhile to mention about breeding. A limiting condition of nuclear power is that the available natural fissile is only uranium-235 and its amount is subtle. In consideration of this, fissile supply must be guaranteed to facilitate the widespread use of nuclear power. This is the impetus for considering breeding. One neutron is used for sustaining fission chain reaction, while the rest are used to convert fertile to fissile. Some designs, such as fast breeder reactors, adopt blanket regions for breeding which surround the core region for energy generation. A similar concept can also be applied to MSR.

There are two different approaches to achieve these two regions of core and blanket by using the same fluid or different fluids. The former is referred to as a "One-fluid two-region MSBR", while the latter as "Two-fluid two-region MSBR". From a viewpoint of achieving better conversion ratio, a two-fluid MSBR is desirable. Conceptual configuration of a two-fluid MSBR is shown in Fig. 5.

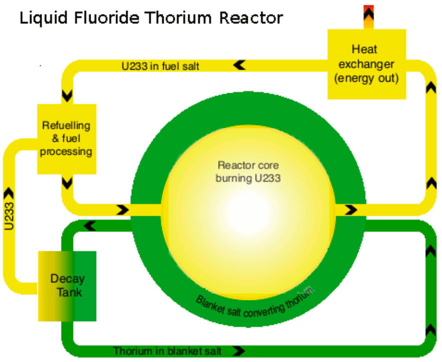


Fig. 5 Schematic of a Two-Fluid Two-Region MSBR.

In the core region, fuel salt (which contains only fissile) is used, in order to utilize neutron for fission reaction by eliminating fertile which acts as a strong neutron absorber. There are some leakages of neutron from the core region to surrounding blanket region, and these neutrons are absorbed by fertile which is added in the blanket salt. As a result of absorption of neutron by fertile, fissile is produced in this blanket salt. Therefore both fertile and fissile exist in the blanket salt. However, the newly produced fissile material is separated by online reprocessing facility (this corresponds to "Decay Tank" in Fig. 5), and fed to fuel salt which flows core region. Thus, the amount of fissile material is not so large in the blanket region.

In the configuration of a two-fluid MSR, it is necessary to separate these two regions by some separators. It has been recognized that it is difficult to find materials which have both corrosion resistivity against molten-salt and radiation resistivity for separators. Even though one-fluid two-region MSBR design was developed after construction of MSRE which has much less engineering difficulties, MSRE was designed under the concept of a two-fluid two-region MSBR. The purpose of MSRE was to demonstrate the core part of a two-fluid two-region MSBR. Therefore, there was no reflector in the MSRE reactor vessel, as can be seen in Fig. 4 (a), in order to utilize leaked neutrons from core region at the blanket region. As mentioned above, there was no thorium in the fuel salt of MSRE because thorium would be added in blanket salt. MSRE had a very large negative temperature reactivity coefficient owing to this kind of configuration. That is, the small dimension, absence of a reflector enabling large leakage and the lack of thorium which acts as a strong neutron absorber.

The latest operated MSR in history is this MSRE which was built 40 years ago. Thanks to the successful experiment, we sometimes hear opinions of small thorium MSRs which have similar configurations akin to MSREs that can be commercialized soon [9]. Nonetheless, more discussions are necessary. As aforementioned, no tests utilizing thorium were conducted with MSRE. This means that at least the following 3 points have not yet been tested. First, there is lack of experience to demonstrate the strong γ-activity emitted from tallium-208 which is a daughter nuclei of thorium in MSR. Thorium has already been used in LWR or high-temperature gas-cooled reactors, but these experiences cannot be simply applied to MSR. This is particularly due to its unique configuration that fuel salt circulates in primary circuit. This lack of experience, for example, causes uncertainty in operation such as maintenance. Second, there are remaining problems of neutronics relating to uranium-233 generated from thorium having a small portion of delayed neutrons, and the flow of precursors of delayed neutrons toward outside of the reactor core. Third, there is lack of experience regarding heat exchanger between molten-salt and steam.

4.3 MSBR

MSBR was a demonstrator of power station targeted to have 1 GW of electricity generation. As mentioned in Section 4.2, at least two regions were considered necessary to achieve breeding. At first, concept of a two-fluid design was discussed due to its easiness of construction for breeding MSR, but it was also recognized that it would be difficult to find a suitable material that can be used as a separator of core and blanket regions. Thus, in parallel, one-fluid design was considered and finally a design was completed, enabling 1.06 of breeding ratio. Its system configuration is shown in Fig. 6.

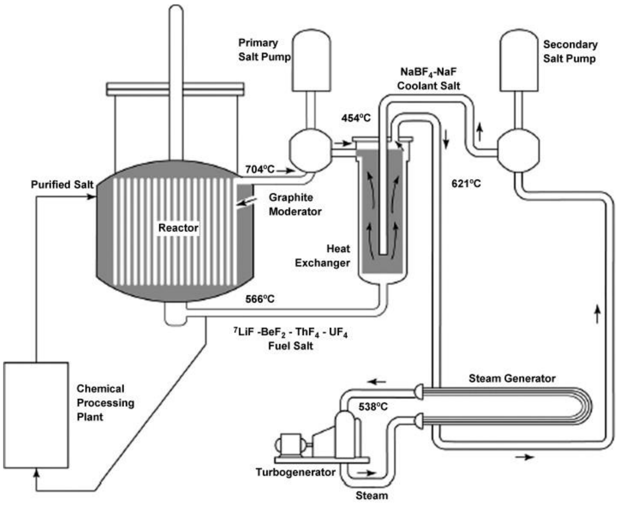


Fig. 6 Schematic of MSBR.

There are 3 points here that enabled breeding by using only one fluid. First, it was an adjustment of the volume ratio between fuel salt and moderator. Volume ratio of fuel salt is small in the core region (13.2 %), and thus the capability of moderation is high. On the other hand, this ratio is large (37 %) in the blanket region to enhance resonance absorption thus promoting conversion of thorium to uranium-233 [22]. In addition, a reflector is used in MSBR in order to cover the blanket region.

A second aspect was the utilization of online reprocessing [25]. This method is, obviously, available in a two-fluid MSR. The aims of online reprocessing are to remove non-gaseous FP and to evacuate protactinium (Pa). Gaseous FP can be removed by any kind of MSR without the mandatory need of any special facility. The purpose of removing non-gaseous FP is to avoid unnecessary loss of neutron by absorption to the FP. The purpose of evacuating Pa is also to avoid neutron loss; nevertheless, the importance of Pa is that Pa converts to uranium-233 via β-decay. The half-life of Pa is long (27 days) and its absorption cross-section of neutron is large (one-third that of uranium-233). Thus, circulation of Pa in the core prevents conversion of Pa to uranium-233. In a two-fluid MSR, Pa can almost completely be separated by processing blanket salt. Uranium-233 can be extracted and added to fuel salt of core region as shown in Fig. 5. Moreover, not all the fuel salt (210 m^3/min) is fed into online reprocessing facility, but only a few portions (3.33 *l*/min) are fed by using a branch line. Therefore, Pa exists in the core region. Online reprocessing facility was not equipped to MSRE and has never been tested. In the R&D program of MSBR, a much simple MSCR (Molten-Salt Converter Reactor) omitting complex online reprocessing from the same core design of MSBR was also proposed [26].

A third point was the replacement of graphite moderator [27]. This was geared toward enhancing the conversion ratio by using high neutron flux in the core. Since there is a limitation of irradiation resistivity of graphite, it was considered crucial to replace all moderators in every 4 years in ORNL's design. It was optimized to replace graphite moderator partially in every 4, 8, 12 years based on difference of irradiation amount in Ebasco's design. This kind of replacement of moderator requires shut down of MSBR and also demands additional facilities such as cranes.

In spite of the attractiveness of the MSBR project, MSBR was not constructed.

5 Pending problems

In history, only two reactors based on MSR concept were built and operated, however there is no actual reactor which is currently available. Problems that are pending during the R&D activities at ORNL will be described in this section.

From a view point of manufacturing, operation and neutronics, the following issues compare to solid fuel reactors such as LWR [28].

a. MSR is a non-sealed radioactive source.
b. Circulation of FP toward outside of core.
c. Loss of precursors of delayed neutron.
d. Fluctuation and temperature reactivity coefficient.
e. Production of tritium.

Regarding concern "a", though it is based on design, it is necessary to use an external primary circuit including pump, heat exchanger having a number of narrow tubes to efficiently remove heat for a large thermal power reactor as mentioned in Section 4. All designs described in the next section after R&D activities at ORNL, except the author's example, utilize external primary circuits. There is therefore a possibility of leakage of radioactive materials from this primary circuit being determined as non-sealed radioactive source.

As regard to concern "b", irradiation of materials and exposure to radiation during maintenance caused by radiation outside of the reactor core are of prime interest. In particular, the γ-rays emitted from thallium-208 are important in the case of using thorium. The fact that thorium was not contained in the fuel salts of ARE and MSRE, indicates that there was hardly any experience on operation of MSR under γ-activity. Fuel salt is not a simple composition of Flibe with nuclear fuel, but rather changes along with time due to nuclear chain reactions. The effects caused by this inconstant composition of fuel salt toward structural materials such as extraction, deposition and corrosion are still not clear. Corrosion test of Hastelloy N which was used for MSRE's structural materials have been carried out. Corrosion rate in a forced convection loop having a temperature difference between 566 ^{o}C and 704 ^{o}C was 0.003 mm in a year [29]. This corresponds to 0.09 mm for 30 years' operation. It was designed to have 0.9 mm thickness for pipes of heat exchanger of MSBR [30] to be about 10 % of depth. Taking account of uncertainty of manufacturing and fabrication of the many pipes in a heat exchanger, it will be necessary to take care of corrosion of pipes. These tests were done for an initial composition of fuel salt of MSBR. Therefore the effect of FP, testing of which is most important, is still not clear.

Concern "c" is pivotal for safety. Delayed neutrons are emitted after a few seconds or a few minutes of nuclear fission. There are 6 groups of precursors based on the range of emission time of delayed neutrons. Delayed neutrons are not important for an atomic bomb, which rapidly attains super-criticality, but they are indispensable for nuclear reactors which are maintained at criticality under human control.

In a solid-fuel reactor, such as LWR, precursors stay at the location where they are produced by fission reaction. Thus, delayed neutrons are produced here so as to contribute to chain reactions. Conversely, in MSR using flowing liquid fuel, the contribution of delayed neutrons changes. Reactor output power, operating condition and difference of fissile are amongst the aspects that affect this contribution. For a large reactor output power, flow rate tends to be high in order to transfer thermal energy toward outside of the reactor core. As a result, the maximum flow velocity of fuel salt at the center of reactor core of MSBR was 2.6 m/s. Core height was 4 m, thus fuel salt passed through the core within a few seconds. If the total pipe length of the primary circuit is short, there is a possibility that delayed neutrons are emitted in the core due to the return of precursors to the core. In reality, it takes 14 seconds for one circulation of the primary circuit of MSBR [22]. Therefore, most precursors (except group 5 and 6 having half-lives shorter than 1 second) emit delayed neutrons outside the reactor core. This is a situation of normal operation, thus contribution of delayed neutrons changes depending on reactor operation. One of the important cases is the termination of circulation of the primary circuit caused by pump trip, since delayed neutrons which are lost outside of core are emitted in the core consequently adding to positive reactivity.

Contribution factor of delayed neutron β is different by fissile and based on JENDL3.2, its value is 0.65 % for uranium-235, 0.216 % for plutonium-239 and 0.269 % for uranium-233, respectively. In MSBR which uses thorium, the effective value of β becomes 0.13 % (which is half the value of uranium-233) due to loss by circulation of fuel salt. The effect of loss of delayed neutrons by circulation is not large for MSRE having a relatively short pipe length outside the core, thus, the effective β value for uranium-233 was about 0.3 % in MSRE.

Delayed neutrons strongly affect nuclear safety. Prompt jump is enlarged by $\beta / (\beta - \rho_0)$, thus it becomes large for small β values. In addition, delayed transition is proportional to $1/\beta$, and therefore it is large for small β. Consideration of β is extremely important for the design of MSR, since prompt super-criticality is caused when $\rho_0 > \beta$. There are several reports regarding this issue after the re-evaluation of MSR since year 2000 [31, 32].

Concern “d” is also important for the safety analysis of MSR. There are several factors that influence reactivity in MSR. Flibe (which contains BeF_2) was proposed as fuel salt for MSBR, and many designs after ORNL activities adopted this composition. Be is added to lower the melting point and reduce the viscosity of LiF which is the main part of fuel salt. Be also acts as a neutron breeder. Solubility of uranium in Flibe is sufficiently high. However, if some oxide such as H_2O is mixed in the fuel salt, UO_2 or PaO_2 precipitates depending on the redox potential governed by ratio of U^{4+}/U^{3+} [25]. There is a likelihood that the precipitated uranium or protactinium deposits in some part of the primary circuit and is removed by fuel salt flow so as to suddenly add large positive reactivity in the core. It is necessary to develop a redox potential control method, albeit a method utilizing supplemental Be was developed previously.

Temperature reactivity coefficient is one of the most important indexes to evaluate the safety of a nuclear reactor. MSR has a general propensity of having a negative temperature reactivity coefficient due to its liquid fuel. The expansion of liquid fuel with increase in temperature, decreases the density of fissile material and salt moderator, thereby reducing both the deceleration of neutrons and fission reaction. MSBE used also graphite moderator, since fuel salt’s moderation ability of neutrons was not sufficient. There is subtle expansion of graphite moderator by increase of temperature. The temperature reactivity coefficient of MSR using a graphite moderator, is a sum of calculated values for fuel salt and graphite moderator. Fuel salt tends to have a negative value while graphite has a positive value. In the case of having fertile in the fuel salt, expansion of fuel salt has an effect of increasing the reactivity since density of fertile (which acts as neutron absorber) decreases. This effect appears more pronounced for thorium, owing to its large neutron absorption cross-section.

Temperature reactivity coefficient is affected by the volume ratio of fuel salt and graphite. That is to say, reactivity becomes positive for a small volume ratio of fuel salt against graphite. The difference of volume ratio between the core region and blanket region of MSBR shown in Section 4.3 indicates this tendency. In a case of large loss of fuel salt from the primary circuit, fission reaction stops because a large portion of fuel is lost. However, in a case of small leakage, fission reaction is sustained because fuel salt is slowly lost from the reactor core. Once fuel salt is gradually lost from the core, the volume ratio of fuel salt atop the core becomes small. Thus, it is possible to have a positive temperature reactivity coefficient locally. It has been pointed out that this kind of unique characteristics of reactivity of MSR should be re-evaluated in future, as there was uncertainty of nuclear data and so on during R&D activities of MSBR [33].

Several examples of temperature reactivity coefficient of MSBR are shown in Table 1. As can be seen from here, the value of α_{total} is negative for ORNL’s evaluation. The result obtained during re-evaluation of MSBR by GIF, in the early 2000s, shows also a negative value for an initial composition of fuel salt. However, it becomes positive for an equilibrium composition. These results surprised researchers of MSR, and a large red cross-mark was added on the illustration of graphite moderator of MSR in the website of GIF [18]. Based on this result, France is proposing a fast spectrum molten-salt breeder devoid of graphite moderator (Section 6).

Table 1 Temperature Reactivity Coefficient

Coefficient (pcm/^{o}C)	$\alpha_{doppler}$	$\alpha_{dilatation}$	α_{salt}	$\alpha_{graphite}$	α_{total}
ORNL value	-4.1	+0.82	-3.28	+2.35	-0.87
EDF Apollo 2 (Salt at equilibrium)	-2.67	+0.99	-1.68	+2.09	+0.41
EDF Apollo 2 (^{232}Th-^{233}U salt)	-3	+0.18	-2.82	+2.36	-0.46

Concern “e” is the same as what is discussed in Section 4.2. This problem does not happen in the case of ARE which does not use lithium in its fuel salt. However, most of the proposed designs of MSR after ORNL utilize lithium in their salt.

The general configuration of an MSR core is significantly different from that of a solid fuel reactor, such as LWR, in which many fuel rods can be recognized as heaters located at fixed position and the heat is removed by fluid. Criticality tendency can be obtained by calculating the dispersion equation;

nonetheless, the basis of this equation (such as distribution of fissile, precursors and fertile) move along with flowing fuel salt. This movement is calculated by heat generation based on neutron flux distribution. In short, criticality, heat generation and flow distribution are related strongly with each other. There is no combined calculation code for these aspects but there are several trials [34, 35].

6 R&D activities after ORNL work

Furukawa *et al.* proposed FUJI in the 1980s [36]. In the concept of FUJI, conversion ratio is targeted to be 1.0. FUJI is not a breeder but a converter, and thus it does not use online reprocessing and graphite moderator is not replaced. FUJI has a reflector, which was not used in MSRE. Though its output electric power is small (200MW), the concept is nearly the same as that of the fixed moderator MSR, which was discussed at ORNL [26]. Therefore, FUJI is still plagued with the same problems mentioned in Section 5. Temperature reactivity coefficient can be negative for a small-FUJI, but the value should be re-evaluated for a super-FUJI having 1GWe [37, 38]. Mini-FUJI, having a small capacity of 10MWe, is a demonstrator of FUJI, and thus its system is not optimized for small MSRs. For instance, it incorporates a secondary circuit and steam cycle, which are not used in WAMSR and UNOMI (mentioned later), thus the cost of mini-FUJI will be high due to its complexity.

Nishibori, who directed Japan Atomic Energy Research Institute from 1958 to 1964, also recommended thorium MSR. Nishibori recognized that problems of MSRs are mainly caused by the heat exchanger located outside the reactor vessel, and proposed his own design having a primary circuit including heat exchanger inside of the reactor vessel [39]. There is no possibility of leakage of fuel salt from pipes in alternating primary circuit with gaps of reflector and inner walls of reactor vessel. Although the system configuration is akin to ART, the reactor core is not a simple annulus channel but rather a combination of multiple cylindrical channels which is similar to that of MSRE. Reactor size is kept small by limiting the thermal output to 10~100MW so as to minimize loss of delayed neutrons. However, it is necessary to use many narrow tubes for internal heat exchanger, thus there is still a remaining possibility of corrosion for this heat exchanger. There are similar proposals from France [40, 41]. Outline of this design is shown in Fig. 7.

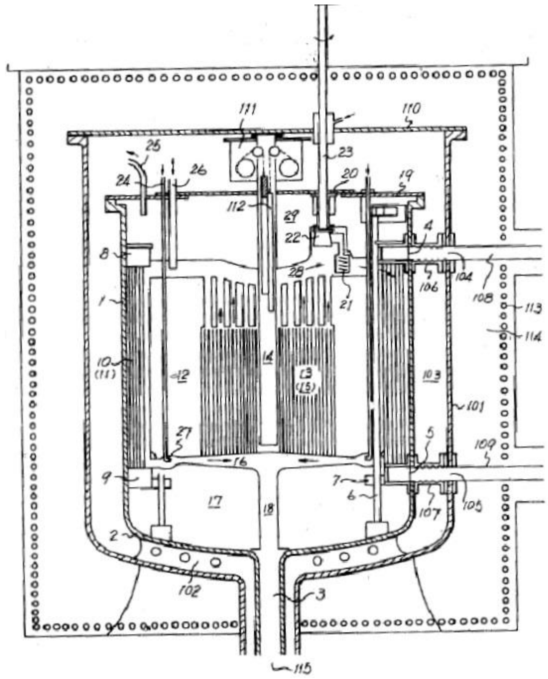


Fig. 7 Small MSR designed by Nishibori.

MSR is one of the candidates of generation IV reactors. During the re-evaluation process of MSBR's safety features with latest nuclear data and calculation code, French people revealed that MSBR has a positive temperature reactivity coefficient. Further, they proposed molten-salt fast reactor (MSFR) which does not use solid moderator in the core in order to attain a large negative temperature reactivity coefficient [42]. In this design, blanket salt, which is separated from fuel salt by a metal pipe, is located around the peripheral of the reactor core to achieve a high breeding ratio. This configuration faces a problem relating to corrosion resistivity of separator, as mentioned. Moreover, MSFR must have a large core in order to minimize neutron leakage, thus, thermal output of the reactor becomes large. It is therefore necessary to have external heat exchangers. The core of MSFR is surrounded by a metallic reflector and the primary circuit is contained in a reactor vessel. Thus the heat exchanger is termed as "internal" heat exchanger. Outline of this design is shown in Fig. 8.

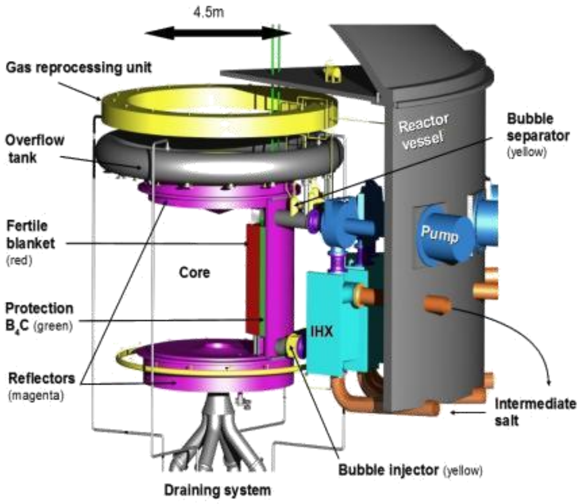


Fig. 8 Configuration of MSFR.

MSFR is suitable for incineration of transuranic (TRU), since MSFR utilizes fast spectrum neutrons. BeF_2 is added in the fuel salt of MSBR but it limits the solubility of Pu to be lower than about 2% [43]. Therefore, MSFR demands a fuel salt composition which does not use Be in order to increase the solubility of PuF_3, even though melting point of fuel salt is high. Russia also has proposed a similar concept named MOSART (Molten Salt Actinide Recycler & Transmuter) [44]. Recently, Hirose has proposed a TRU burner for processing Japan's spent nuclear fuel [45]. Since this is a burner, there is no blanket such as that used in MSFR and thorium is not contained in the fuel salt. TRU is used as fissile, thus, FLiNaK (LiF-NaF-KF: 46.5-11.5-42 mol%) is used to bypass the problem of TRU's solubility.

Sekimoto's proposal is also a fast molten-salt reactor in order to avail a large negative temperature reactivity coefficient. His design adopts a pool-type reactor vessel. The purpose of using a pool-type vessel is to avoid leakage of fuel salt from primary circuit which is usually adopted in most MSR designs such as MSFR. Due to its large thermal output power, a heat exchanger is needed. The heat exchanger is dipped in the fuel salt pool, a concept similar to that applied in ART. It is generally said that one of the advantages of MSR is its small fuel inventory. However, fuel inventory in the Sekimoto's design is large. Since this is a fast reactor, chloride salt is used instead of fluoride, but there is no corrosion-resistive structure material. Outline of this design is shown in Fig. 9 [46].

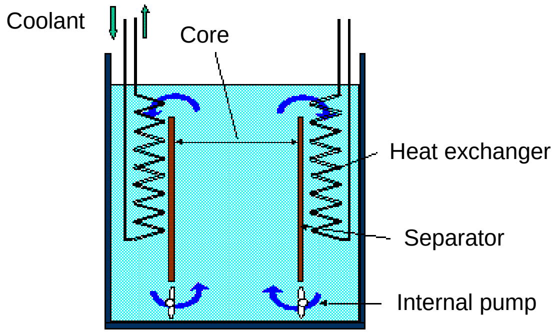


Fig. 9 Schematic design of Sekimoto's MSR.

LeBlanc proposed a two-fluid two-region MSR named "Tube-In-Tube" for breeder reactor, as shown in Fig. 10 [47]. He, however, has already terminated the study of this design, since there is no suitable material which can be used as separator.

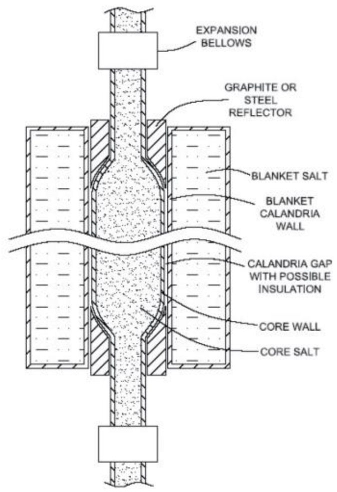


Fig. 10 "Tube-in-tube"-type MSR by LeBlanc.

LeBlanc is now designing an orthodox graphite-moderated one-fluid MSR [48]. The concept is shown in Fig. 11. In this design, breeding ratio is not a focal point. Thermal output power ranges from 60 to 650 MW. Reactor core is located at the lower part of the containing vessel, and heat exchanger is located at the top of the core region. Fuel salt flows down from the heat exchanger and reaches the bottom of the core after generating heat at the core part. The fuel salt then flows upward in the gap formed by the core separator and containing vessel. This configuration is essentially similar to that of ART, except the location of heat exchanger. Therefore, this new design also is encumbered with the common problem of corrosion resistivity of pipes for heat exchanger.

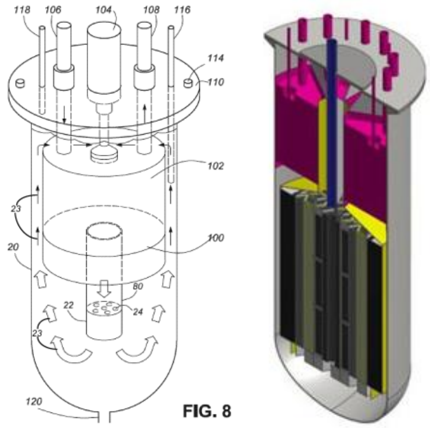


Fig. 11 LeBlanc's design of MSR.

Massie and Dewan proposed a WAMSR (Waste-Annihilating Molten Salt Reactor) for incineration of spent nuclear fuel [49] and established a new company named "Transatomic Power". WAMSR is a small reactor having 100 MW of thermal output. Its system configuration is shown in Fig. 12. WAMSR is a thermal spectrum reactor using $ZrH_{1.8}$ as moderator. Thermal energy is removed by an external heat exchanger. Gas-Brayton cycle is adopted. Although there is no intermediate circuit, concerns "a, b, e" mentioned in Section 5 still have to be considered.

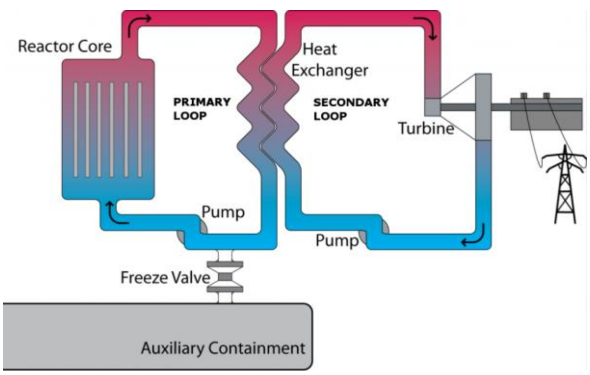


Fig.12 Configuration of WAMSR.

China officially announced that they started the development of thorium MSR in January 2011 [50]. China, at first, developed an orthodox MSR based on the technical background of ORNL's MSRE, but conducted tests both adding thorium and steam cycle which were not demonstrated in MSRE. They are going to build a 2MWth experimental reactor in 2015, a 10MW pilot plant in 2020, and finally commercialize a 100MW reactor in 2030. These designs do not use online reprocessing and are not breeders. They also started additional program to develop fluoride salt- cooled high-temperature reactor (FHR) in 2012 [51].

It is well-known that India has focused on thorium utilization. Since thorium does not contain fissile, India takes three stages for developing thorium nuclear power. In the first stage, natural uranium is used in heavy water reactors to produce plutonium. This plutonium is used in fast breeder reactors with thorium blanket to produce uranium-233. This uranium-233 is used in advanced heavy water reactors in the third stage with thorium to complete a closed thorium-uranium-233 fuel cycle. MSR has already been included amongst the candidates of third stage's reactor. They organized a thorium MSR conference named "CMSNT" in January 2013 [52].

The author is developing a transportable thorium MSR named "UNOMI (Universally Operatable Molten-Salt Reactor Integrated)" [53]. Its system configuration is shown in Fig. 13.

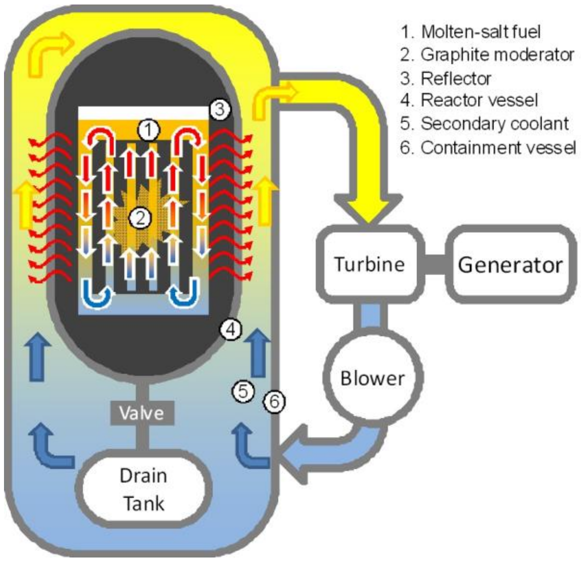


Fig. 13 System configuration of UNOMI.

The total system including both reactor core and heat exchanger must be small to enable transportation, thus thermal output is kept at a few MW. To achieve a small dimension of the reactor, fast spectrum cannot be used. A graphite moderator and reflector are used for thermal spectrum. Since the thermal output power is small, the generated heat is removed from the surface of the reactor vessel without using primary circuit pipes requiring fuel salt flow outside the reactor core. Gas-Brayton cycle will be adopted. The elimination of primary piping enables this reactor to serve as a "sealed radioactive source" to solve concern "a". Thus, FP does not circulate outside of the core (corresponding to concern "b"). Loss of delayed neutrons can be minimized (concern "c") and temperature reactivity coefficient has a negative value (concern "d"), owing to its small size (same as MSRE). Tritium is produced if lithium is used as component of fuel salt; however, it does not permeate the reactor vessel because the reactor vessel is covered inside by graphite reflector (solving concern "e"). It can be clearly seen that this design addresses all the problems listed in Section 5. Detailed works of thermo-hydraulic and neutronics are underway and will be published in near future.

UNOMI is not a breeder, thus online reprocessing is not used. Uranium-233 is produced outside of the UNOMI by using Mitrailleuse accelerator based on D-Be reaction [54].

7 Conclusion

In this paper, the concept of thorium MSR, its problems and several designs were introduced. Recently, there are several new activities on thorium MSR, but they should recognize the existing problems and address them based on scientific discussion. For example, a hearing was held on 9th May, 2013 at Japan Atomic Energy Commission regarding thorium MSR [56]. Addressed at the hearing were only the advantages, but hardly any description about related problems, as explained here, were presented. It is then not surprising that Chairman Kondo gave this comment: "it is not realistic to repeat fundamentalist-like discussions" at the end of the hearing.

In spite of this kind of unscientific movement, thorium MSR itself has potential as a safe nuclear power. Fukushima accident revealed that LWR has a possibility to release radioactive materials inside the reactor core to the environment. In the case of destruction of reactor vessels, fuel salt mostly maintains radioactive materials (including cesium which is a salt-seeker) inside of its bulk except for some volatile element. Material dispersion tests for reactor vessel explosion will be carried out during collaborative development of UNOMI at Kazakhstan by using non-radioactive isotope. Thus, the significance of developing thorium MSR is still large. Although it is necessary to consider the source of fissile and disposal of radioactive waste for commercialization of thorium MSR, it is not mentioned here for brevity reasons. A comprehensive research and development including the front-end and back-end processes will be required.

Nomenclature

ARE	Aircraft Reactor Experiments
ART	Aircraft Reactor Test program
CR	Conversion Ratio
FHR	Fluoride salt-cooled High-temperature Reactor
FP	Fission Products
GIF	Generation IV International Forum
ICBM	InterContinental Ballistic Missile
LWR	Light-Water Reactor
MOSART	MOlten Salt Actinide Recycler & Transmuter
MSBR	Molten-Salt Breeder Reactor
MSR	Molten-Salt Reactor
MSRE	Molten-Salt Reactor Experiment
ORNL	Oak-Ridge National Laboratory
R&D	Research and Development
SMR	Small Modular Reactor
UNOMI	UNiversally Operatable Molten-salt reactor Integrated
WAMSR	Waste-Annihilating Molten Salt Reactor

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