

Thorium Molten Salt Nuclear Energy Synergetic System (THORIMS-NES).

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Abstract

The authors have been promoting nuclear energy technology based on thorium molten salt as Thorium Molten Salt Nuclear Energy Synergetic System (THORIMS-NES). This system is a combination of fission power reactor of Molten Salt Reactor (MSR), and Accelerator Molten Salt Breeder (AMSB) for production of fissile ^{233}U , with connecting chemical processing facility.

In this paper, concept of THORIMS-NES, advantages of thorium and molten salt, recent MSR design results such as FUJI-U3 using ^{233}U fuel, FUJI-Pu, large sized super-FUJI, pilot plant miniFUJI, AMSB, and chemical processing facility are described.

1. Introduction

After the 21st century began, the world is facing rapid economic growth mostly in Asia and in Africa. One of the reasons is an increase of population, and another reason will be modernization or industrialization in these countries. As a result of this fact, the world energy demand is increasing very rapidly. Meanwhile, environmental issues will become crucial.

One of the solutions is considered as a nuclear technology. Based on a hypothetical prediction, large amount of nuclear energy as high as 10 TWe (:10,000 plants of 1 GWe size) will be required in 21st century [1].

But, in March 11, 2011, severe accidents occurred at Fukushima in Japan. A huge earthquake and tsunami struck 6 reactors at Fukushima site, and No.1 through No.4 reactors had severe damages due to loss of electricity and cooling systems. Therefore, safety issue in nuclear technology becomes more important than before.

Looking back the history, after the World War II, application of nuclear energy to power generation has started. Many different types of reactors were discussed at Oak Ridge National Laboratory (ORNL). One of them was a Molten Salt Reactor (MSR). The first experimental MSR was Aircraft Reactor Experiment (ARE) in 1954. After the success of ARE, the second experimental MSR, which was named Molten Salt Reactor Experiment (MSRE), showed extremely successful operation from 1965-1969 at ORNL. Based on MSRE experience, ORNL issued a conceptual design report for a 1,000MWe (2,250 MWt) large sized power generating plant in 1971 [2], which was called Molten Salt Breeder Reactor (MSBR). This MSBR was planned to use 700 deg.-C molten salt of $^7\text{LiF-BeF}_2\text{-ThF}_4\text{-UF}_4$ (72%-16-12-0.4), aiming to achieve a Breeding Ratio (BR) of 1.06 by two important assumptions. One is on-line reprocessing, and another is replacing graphite at every 4-years. Alvin Weinberg conducted these developments as a director of ORNL for almost 20 years [3]. But, the development plan of MSBR was cancelled in 1970s.

2. Synergetic Nuclear System: THORIMS-NES

In this paper, we propose and describe Thorium Molten Salt Nuclear Energy Synergetic System (THORIMS-NES), which is a combination of fission power reactor of Molten Salt Reactor (MSR), and Accelerator Molten Salt Breeder (AMSB) for production of fissile ^{233}U . In between these two facilities, fuel chemical processing facility is placed. AMSB is similar to Accelerator Driven subcritical System (ADS). AMSB utilizes a high-intensity proton accelerator with an energy of about 1 GeV, directed to a spallation target made of molten salt of thorium (Th), and converts Th to fissile ^{233}U . Fusion technology might someday become available to produce the needed ^{233}U fuel.

THORIMS-NES has the following three unique features.

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| <ol style="list-style-type: none">1) Using Th instead of U as fertile element, that is Th cycle instead of U-Pu cycle.2) Using liquid fuel (molten fluoride salt fuel) instead of solid fuel.3) Separation of electric power station (MSR-FUJI) and fissile breeding facility (AMSB). |
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The reasons why these features are applied are explained below, and this concept is also described in K. Furukawa's several papers [1] [4] [5] [6] [7] [8].

THORIMS-NES concept includes a plan as follows. First stage is a construction of mini-FUJI, which is a 7 MWe small sized demonstration pilot plant. This mini-FUJI is based on 1960s' MSRE experience at ORNL, and can be developed in a short time. Second stage is a construction of FUJI reactor, which is typically a 100-200 MWe MSR. Third stage is an establishment of Regional Center, which is composed of chemical processing facility and AMSB for ^{233}U production.

2.1 Why thorium?

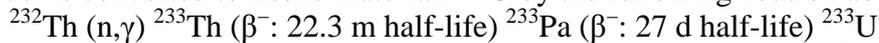
2.1.1 Abundant resource

Thorium (Th) is geo-chemically 3-4 times more abundant than uranium (U) in the earth crust. Th co-exists with rare-earth elements in most cases, and they are produced in most countries. Reasonably Assured Reserves (RAR) category of Th is less than 3 million tons. This number is smaller than expected, because Th was not explored as U. Based on the estimate that U resource is about 5 million tons, Th resource would be 15-20 million tons.

Since 1 GWe MSR with 75% load factor consumes 0.7 ton of Th per year [9], required Th resource to operate 10,000 MSRs of 1 GWe is 7,000 tons per year. Therefore, the above estimated amount of Th would be enough to operate 10,000 MSRs of 1 GWe for more than 2,000 years.

2.1.2 Small production of plutonium and Minor Actinides

Natural Th is composed of only one nuclide ^{232}Th , except about 10ppm ^{230}Th which is rich in Th from U-ores. Chemically refined Th is added directly to molten salt. Then, fertile material: ^{232}Th in reactor fuel is converted to fissile material: ^{233}U by the following neutron absorption reaction:



Repeating successive neutron absorptions, ^{233}U is converted to ^{234}U , ^{235}U , ^{236}U , ^{237}Np , ^{238}Pu , and finally to ^{239}Pu . Since this is a long process, production of Pu is very small compared with U fuel case.

This small production of Pu makes Th-fueled reactor a nuclear weapon proliferation-resistant technology. Also, production of long-lived trans-uranium elements (TRU, or Minor Actinides: MA) such as Np (neptunium), Cm (curium) and Am (americium) is very small, because these isotopes are mostly produced by neutron absorption of Pu isotopes. Actual data for production of Pu and MAs are shown in Section-3.

2.1.3 Proliferation resistance

Nuclear proliferation or nuclear terrorism is one of the most crucial issues in the world. Of course, since ^{233}U is a fissile material, if pure ^{233}U can be collected from MSR, it may be possible to use ^{233}U as a nuclear weapon. But, ^{233}U fuel is accompanied by ^{232}U , which is produced by several neutron reactions. And, strong gamma activity is accompanied by daughters of ^{232}U , such as 2.6 MeV gamma emission from ^{208}Tl (thallium) and 1.8 MeV gamma emission from ^{212}Bi (bismuth). This fact makes handling of ^{233}U very difficult, and also makes it easy to detect illegal storage or transportation of ^{233}U .

2.2 Why liquid fuel? (Why molten salt fuel?)

Coming back to the history of MSR, Eugene Wigner, who was the first research director of ORNL

and the Nobel Prize winner in 1963, gave a prospect that the reactor should be a “Chemical Engineering Device” using liquid fuel because of its simplicity, and not a “Mechanical Engineering Device” using solid fuel.

Based on Wigner’s suggestion, ORNL people investigated candidates for liquid fuel, and they finally reached the conclusion that molten salt is a best solution. This is because molten salt is chemically stable and inert, and especially it can be used at very low pressure owing to its very high boiling temperature and very low vapor pressure, also it can dissolve most nuclear materials such as U and Th. Since most molten salts have high melting temperature, if leakage of molten salt occurs, it will freeze and do not release radioactive materials.

Among many molten salts, they finally proposed an eutectic mixture of lithium fluoride and beryllium fluoride called “FLiBe” (LiF-BeF_2), with fertile Th and fissile U/Pu dissolved as molten fluoride salt. This liquid serves as fuel element, as heat transfer medium, and as fuel processing medium. Each of these functions is described in the following sections.

2.2.1 Molten salt as liquid fuel element

In MSR, fissionable isotopes, fertile isotopes, most Fission Products (FPs) and heavy elements reside as ionic elements dissolved in molten salt. When fission reaction occurs in the core, molten salt is heated and it circulates through pumps, heat exchangers and then returns back to the core. Fission reaction occurs only in the core, because the reactor is only critical when graphite exists as a neutron moderator.

For solid fuel reactor, there is a mechanical limitation due to radiation damage on fuel pellet and cladding tube, internal pressure due to accumulated gaseous FPs, fuel pellet swelling, outer oxidation of cladding tube, and so on. Also, there is another limitation due to depletion of fissile material. Due to these effects, periodic replacement of fuels is required for solid fuel reactor.

But in molten salt fuel, there is no radiation damage, because it is an ionic liquid and not affected by neutron or gamma radiation. Also, there is no requirement to replace molten salt. It is easy to adjust molten salt composition by adding molten salt containing fissile U/Pu or fertile Th in order to maintain an optimum fuel composition.

An important advantage of a liquid fuel is that radioactive gasses produced as FP can be removed from liquid fuel. In solid fuels, radioactive gasses such as xenon and krypton are accumulated within cladding tubes, and they are released at severe accident condition such as Fukushima accident.

Among these nuclides, ^{135}Xe has a very large neutron absorption cross-section, and it deteriorates neutron economy at normal operation. Also, xenon oscillation issue must be considered for PWRs.

But, in liquid fuels, these gasses are insoluble in molten salt. Therefore, they can be removed from molten salt, and captured by a charcoal filter system, then stored for enough decay periods. Especially, removal of ^{135}Xe makes load-following capability easier, and also removes the above xenon oscillation issue. In order to accelerate removal of radioactive gasses, a helium (He) gas bubble injection system was proposed at MSBR design, and this system collects radioactive gasses more efficiently.

Molten salt can dissolve fissile/fertile materials. It is chemically stable and inert, and especially it can be used at very low pressure owing to its very high boiling temperature and very low vapor pressure. Normal operating pressure for MSR is 0.5 MPascal (5 atm), which is very low compared with 15 MPa (about 150 atm) for PWR. This low pressure reduces the danger of reactor vessel break or pipe break due to high pressure such as is LWR case. Higher temperature brings higher thermal efficiency of the plant. Outlet temperature of MSR is typically as high as 700 deg.-C, compared with 330 deg.-C of PWR case. Liquid fuel design does not need fuel manufacturing such as solid fuel case. These features make reactor design simple and economical.

ORNL made a choice of a fuel salt based on $^7\text{LiF-BeF}_2$ (FLiBe), and $\text{NaBF}_4\text{-NaF}$ as a secondary coolant. Properties of these molten salts are summarized in ORNL report [10]. Recent information

is shown in another document, where it is described that molten fluoride salts are intrinsically not corrosive to constituent elements in containment alloys, primarily because of greater thermodynamic stability of molten fluoride salt constituents as compared with fluoride of transition metals [11]. Compatibility to structure materials is explained later.

Fuel salt of FLiBe has an appropriate melting temperature and viscosity. Also, it has a very high boiling temperature of 1,430 deg.-C.

From nuclear standpoint, FLiBe has a small neutron absorption cross-section, but in order to improve more, natural Li is enriched to ^7Li , because natural Li includes 7.5% of ^6Li of large neutron absorption cross-section.

2.2.2 Molten salt as heat transfer medium

Thermo-physical properties of molten FLiBe indicate that FLiBe has significant and acceptable characteristics as a heat transfer medium. Such excellent characteristics are based on (1) low pressure, (2) high boiling temperature, (3) high heat capacity owing to small ions, (4) appropriate viscosity, and (5) suitable Prandtl number. The Prandtl number of FLiBe is 10-20 in temperature range of 700-500 deg.-C, which is very high than liquid Na of 0.004, and it means that FLiBe is a relatively adiabatic liquid. This fact mitigates a thermal shock for quick and inadvertent temperature change.

2.2.3 Molten salt as fuel processing medium.

Today, most spent fuel reprocessing is a hydrometallurgical process called PUREX (Plutonium and Uranium Recovery by EXtraction). This process uses organic solvent extraction from aqueous solutions, which result from dissolution of spent fuel. On the other hand, recent process, which does not use water or organic liquids, is called Pyro-processing or Dry-processing. Since this process does not use water or organic liquids, there is less danger of critical accident, and can be more compact than PUREX plant.

In THORIMS-NES concept, reprocessing medium is FLiBe, which is the same medium of MSR fuel salt. Dry-processing technology for molten salt fuel has been proposed and developed at MSBR development stage in 1970s, and later in European countries where they tried to apply this technology starting from solid fuels. This history is described below.

In 1970s, ORNL proposed on-line fuel processing of fuel salt. This system consists of two parts: (1) removal of uranium (U) and protactinium (Pa) from fuel salt and reintroduction of U to fuel salt, and (2) removal of rare-earth fission products (FPs) from fuel salt [2].

Although THORIMS-NES does not adopt on-line processing, its basic concept can be applied. The reasons why THORIMS-NES eliminates continuous chemical processing of molten salt are because (1) we have to deploy large number of reactors and we need highest simplification for these reactors, and (2) we adopt AMSB instead of reactors for producing fissile fuel. Chemical processing of spent fuel from MSR is considered in THORIMS-NES concept as a separate operation from reactor operation. Fuel processing at MSR during power operation is limited to removal of gaseous FPs.

In 1980s, Czechoslovakia and former Soviet Union jointly developed a reprocessing technology, called "FREGAT". This technology is based on a fluorination flame reactor technology used in France to produce UF_6 gas for uranium enrichment from uranium oxide.

FREGAT technology is applying a fluoride volatility process based on separation of U and Pu in the form of volatile fluoride from FPs, the majority of which are non-volatile fluorides. This process was originally developed to reprocess spent fuels of FBR, but it can be applied to LWR fuels [12].

As a conclusion, molten salt, especially FLiBe, has many excellent features such as very small neutron absorption cross-section, no radiation damage, large heat-capacity, low vapor pressure, single phase liquid, appropriate melting temperature, high boiling temperature, chemically inertness, good compatibility with structural material, compatibility with graphite, high solubility for

fissile/fertile and FPs, no-solubility for gaseous FPs such as Xe and Kr, and very limited radioactivity release in accident.

2.3 Why separation of reactor and fuel breeder

At the early stage of nuclear power development, a breeder reactor was proposed, which produces not only electricity but also produces more fissile fuel than it consumes. A breeding ratio (BR), that is a ratio of produced fissile amount against consumed fissile amount, becomes higher than 1.0, especially higher at fast neutron spectrum. Therefore, a Fast Breeder Reactor (FBR) cooled by liquid metal such as liquid sodium was developed since then. But, BR is not high enough to supply the required fissile fuel to a large amount of reactors, because one fission reaction produces only 2 to 3 neutrons, and an excess neutron is not enough.

Therefore, AMSB is required to produce enough fissile fuel, because AMSB utilizes spallation reaction, which produces 20 to 30 neutrons/protons by one reaction.

On the other hand, AMSB has two weaknesses. One is that spallation reaction produces about 1/10th energy of fission reaction. Another one is that continuous operation of accelerator is very difficult, this means AMSB is not appropriate for power station, but acceptable as a fissile producer plant which does not require continuous operation. Also, AMSB is a high technology system, and requires a long-time to develop. But, at the initial stage of THORIMS-NES, Pu from LWR spent fuel can be used as initial start-up fissile of MSR-FUJI, and a long-time development of AMSB is acceptable. Fusion technology might someday become available, and be an even better fissile producer.

As a conclusion, a fission reactor is energy rich but neutron poor, and an AMSB is neutron rich but energy poor, and then, these two functions are separated.

3. Molten Salt Power Reactor FUJI

3.1 FUJI-U3 (using ^{233}U as fissile)

Conceptual design of FUJI has started in late 1980s based on ORNL studies, and its latest design is shown in references [9] [13].

FUJI is size-flexible, but typical value is 100-200 MWe. The latest design: FUJI-U3 is 200 MWe and thermal output of 450 MWt. This means a thermal efficiency of about 44% owing to its high exit temperature, compared with LWR case of 33%.

Schematic diagram of MSR-FUJI is shown in Figure-1. A reactor vessel and pipes are made of Hastelloy N. There is a graphite moderator within a reactor vessel. Fuel salt flows upwards by primary pumps, and then goes to heat exchangers to transfer the heat to secondary coolant salt. The heat of secondary salt is transferred to the steam through a supercritical steam generator, and then to a turbine/generator to produce electricity. Based on an optimized core design, the graphite moderator does not require replacement during a reactors' lifetime of 30 years.

Fuel salt of FUJI-U3 is ${}^7\text{LiF}\text{-BeF}_2\text{-ThF}_4\text{-UF}_4$ (69.78-18-12-0.22 mol%). Although fuel salt is based on ${}^7\text{LiF}\text{-BeF}_2$ (FLiBe), secondary salt is selected as $\text{NaBF}_4\text{-NaF}$ (92-8 mol%), same as MSBR design, because of cost performance. Both salts have good compatibility with Hastelloy N.

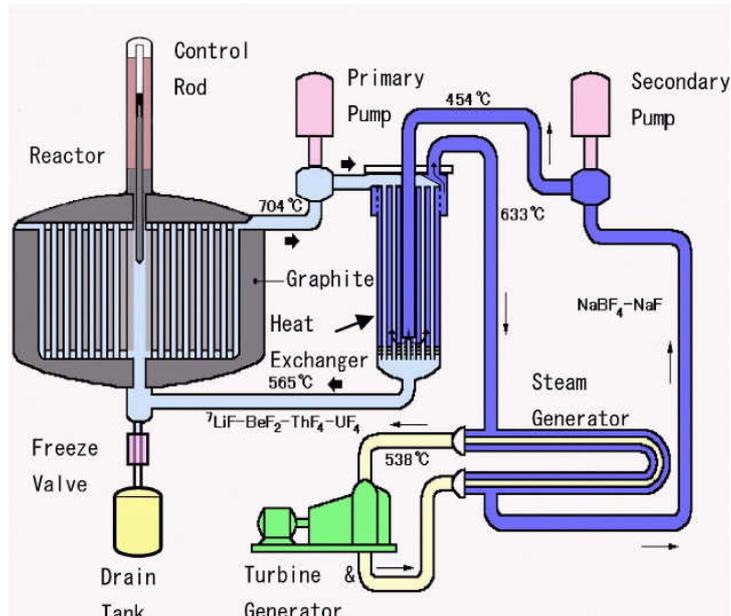


Figure-1. Schematic diagram of MSR-FUJI (One loop is shown,)

Vertical cross section of the primary system of FUJI is shown in Figure-2. If fuel salt is overheated, fuel salt goes down to a drain tank, which is located below the reactor, through a freeze valve. Since there is no graphite moderator in a drain tank, there is no danger of re-critical accident. Also, in case of pipe break, leaked fuel salt is collected to an emergency tank, which is also located below the reactor.

Continuous chemical processing of fuel salt is not performed in FUJI, but radioactive Xe, Kr and T(tritium) are continuously removed from the reactor not only to improve Conversion Ratio (CR), but also to prevent their release in a pipe break accident.

One of the design parameter is a graphite fraction within the core ($= \frac{\text{[graphite area]}}{\text{[graphite area + fuel path area]}}$, see Figure-4), and MSBR proposal was 0.87. But FUJI-U3 has a slightly lower value of 0.64 in order to achieve higher CR by harder neutron spectrum. Also, in order to improve CR more, all fuel salt is assumed to clean up at every 7.5 years operation.

As a result, total required fissile ^{233}U is 1.48 ton for 30 years operation with 75% load factor, and remaining fissile at the end of reactor life is 1.50 ton. This means $\text{CR}=1.01$, and therefore self-sustaining is achieved. Th is consumed 3.9 tons for 30 years operation. Owing to Th cycle, production of Pu and Minor Actinides (MA) are very small. Total production of Pu is 0.3 kg for 30 years operation, which is only 0.03% of the same sized Boiling Water Reactor (BWR). Also, production of MA is only 5.3 kg, which is only 5 % of the same sized BWR.

Bird-eye view of FUJI is shown in Figure-3. This reactor design has a three level containment safety, as is proposed in MSBR design. First level is a reactor vessel and pipes made of Hastelloy N. Second level is a high-temperature containment composed of three layers, which contains a reactor vessel, pipes, and heat exchangers. Third level is a reactor building composed of two layers.

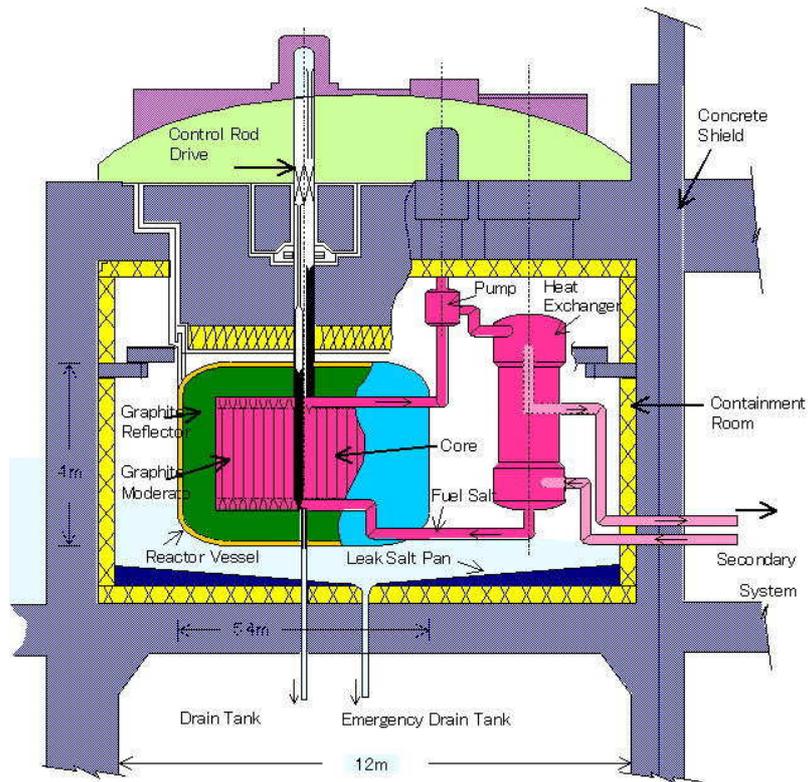


Figure-2. Cross section of the primary system of MSR-FUJI

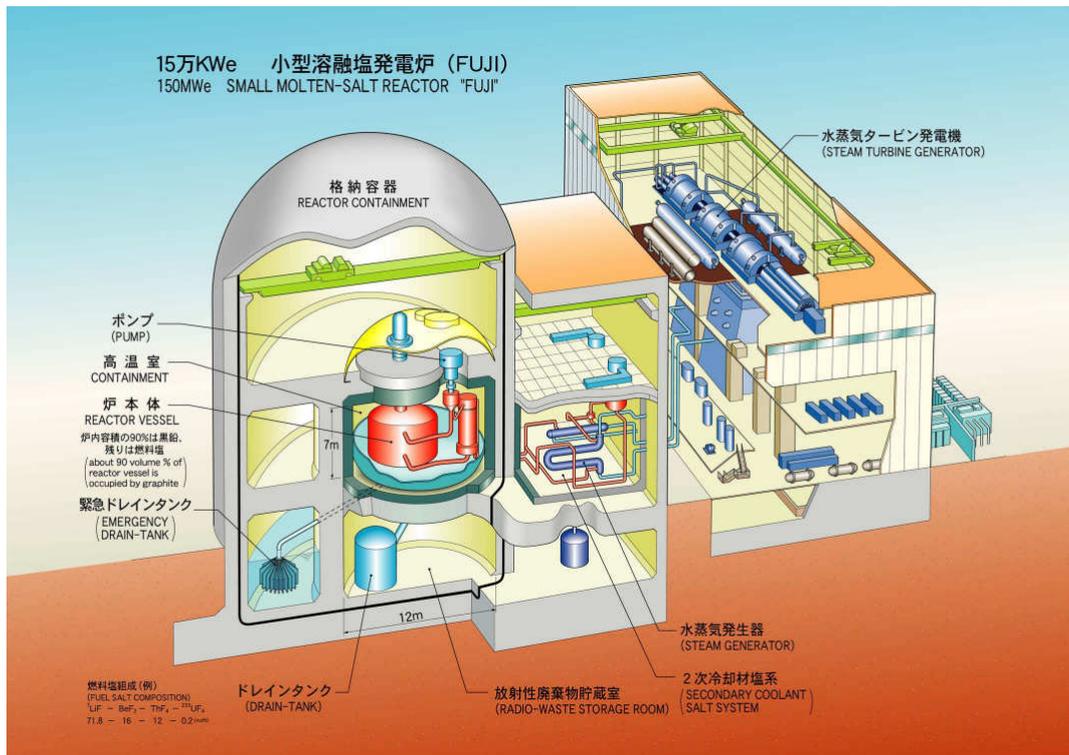


Figure-3. Bird-eye view of MSR-FUJI

3.2 FUJI-Pu (using Pu as initial fissile)

At the initial stage of Th cycle, MSR needs fissile material for initial startup, because natural Th does not contain ^{233}U . Before we establish AMSB for fissile production, one idea is to utilize Pu

from LWR spent fuel, Therefore, Pu burner version: FUJI-Pu was studied [14]. FUJI-Pu can be applied to incinerate recovered Pu from LWR spent fuels and convert Th to ^{233}U . Assuming 100 MW (electric) and 250 MW (thermal) output, and initial fuel composition of $^7\text{LiF}\text{-BeF}_2\text{-ThF}_4\text{-PuF}_3$ (71.8-16.0-12.0-0.25 mol%), FUJI-Pu can consume 244 kg of Pu-fissile, and produce 112 Kg of U-fissile for 900 days full power operation.

3.3 Transmutation of Minor Actinides by MSR-FUJI

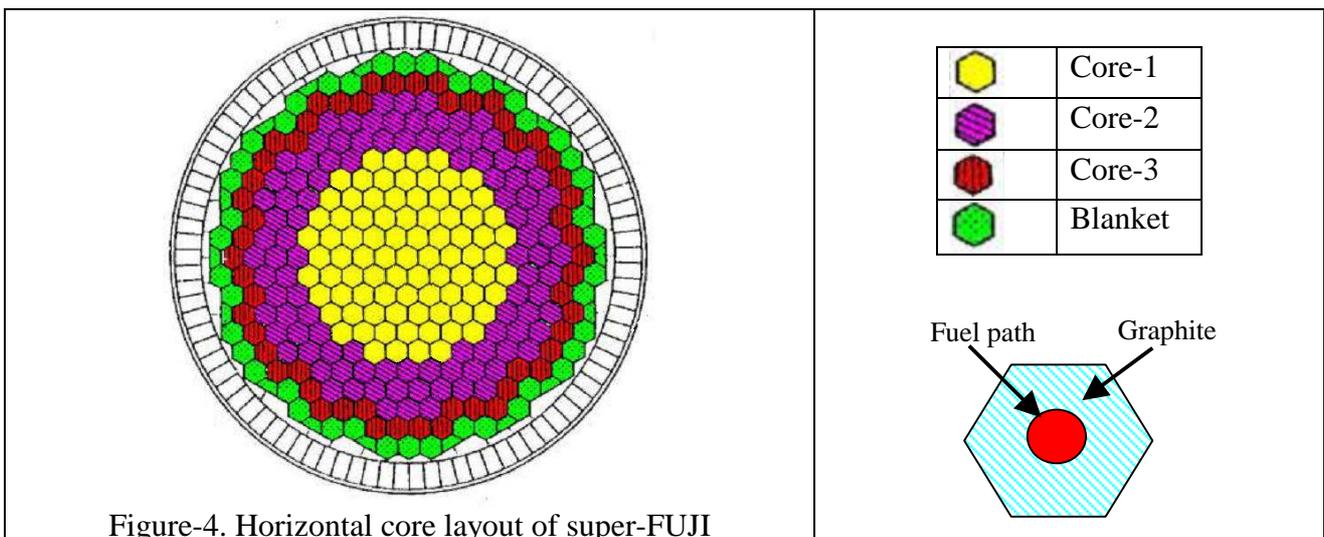
As is shown above, production of Minor Actinides (MA) in MSR-FUJI is very small. Since these MAs have certain absorption cross-sections for thermal neutrons, MAs from LWR spent fuel can be efficiently transmuted to stable or shorter lifetime elements. A preliminary study was performed on 1,000 MWe (2,250 MWt) FUJI case. Assuming totally loaded MA of 5,940 kg, it is calculated that 85% of MA is transmuted for 30 years operation. This means 170 kg MA is consumed yearly [15].

3.4 super-FUJI (large sized plant)

FUJI was originally designed as a relatively small sized plant, because it is aimed to deploy widely in the world. But, large sized plant as 1,000 MWe is also possible, as was proposed as 1,000 MWe MSBR by ORNL. Since MSR does not need a high-pressured vessel, it will be easier to design a large sized plant, of course, large sized equipments such as molten salt pump or graphite have to be developed.

A preliminary design was performed as “super-FUJI”, assuming 1,000 MW (electric) and 2,272 MW (thermal) output [16].

As is shown in Figure-4, the core is composed of graphite rods of hexagonal shape, and fuel salt passes through center holes of each graphite rods. Graphite fraction can be optimized by changing fuel path diameter, and the average graphite fraction is 0.87 same as MSBR. Initial fuel composition is $^7\text{LiF}\text{-BeF}_2\text{-ThF}_4\text{-}^{233}\text{UF}_4$ (71.8-16.0-12.0-0.2 mol%), and initial $^{233}\text{UF}_4$ inventory is 1.5 ton. This is a somewhat old design, but fissile inventory is minimized owing to higher graphite fraction and softer neutron spectrum. Initial Conversion Ratio (CR) was 0.98, and lifetime average CR will be lower than this value. In general, a large sized plant has better neutron economy, and CR can be improved. But, initial fissile inventory and CR are trade-offs in most cases.



3.5 mini-FUJI (pilot plant)

Almost all reactor engineering was established at ORNL for MSRE and MSBR design. But, it was 40 years ago, and technology has to be rebuilt by a pilot-plant “mini-FUJI”.

Also, there are two major items, which were not demonstrated in MSRE. One item is to use Th in fuel salt. Fuel salt of MSRE did not contain Th, but this will be easily demonstrated in mini-FUJI. Another issue is development of a steam-generator (SG), because the heat in secondary salt of MSRE was released to the open air. This SG is a new technology, but SG development experience for FBR can be applied. Table-1 shows basic design parameters for FUJI-U3 in Section 3-1, mini-FUJI [17] and MSRE [18].

Table-1. Design parameters of three MSR's (FUJI-U3, mini-FUJI and MSRE)

	FUJI-U3	mini-FUJI	MSRE
Electric output (Mwe)	200	7	-
Thermal output (MWt)	450	17	8
Thermal efficiency (%)	44	41	-
Graphite fraction (vol%)	55-73	70	77
Fuel conversion ratio	1.01	0.6	-
233U inventory (kg)	1,130	27	32
232Th (ton)	56	0.65	-
Fuel salt	⁷ LiF-BeF ₂ -ThF ₄ -UF ₄	⁷ LiF-BeF ₂ -ThF ₄ -UF ₄	⁷ LiF-BeF ₂ -ZrF ₄ -UF ₄
Secondary salt	NaBF ₄ -NaF	NaBF ₄ -NaF	LiF-BeF ₂
Total fuel salt volume (m ³)	39	4.5	2.1
Core flow rate (m ³ /min)	42	1.6	4.5
Temperature in-out (deg.-C)	565-704	565-704	632-654
Graphite inventory (ton)	163	8.8	3.7

3.6 Structural alloy: Hastelloy N

Structural alloy Hastelloy N (Nickel:71wt%, Molybdenum:16wt%, Chromium:7wt%, Iron:5wt%, others:1wt%) was used as a container material in MSRE. Its data is shown in supplier's web site [19]. Figure-5 shows a photo of a sample plate.

Corrosion of Hastelloy N was investigated using irradiated specimens in MSRE. As far as suitable purification is performed, there is no water or oxygen in molten fluoride salt. Therefore, Hastelloy N corrosion is very small. Estimated corrosion rate of Hastelloy N was reported by J. W. Koger as 0.02 mil/year [20].

Regardless of apparent success in MSRE, standard Hastelloy N exposed to fuel salt under irradiation revealed material embrittlement due to inter-granular attack, where grain boundaries were degraded due to existence of tellurium (Te) which was produced as a fission product, although not penetrating deeply. To solve this issue, a modified Hastelloy N was developed where Mo, Si and B are reduced and Nb (1-2%) is added, and another solution is to control a redox potential, that is a U⁴⁺/U³⁺ ratio. It was observed that this Te cracking becomes very small at a U⁴⁺/U³⁺ ratio is less than 60. This U⁴⁺/U³⁺ ratio can be easily controlled in a suitable range by varying dissolution of Be into fuel salt [21].

This modified Hastelloy N was proposed as a container material in MSBR, and this alloy has the same or better corrosion resistance as standard Hastelloy N [2].

In MSBR design, an anticipated service lifetime was 30 years, and the most highly irradiated portion of the reactor vessel would be exposed to a fast-neutron ($E > 0.1$ MeV) fluence of less than 1×10^{21} neutrons/cm² and a thermal neutron fluence of about 5×10^{22} neutrons/cm². Although

irradiation effect on Hastelloy N is small until these values, these values are considered as design limit for MSR-FUJI.



Figure-5. Hastelloy N equivalent (courtesy of MMC)

3.7 Graphite

Graphite is the only one component material within the reactor core, and it serves as a neutron moderator and a reflector. Since graphite is irradiated and is facing directly to fuel salt, there are several requirements.

First requirement is irradiation stability. When graphite is irradiated, it shows shrinkage at first, and then volume expansion (:swelling) begins. In MSBR design, fluence limit of fast neutrons ($E > 50$ keV) was proposed as 3×10^{22} neutrons/cm², where the volume returns back to an initial value [22]. This value is applied to MSR-FUJI design.

Second requirement is to prevent penetration of fuel salt into graphite. If fuel salt penetrates into graphite, fission reaction causes additional radiation damage to graphite. But, since molten salt does not wet graphite, this can be easily prevented by manufacturing a pore-diameter of graphite less than 1×10^{-6} m, owing to a surface tension of molten salt.

Third requirement is to prevent absorption of Xe and Kr to graphite. Especially, ¹³⁵Xe absorbs neutron due to its very large neutron absorption cross-section, and it deteriorates neutron economy. One solution is to seal graphite surface by pyro-carbon. Another solution is to apply a He-bubble injection system. This will collect Xe and Kr efficiently, and Xe/Kr absorption to graphite will not affect neutron economy.

3.8 Reactor chemistry

The chemistry of MSR, mostly related to molten salt, is summarized by ORNL [23], and many source documents are shown in its references.

Regarding behavior of Fission Products (FP), there are three types [8].

Group-1 is a noble FP gas such as Xe, Kr and T (tritium), which are essentially collected from molten salt by a gas-removal system. Although the path of T is complicated, recovery of T can be performed efficiently, as is described in K. Furukawa's paper [24].

Group-2 is soluble elements such as Rb/Cs/Sr/Ba/Y/Zr/Br/I/La and so on. Among them, Rb/Cs/Sr/Ba/Y have noble-gas precursors, which will be isolated to gas-phase, except several short half-life nuclides. Other elements stay in molten salt as fluoride.

Group-3 is noble and semi-noble FPs such as Ge/As/Nb/Mo/Ru/Rh/Pd/Ag/Cd/Sn/Sb and so on. Their amount is small, and they will exist in an elemental state, and will be collected by filters.

Total amount of concentration of three-valence elements such as Nb/Ce/Pr/La/Pm/Y/Gd should not exceed 1.3%, due to its solubility limitation [6].

As for the valence control for fuel salt from the standpoint of corrosion, it was described at Section-3.6.

3.9 Safety of MSR-FUJI

MSR-FUJI has a very high safety, and severe accidents are essentially impossible. These safety features are listed as follows, which are summarized from a detailed reference [7].

Some of major accidents were analyzed so far [25] [26] [27], and safety criteria and safety guidelines are being studied by the authors.

- (1) The primary and secondary loops are operated at a very low pressure (5 atm), which reduces the danger of system rupture or salt leak due to high pressure.
- (2) Molten salt is chemically inert and has no flammability.
- (3) Molten salt is an ionic liquid, and irradiation damage or fuel failure does not occur.
- (4) There is no possibility for pressure increase in the primary loop, because boiling temperature of fuel salt is very high (ca. 1400 deg.-C) compared with operating temperature (ca. 700 deg.-C). In addition to this, there is no water within the containment, and this fact eliminates accidents due to water evaporation.
- (5) Fuel salt is critical only where graphite exists in an appropriate fraction. In an accident, fuel salt exhausted from the core to a drain tank cannot cause re-criticality accident.
- (6) MSR-FUJI has a large negative reactivity coefficient on fuel salt temperature, which can suppress an abnormal change of reactor power. Heat capacity of graphite is large and temperature rise is slow; therefore, it is easy to control, even though temperature coefficient of graphite is positive.
- (7) Gaseous FPs such as Xe/Kr/T(tritium) can be effectively trapped or collected by activated charcoal filter or other system, and contained for enough decay time. This system decreases radioactivity release in case of severe accident.
- (8) Since fuel composition can be easily adjusted when necessary, an excess reactivity, which must be compensated by control rods, is small. Therefore, inadvertent reactivity insertion is small.
- (9) Delayed neutron fraction of ^{233}U is lower than that of ^{235}U , and some (*) of delayed neutrons are generated outside the core (*: for example, about 10% for FUJI-U3). However, safe control of the reactor is possible because of a large negative reactivity coefficient on fuel salt temperature, large prompt neutron lifetime, and small reactivity insertion.
- (10) There is no possibility of graphite fire. One reason is because there is no oxygen within the containment. Another reason is because there is no heat source near the graphite, after fuel salt is transferred to a drain tank or an emergency tank.

Based on an investigation report on Fukushima accident, which was a severe accident occurred on March 11 2011, we studied whether MSR is still safe against Fukushima type accident [28]. Its conclusion is referred as follows.

- (1) MSR is safe, in case of loss of all electricity and/or loss of all core cooling, because natural circulation cooling is possible. In addition, fuel salt is drained to a drain tank if required.
- (2) Drain tank is equipped with cooling system, which does not need electricity.
- (3) There is no fuel damage or fuel melt-down, owing to its intrinsic feature of molten salt.
- (4) There is no danger on reactor vessel or containment, because there is no overpressure possibility due to vaporization of molten salt or water.
- (5) There is no danger of hydrogen explosion, because there is no water or zirconium within the containment.
- (6) Spent fuel salt is drained to a spent fuel tank, which is cooled same as a drain tank.
- (7) Seismic design and countermeasures for tsunami are required same as LWR.
- (8) Even in a worst scenario, gaseous radioactive Fission Products (FPs) are not released, because they are already removed in normal operation. Other FPs stay within fuel salt, and solidify to glassy material below melting temperature.

4. Accelerator Molten-Salt Breeder (AMSB) for ^{233}U production

The basic concept of applying accelerator to produce fissile material was originally proposed by a Canadian scientist W. B. Lewis in 1952 [29]. In 1981, K. Furukawa proposed Accelerator Molten-Salt Breeder (AMSB) as a ^{233}U producing facility [30]. Recently, it is studied in various countries as Accelerator sub-critical Driven System (ADS).

AMSB is composed of three parts: (1) 1 GeV x 200-300 mA proton accelerator, (2) single-fluid molten-fluoride target system and (3) heat transfer and electric power recovery system. A diagram of this system is shown in Figure-6. The size of target salt bath is 4.5 m in diameter and 7 m in depth. A Hastelloy N vessel is protected by a graphite reflector.

The salt flows into the top of bath, forming a vortex of about 1 m in depth. Proton beam is injected in an off-centered position, hitting near the bottom of the vortex to minimize neutron leakage and to improve generated heat dispersion. This target molten-salt system is sub-critical and is not affected by radiation unlike solid target. This makes heat removal easier, and does not need target shuffling. The beam injection port will be designed by an improved gas-curtain technology. A high-energy proton accelerator will utilize multi-beam funneling. Spallation neutrons transmute Th to ^{233}U , and also cause fissions in the target.

Target salt is ${}^7\text{LiF}\text{-BeF}_2\text{-ThF}_4\text{-}^{233}\text{UF}_4$ in equilibrium condition. Since there is no fissile ^{233}U in this salt at the start-up stage, one idea is to add PuF_3 as an initial fissile same as FUJI-Pu, that is AMSB-Pu [1].

In order to operate the accelerator of proton beam of 1 GeV x 300 mA, about 600 MWe (1,400 MWt) is required, and this electricity is recovered by fission energy in target salt. Since Conversion Ratio (CR) of target salt is high and close to 1.0, consumption of ^{233}U by fission is almost compensated by chain reaction, and its influence is small.

Assuming 80% load factor of the accelerator, annual net production rate of ^{233}U is about 800 kg/year. As is shown in Section-3.4, one 1,000 MWe super-FUJI requires 1.5 ton of initial ^{233}U , and this fissile amount will be supplied by one year operation of 2 AMSBs.

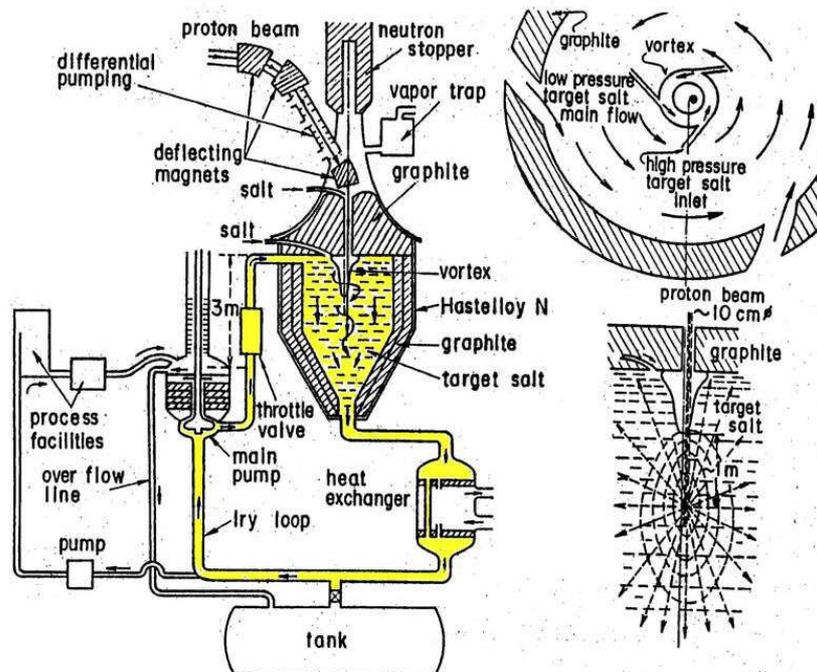


Figure-6. Schematic diagram of Accelerator Molten-Salt Breeder (AMSB).

5. Regional Center for Chemical Processing and Fissile Production.

As is proposed in Section-2, chemical processing facility connects Molten Salt Reactor (MSR) and Accelerator Molten Salt Breeder (AMSB) by utilizing common molten fluoride salt. Regional Center is mainly composed of this chemical processing facility and AMSB. These two facilities are highly sensitive to proliferation issue, because they produce large amount of fissile materials such as ^{233}U or ^{239}Pu . Therefore, this Regional Center should be placed at highly safeguarded area under international supervision throughout the world.

Figure-7 shows a layout of Regional Center and relation among chemical processing facility, AMSB, MSR and external LWR.

As is described in Section-2.2.3, LWR spent fuel can be reprocessed by dry-processing technology, and plutonium (Pu) can be transferred to a chemical processing facility in this Regional Center, and produced PuF_3 can be used as initial fissile material for the first stage MSR.

This first stage starts from mini-FUJI, which is a pilot plant for demonstration. Following after mini-FUJI, MSR-FUJI or probably FUJI-Pu starts in operation, which utilizes recovered Pu from LWR spent fuel. Spent fuel salt from MSR is returned back to a chemical processing facility for further recycling. Also, Minor Actinides (MA) from LWR spent fuel can be transmuted in some MSRs. Radioactive wastes can be processed in radio-waste plant in Regional Center. After several decades' development and when a huge amount of energy is required, AMSB is built for ^{233}U fissile production, and its fissile is used in MSR-FUJI.

Significant aspects of this fuel cycle are as follows: Total system is simply integrated by a single phase of molten fluorides based on $\text{LiF}-\text{BeF}_2$ solvent, after doing some make-up in each component facilities. Each Regional Center for chemical processing and fissile production will accommodate 4 to 40 AMSBs, two chemical processing plants and one radio-waste processing plant. It is planned that several tens of centers will be built in different locations in the world.

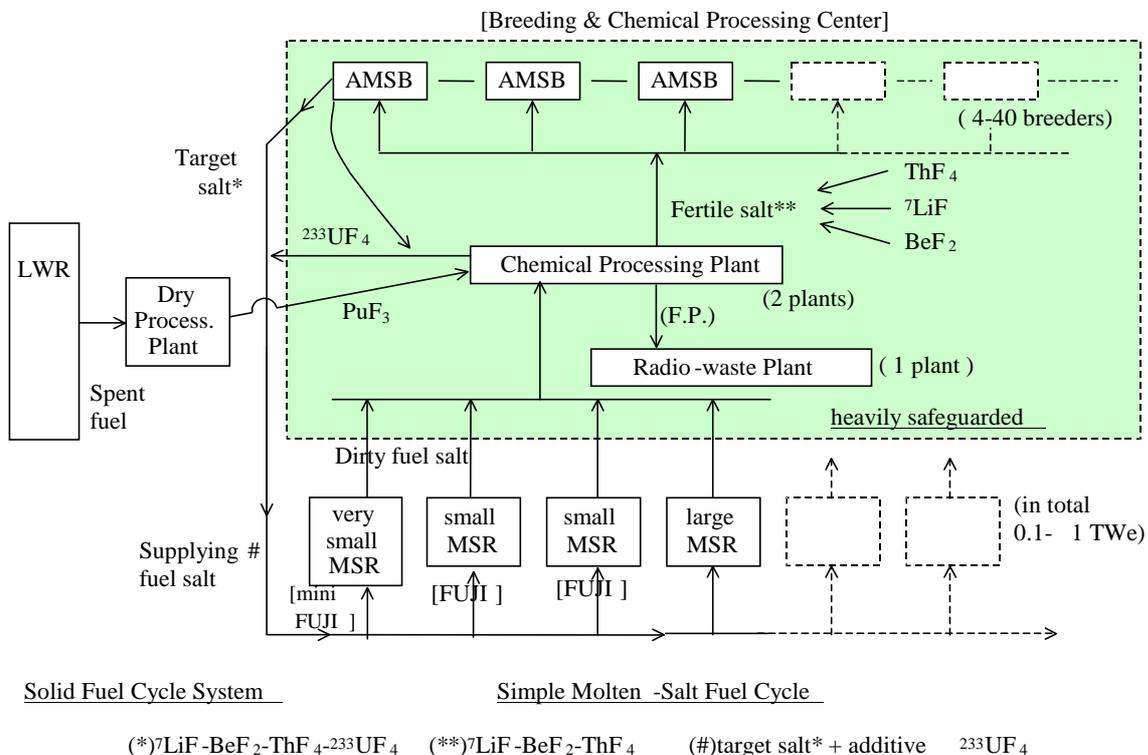


Figure-7. Regional Center for chemical processing and fissile production

6. Conclusion.

One of the most promising philosophical and technical strategies for the world survival in this century is presented as Thorium Molten Salt Nuclear Energy Synergetic System (THORIMS-NES), which is composed of Molten Salt Reactor MSR-FUJI, Accelerator Molten-Salt Breeder (AMSB), and chemical processing facility within Regional Center. These three facilities are using molten fluoride salt, and showing an ideal application of properties of molten salt into nuclear technology. THORIMS-NES can satisfy huge future energy demand, can solve the uranium resource issue, and also can solve the plutonium (Pu) issue. Production of Pu and Minor Actinides (MA) in MSR are very small, compared with LWR. Besides that, MSR can utilize Pu from LWR spent fuel, and transmute Pu and MAs. This means we can terminate U-Pu cycle, and can start a new “Thorium Era”.

As is described in this paper, MSR has a very high safety, which cannot cause severe accident like Fukushima accident.

Regarding the economy, MSR has higher thermal efficiency, and its core is simple. No refueling or shuffling is required, and then a higher load factor is possible. Self-sustaining is possible owing to superior ^{233}U -Th nuclear characteristics.

MSR has superior proliferation resistance, owing not only to the characteristics of very little Pu production, but also to its strong gamma emission by daughters of ^{232}U accompanied with ^{233}U .

Although the experimental reactor MSRE proved feasibility of MSR, and ORNL produced a commercial reactor design of MSBR, there remain technical items, which we have to develop. Some of them are maintenance equipments, steam-generator, control-rod design, long-term demonstration and so on. Regarding this subject, one of MSBR design reports shows uncertainties under its development [2].

Development of MSR and AMSB/ADS are starting in many countries, and international cooperation should be promoted.

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