# The Concept of Fuel Cycle Integrated Molten Salt Reactor for Transmuting Pu+MA from Spent LWR Fuels.

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## ABSTRACT

Japan should need a new fuel cycle, not to save spent fuels indefinitely as the reusable resources but to consume plutonium and miner actinides orderly without conventional reprocessing. The key component is a molten salt reactor fueled with the Pu+MA (PMA) separated from LWR spent fuels using fluoride volatility method. A double-tiered once-through reactor system can burn PMA down to 5% remnant ratio, and can make PMA virtually free from the HAW to be disposed geometrically. A key issue to be demonstrated is the first of all solubility behavior of trifluoride species in the molten fuel salt of $^{7}\text{LiF-BeF}_{2}$ mixture.

*Keywords: LWR Spent Fuels, Transmuting of Pu+MA, Molten Salt Reactor, $^{7}$LiF-BeF$_{2}$ Molten Salt Mixture, Solubility of Trifluorides.*

## 1- INTRODUCTION

51 LWRs are operating, 13 LWRs (4 under construction) are to be operated during the next decade, and at least 7 more LWRs are preparing to be operated by 2015 with the total capacity of 68.5 GW(e) in Japan. The nuclear power capacity in Japan increases at a linear rate of 1.5 GW(e)/y as shown in Figure 1. It is predictable that the total nuclear power capacity will reach the maximum of 90 GW(e) in 2030. Because the existing and forthcoming LWR plants will be operable for 60 years at 80% capacity factor, owing to intensive technological endeavor by utility companies and the Government, the power capacity will stay at the constant on going, if the supplement will be kept at the rate of 1.5 GW(e)/y. This life extension of LWRs can provide Japan with almost double power producing capacity and in consequence double amount of spent fuel comparing with the classical assumption of 30 years life with 80% operability. Operation of LWRs from 1970s until 2090 should produce 124,200 MTU of spent fuels. 5,630 MTU have been reprocessed in La Hague and Sellafield, and 950 MTU have been reprocessed in Tokai. About 36,600 MTU would have been reprocessed including 30,000 MTU by Rokkasho plant by 2045. Japan will consume 392 MT of recovered plutonium as 8,400 MT of MOX fuel in LWRs. Eventually, the total amount of accumulated spent fuel from LWRs will be 87,600 MT of HE including 79,200 MTU of $UO_2$ fuel by 2090. The $UO_2$ fuel will contain 847MT of plutonium, and 86MT of MA. It is assumed that the spent MOX fuel contains 70% of initially loaded plutonium and the miner actinides equivalent to 10% of initially loaded plutonium. Therefore, the spent MOX fuel will contain 275MT of plutonium and 39MT of MA. Totally, Japan will have 1,122MT of plutonium and 125MT of MA accumulated in the spent fuels from LWRs by the end of 2090.
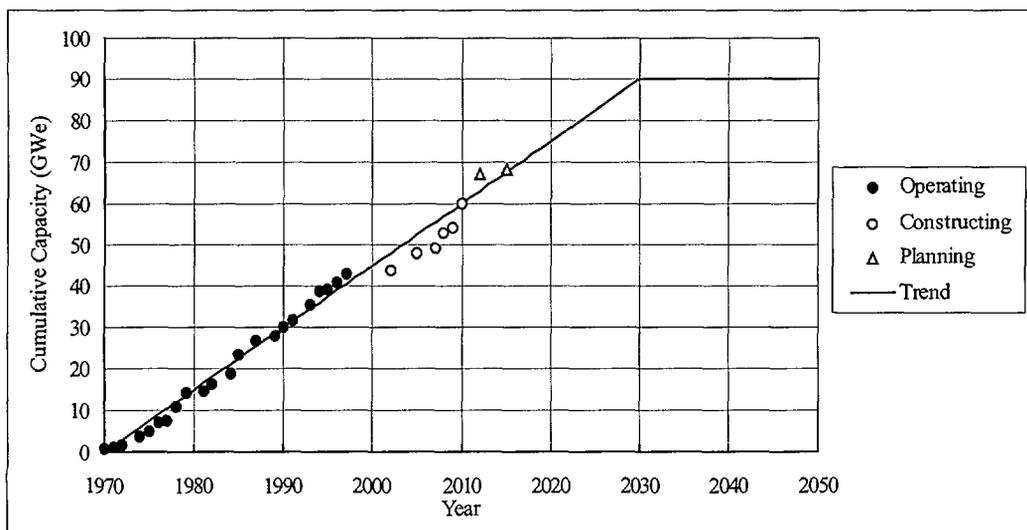


*Figure 1. Cumulative Nuclear Power Capacity in Japan*

Relying future demand of energy upon the FBR using U-Pu fuel cycle is the national policy. How much plutonium do we need to realize the goal. A typical 1-GWe LMFBR (80% capacity factor) annually requires 1.63MTPu and breeds 0.265MTPu[1]. Assuming six years of a turn around time, two years in core and four years for cooling-reprocessing-refabrication, the plutonium inventory in a system is 9.78MTPu/GW(e). In order to make up for the decreasing rate of the LWR of 1.5GW(e)/y from 2030, 55.4GW(e) of self-sustained FBR capacity will be required. The amount of plutonium required to implement the self-sustained FBR fleet is 542MT. Thus, Japan will have at least 580MT of surplus plutonium, which shall not be required by the FBR deployment and cannot be consumed easily, therefore shall become a heavy burden to the future generation in addition to the highly radioactive waste containing long life minor actinides due to the plutonium separating process from the spent LWR fuels. There might be various scenarios, in which less plutonium will be required and more plutonium shall be managed.

In Japan, any fissile/fertile contents in the spent fuels have been considered as reusable resources to be saved indefinitely, however, we advocate a concept to utilize energy from the unnecessary and troublesome resources (Pu+MA, or PMA) and much emphatically to eliminate them as far as possible at the end of LWR era. Various concepts to transmute PMA as nuclear wastes have been proposed basing upon quite a variety of technologies. We have been particularly interested in an accelerator-driven system associated with a thermal spectrum molten salt subcritical blanket consisting of two tiers, proposed by C. D. Bowman[2,3]. Tier-1 ADS would destroy most of the PMA while recovering the fission energy as electric power. The system is a continuous flow-through system with the feed entering and the discharge leaving at the same rate. The PMA mixture is significantly transformed before it leaves. The system requires no back-end separations and only modest front-end separations compared to current reprocessing technology. The out-put from this Tier-1 ADS is sent to the Tier-2 ADS where the PMA would be essentially completely eliminated. The Tier-2 system, however, would require full front-end and back-end separations. The Tier-1 ADS is characterized by fueled with PMA from the LWR spent fuels dissolved in $ZrF_4$-NaF mixture as the carrier salt, meanwhile, the Tier-2 ADS is fueled with the remnant PMA from the Tier-1 ADS dissolved in $^7LiF$-$BeF_2$ mixture as the carrier salt. It has looked to satisfy various requirements favorably such as (1) higher one-pass burn-up of PMA, (2) lower PMA inventory in the system, (3) least leakage of PMA into HAW, (4) safety, nuclear proliferation resistance, economy, and cleanliness (5) technological feasibility and viability based upon well established experiences. A feasibility to be a critical reactor without an accelerator suggested by the Kurchatov Institute[4] has stimulated our concept to select the combination of thermal spectrum molten-salt reactor without fertile materials and associated pyro-chemical processing, which is designated as the Fuel Cycle Integrated Molten Salt Reactor for Transmuting PMA from LWR Spent Fuels (IMSR-TPMA).
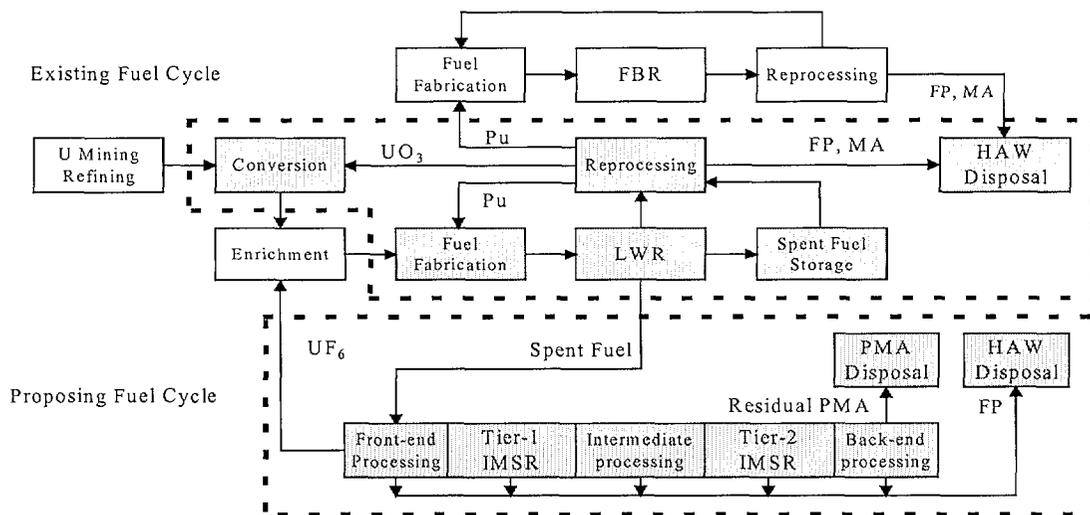


*Figure 2. IMSR-TPMA for the End of LWR Era*

2- GENERAL DESCRIPTION OF IMSR-TPMA

The IMSR-TPMA system consists of Front-end Processing; dismantling of fuel assembly, uranium recovery as $UF_6$ and PMA fuel salt preparation for the Tier-1 IMSR by FP separation, Tier-1 IMSR; a molten fluoride salt reactor fueled with a $^7LiF$-$BeF_2$-$PMAF_3$ mixture and operated as the once-through mode associated with an in-line FP (rare gas and noble metal) removal, Intermediate Processing; FP removal from the discharged fuel salt of

the Tier-1 IMSR and PMA fuel salt make-up for the Tier-2 IMSR, Tier-2 IMSR; essentially the same as the Tier-1 IMSR except salt isotopic composition, and Back-end Processing; separating and conditioning of residual PMA from the Tier-2 IMSR and accumulated FP from all over the system. Figure 2 illustrates the feature of the IMSR-TPMA in contrast to the existing fuel cycle in Japan, in which a wide scope of the parts, such as spent fuel storage, reprocessing, fuel re-fabrication, and MOX burning in LWR are replaced with the IMSR-TPMA. Much effective separation of MA from the fission product streams can mitigate the issues associated with the high level nuclear waste disposal.

## 3- CHEMISTRY OF IMSR-TPMA

Molten salt fuel for a reactor should melt at a low temperature as well as have a low parasitic neutron capture cross section. A $^7$LiF-BeF$_2$ mixture is virtually the only acceptable carrier salt to satisfy the requirements having . favorable chemical compatibility with metallic construction materials. It can dissolve ThF$_4$ and UF$_4$ in appropriate concentration for a molten salt breeder application developed in the Oak Ridge National Laboratory. In contrast to the previous case, where the on-line chemical processing was applied, the molten salt fuel for IMSR-TPMA should dissolve significant amount of tri-fluorides of Pu, MA, as well as lanthanide elements. What is more the concentration of fissile materials in the fuel salt is virtually only tool to control the reactivity of the reactor.

Solubility behavior of tri-fluorides in molten salts was investigated for two purposes. Firstly, as a process mean to remove fission product lanthanides with high cross section. Secondly, to make available Pu as the starting fissile material for the molten salt breeder/converter reactors. Consequently, in addition to the dependence on chemical composition of the solvent and on temperature, several regularities in solubility behavior of tri-fluorides have been studied. (1) The solubility of each solute in the molten solvent was dependent on the concentration of the solute in the solid solution and its solubility in the molten solvent in the absence of the other solute[5]; (2) The solubility of AmF$_3$ in the molten solvent was the same as that of PuF$_3$ since the AmF$_3$/PuF$_3$ mole ratios in the solid and liquid phases were found to be about equal[5]; (3) There was a relationship between solubility and ionic radius in a particular molten salt solvent at a specific temperature[6].



$$y = -57.765x^2 + 112.13x - 52.161$$
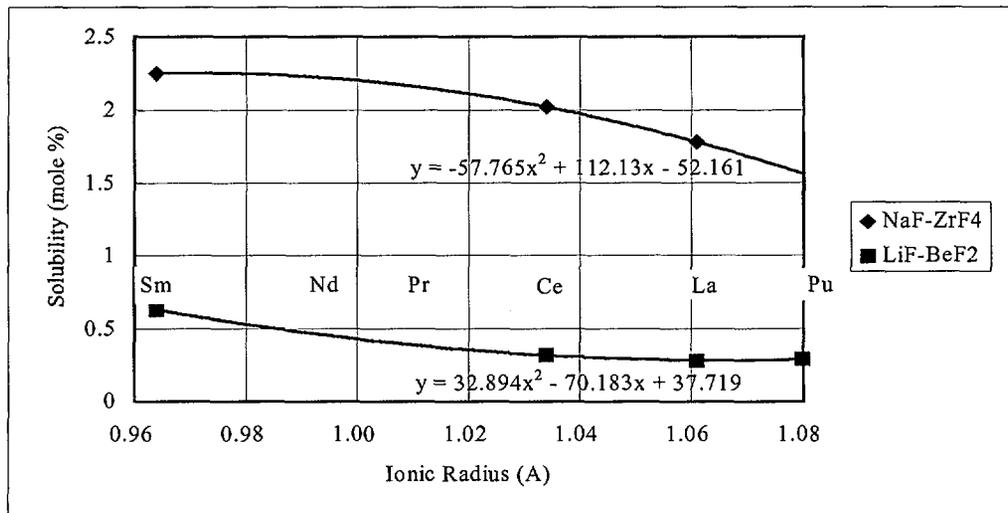
$$y = 32.894x^2 - 70.183x + 37.719$$

Figure 3. Effect of Cationic Radius on the Solubility of Tri-fluorides in Molten Fluorides at 550°C

Figure 3 illustrates the relationships between cationic radius and available measured solubility at 550°C for several tri-fluorides in 0.50NaF-0.46ZrF$_4$-0.04UF$_4$[7] and in 0.628LiF-0.364BeF$_2$-0.008UF$_4$[7], except that of PuF$_3$ which was in 0.67LiF-0.33BeF$_2$[5]. Table I shows the relative solubility referring to that of PuF$_3$ calculated based on the approximated equation shown in the Figure 3. Since, relative fission yield for lanthanide elements are constant, the weighed average of solubility for fission product lanthanide tri-fluorides comparing with PMA tri-fluorides can be calculated. Figure 4 illustrates the effect of lanthanide tri-fluorides on the solubility of co-existed PMA tri-fluoride, assuming that the relationship can be commonly applied on LiF-BeF$_2$ mixtures. The molar fraction of lanthanide tri-fluorides correlates to the one-pass burn-up ratio of PMA in a once-through PMA transmuter. Measured solubility of PuF$_3$ at 550°C in 0.67LiF-0.33BeF$_2$ was 0.29 mole%[5] and that in 0.71LiF-0.29BeF$_2$ was 0.56 mole%[8]. If the one-pass burn-up is 87.5%, the solubility of PMAF$_3$ will be 0.15 mole% and that of fission product lanthanide tri-fluoride will be 0.18 mole% in 0.67LiF-0.33BeF$_2$, in the meanwhile, the solubility of PMAF$_3$ will be 0.29 mole% and that of fission product lanthanide tri-fluoride will be 0.39 mole%.

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Table I. Solubility of Tri-fluorides at 550°C in Molten LiF-BeF₂ mixtures.

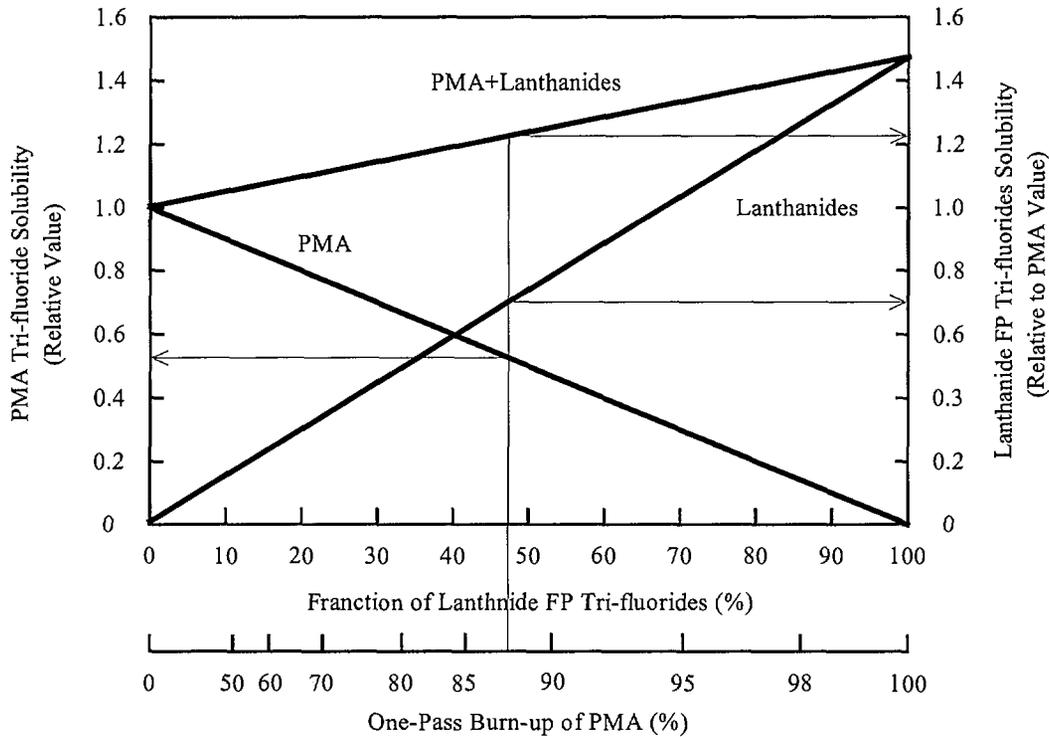| Cations | Cationic Radius $(10^{-8}m)$ | Measured Solubility (mole %) | Calculated Solubility (mole %) | Relative FP Yield |
|---|---|---|---|---|
| $Sm^{3+}$ | 0.964 | 0.63[7] | 0.630 | 0.0910 |
| $Nd^{3+}$ | 0.995 | | 0.456 | 0.4227 |
| $Pr^{3+}$ | 1.013 | | 0.381 | 0.1083 |
| $Ce^{3+}$ | 1.034 | 0.32[7] | 0.319 | 0.2457 |
| $La^{3+}$ | 1.061 | 0.28[7] | 0.284 | 0.1323 |
| Weighed Average for Lanthnides | | | 0.422 | 1.0000 |
| $Pu^{3+}$, $MA^{3+}$ | 1.08 | 0.29[5] | 0.289 | |



Figure 4. Solubility of a Binary Mixture of Tri-fluorides in a Molten Fluoride Solvent

The highest fuel temperature of the molten salt breeder reactor was limited at 704°C due to the compatibility with structural materials. The lowest fuel temperature of 566°C by selecting 0.72LiF-0.16BeF₂-0.12ThF₄ (liquidus temperature=500°C) as the fuel solvent could allow the system with 44.4% heat efficiency[9]. In the IMSR, the lowest fuel temperature should be 600°C, 50°C higher than the temperature of the fuel solvent at which solids (LiF and/or any solutes) will start to deposit. This allow to use not only LiF-BeF₂ binary system but also LiF-BeF₂-ZrF₄ ternary system, so far as their liquidus temperatures are lower than 550°C. In fact, the solubility of PuF₃ was 1.35 mole% at 558°C in 0.7LiF-0.1BeF₂-0.2UF₄, and the analogous substitution effects of BeF₂ by ThF₄ and ZrF₄ were confirmed[8].

Table II shows that the fluorine atoms combined with stable fission product cations from the fission reaction of a ²³⁹PuF₃ molecule are 2.64 out of 3.00 available. ⁷LiF will react with neutron to produce ³H, and will release fluorine at a rate of 0.09 moles per unit mole of fission. This means that the excess fluorine will accumulate at a rate of 0.45 moles per a unit mole of fission. Thus, the redox potential of fuel salt should be controlled by some means at a slightly reduced condition, in order to maintain appropriate compatibility with structural materials. Any redox controlling means should need a certain redox buffer. $U^{3+}/U^{4+}$ couple as much as 5% of PMA attributed to 99.99% of uranium separation efficiency in the front-end processing may be sufficient. In contrast to the $U^{3+}$ behavior, disproportionation reaction[10] is not a case for $Pu^{3+}$. Therefore, much lower redox potential of fuel salt can be acceptable comparing with that in the UF₄ system. This may suggest the possibility to use a stainless steel as the construction material instead of the Hastelloy N, which has typically composed of Ni (base), Mo 12%, Cr 7%, Ti+Nb 2%, and has been developed exclusively for fluoride molten salt reactors[11].

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*Table II. Fluorine Balance for Fission of $^{239}PuF_3$ in the Thermal Neutron Spectrum*

| Fission Product Elements | Valency of Elements in Molten LiF-BeF$_2$ | Fission of $^{239}$PuF$_3$ | |
|---|---|---|---|
| | | Fission Product Yield | Combined Fluorine |
| Halogens | -1 | 0.03 | -0.03 |
| Xe, Kr | 0 | 0.25 | 0 |
| Rb, Cs | 1 | 0.21 | 0.21 |
| Sr, Ba, Cd | 2 | 0.09 | 0.18 |
| Sb, Y, Lanthanides | 3 | 0.46 | 1.38 |
| Zr | 4 | 0.22 | 0.88 |
| Rare Metals | 0 | 0.75 | 0 |
| Total | | 2.00 | 2.62 |

## 4- NUCLEAR PHYSICS OF IMSR-TPMA

The IMSR is principally a critical reactor version of the thermal spectrum two-tiered ADS. Table III shows comparison in the nuclear performances between the ADS and the IMSR. Calculations were made by C. D. Bowman for homogeneous systems, using one-group cross sections for the ADS Tier-1[2], and using those derived from Monte Carlo Code MCNP-4B calculation for the ADS Tier-2[3]. Several modifications are made for the original calculations to make appropriate comparison.

The normalized system power of 750MW(t) is equivalent to the fission energy from the PMA produced by a 1000MW(e) LWR. The system is assumed that the salt volume out of flux (in plenum, pump, heat exchanger, etc.) is the same as in flux. The core PMA density is calculated from fission density, core neutron flux, and fission cross section. The effective neutron flux is averaged over whole fuel salt volume in the system, that why it is a half of the core flux. The weighed average values of fission and capture cross section of PMA are given based on the molar isotopic composition. The effective capture cross section of fission products is specific value depending upon neutron fluence. The core FP density excludes the rare gas fraction. The microscopic cross sections are adjusted at 650°C. The neutron multiplication factor keff = $\nu\Sigma f/\Sigma a$. The manual calculations have not taken into account of neutron leakage, however, the recent Monte Carlo Code MCNP-4B calculation[3] for homogeneous Tier-1 system suggested that a neutron leakage fraction was 6%, and it should be possible to reduce by a factor of two by increasing the graphite reflector thickness from 30cm to 90cm. Therefore, it is assumed that larger than 1.03 of k$_{eff}$ would represent a nuclear criticality of the IMSR. The PMA remnant ratio is defined as (PMA inventory moles)/ (fission moles per average resident time + PMA inventory moles). However, this remnant ratio is lower than that calculated by C. D. Bowman[2] using the Bateman equation by a factor of two. The concentration and solubility of solutes are compared at the same temperature of 550°C.

Comparing with the ADS Tier-1, the IMSR Tier-1 seems to be feasible nuclear physically. A minor change in the core to salt volume ratio is required, in order to adjust the PMA concentration to be accommodated by the solubility of LiF-BeF$_2$. All systems are once-through mode, in which no back flow does except the ADS Tier-2. In the ADS Tier-2, whole PMA inventory will stay in the core while fission product will have been removed by on-line processing at the rate of once an average residence time. All PMA fed, except processing loss during fission product removing, will be in the system at the equilibrium isotopic composition. C. D. Bowman assumes that the final remnant ratio in the system will be 0.00062[3]. ADS Tier-2 seems to be hardly feasible because the PMA concentration significantly exceeds the solubility. Thus, the IMSR Tier-2 will be once-through mode using LiF-BeF$_2$-ZrF$_4$ ternary carrier salt with higher PMA solubility (hopefully 1.35 mole% at 550°C, as measured for PuF$_3$ in 0.7LiF-0.1BeF$_2$-0.2UF$_4$[8]), though much higher final remnant ratio of 4% should be tolerated. A part of ZrF$_4$ may be substituted by ThF$_4$, in order to provide internally bred fissile material to keep neutron balance high, and consequently PMA remnant ratio low. This option is possible when the system will be more orientated to reduce PMA remnant, and to produce energy. Presence of external materials, such as ZrF$_4$ or ThF$_4$, in the fuel salt to be chemically processed, will be economically tolerable in the once-through operation, but not in the system with on-line processing.

C. D. Bowman has pointed out that the possibility to optimize the system heterogeneously, and that will improve neutron economy significantly[2]. O. Meplan suggested 93% of one-pass burning for the optimized Bowman' ADS Tier-1 by multi-group heterogeneous calculation[12]. This part of development is crucial not only for the ADS but also for the IMSR. Higher burn-up means higher fission product concentration as well as higher PMA concentration due to lower fission cross section. Reactor performance calculation must observe the solubility

behavior of tri-fluorides seriously.

*Table III. Comparison of Nuclear Performances between ADS and IMSR*

| Items | ADS Tier-1[2] | IMSR Tier-1 | ADS Tier-2[3] | IMSR Tier-2 |
|---|---|---|---|---|
| Operation Mode | Once-through | Once-through | FP removing PMA recycling | Once-through |
| Thermal Power MW(t) | 750 | 750 | 750 | 750 |
| Power Density W/cm$^3$ | 19.5 | 19.5 | 19.5 | 19.5 |
| Fission Density fission/cm$^3$-s | $5.933 \times 10^{11}$ | $5.933 \times 10^{11}$ | $5.933 \times 10^{11}$ | $5.933 \times 10^{11}$ |
| Core Volume m$^3$ | 38.46 | 38.46 | 38.46 | 38.46 |
| Core to Salt Volume Ratio | 0.10 | 0.117 | 0.10 | 0.10 |
| Carrier Salt | NaF-ZrF$_4$ | $^7$LiF-BeF$_2$ | $^7$LiF-BeF$_2$ | $^7$LiF-BeF$_2$-ZrF$_4$ |
| Carrier Salt Composition mole% | 50-50 | 71-29 | 71-29 | 74-16-10 |
| Salt Volume in Core m$^3$ | 3.846 | 4.500 | 3.846 | 3.846 |
| Salt Volume in System m$^3$ | 7.692 | 9.000 | 7.692 | 7.692 |
| Salt Residence Time s | $1.5 \times 10^8$ | $1.5 \times 10^8$ | $1.0 \times 10^8$ | $1.5 \times 10^8$ |
| Core Neutron Flux n/cm$^2$-s | $4 \times 10^{14}$ | $4 \times 10^{14}$ | $6 \times 10^{14}$ | $4 \times 10^{14}$ |
| Effective Neutron Flux n/cm$^2$-s | $2 \times 10^{14}$ | $2 \times 10^{14}$ | $3 \times 10^{14}$ | $2 \times 10^{14}$ |
| Neutron Fluence n/cm$^2$ | $3 \times 10^{22}$ | $3 \times 10^{22}$ | $3 \times 10^{22}$ | $3 \times 10^{22}$ |
| Core PMA Density atoms/cm$^3$ | $1.246 \times 10^{19}$ | $1.246 \times 10^{19}$ | $7.548 \times 10^{19}$ | $3.746 \times 10^{19}$ |
| Core FP Density atoms/cm$^3$ | $3.761 \times 10^{19}$ | $3.761 \times 10^{19}$ | $2.507 \times 10^{19}$ | $3.761 \times 10^{19}$ |
| Microscopic Cross Sections $10^{-24}$cm$^2$ PMA Fission PMA Capture FP Capture | 119 169 5.99 | 119 169 5.99 | 13.1 27.9 5.99 | 40.0 63.6 5.99 |
| Macroscopic Cross Sections cm$^{-1}$ PMA Fission PMA Capture FP Capture Graphite Capture Fuel Salt Capture | 0.001483 0.002106 0.000225 0.000251 0.000394 | 0.001483 0.002106 0.000225 0.000246 0.000180 | 0.000989 0.002106 0.000150 0.000251 0.000154 | 0.001483 0.002382 0.000225 0.000251 0.000262 |
| Neutron Yield per a Fission | 2.96 | 2.96 | 3.45 | 3.20* |
| Neutron Multiplication Factor | 0.9845 | 1.0353 | 0.9348 | 1.0310 |
| PMA Remnant Ratio (Cumulative) | 0.1228 | 0.1228 | 0.005 (0.0006) | 0.2964 (0.036) |
| Atomic Density in Fuel atoms/cm$^3$ Carrier Salt (550°C) PMA FP Gross Fuel | $1.888 \times 10^{22}$ $1.246 \times 10^{20}$ $3.761 \times 10^{20}$ $1.938 \times 10^{22}$ | $3.581 \times 10^{22}$ $1.065 \times 10^{20}$ $3.215 \times 10^{20}$ $3.624 \times 10^{22}$ | $3.581 \times 10^{22}$ $7.548 \times 10^{20}$ $2.507 \times 10^{20}$ $3.682 \times 10^{22}$ | $3.597 \times 10^{22}$ $3.746 \times 10^{20}$ $3.761 \times 10^{20}$ $3.672 \times 10^{22}$ |
| Concentration in Fuel (550°C) mole% PMA Lanthanides | 0.643 0.582 | 0.294 0.266 | 2.050 0.204 | 1.020 0.307 |
| Solubility in Fuel (550°C) mole% PMA tri-fluorides Lanthanide tri-fluorides | 0.819 1.001 | 0.294 0.388 | 0.509 0.074 | 1.020 0.486 |
| System PMA Inventory kg | 384 | 384 | 2362 | 1172 |

*(Arbitrary number)

As the IMSR is a critical reactor which does not contain fertile materials in the fuel salt, it has been pointed out that heterogeneous configuration should be carefully adjusted in order to ensure the negative temperature coefficient of reactivity[13]. Computational verification should be done simultaneously with burn-up calculation.

## 5- ENGINEERING OF IMSR-TPMA

The features of the IMSR plant inherit principally those of the Molten-Salt Breeder Reactor, for which a conceptual design was developed during 1970s by the Oak Ridge National Laboratory[9]. Despite the fuel salt chemistry without uranium or thorium becomes much simpler, there will be new engineering issues to be demonstrated, such as, a higher neutron flux should cause irradiation damage in graphite moderator, a lower redox potential in the fuel salt should cause more penetrating elemental tritium[14], and a higher liquidus temperature of fuel salt should need more efficient heat exchanger. Different from the case fueled with $UF_4$, it is known that oxides does not precipitate from the $PuF_3$ solution even if large amount of oxide species are accidentally inserted[15]. As $LiF$-$BeF_2$ mixtures are thermodynamically stable than any fluoride of alloying elements in construction metals, corrosion does essentially not occur, however presence of moisture can cause corrosion[11]. It is privilege given not to chloride but to fluoride to capable to remove moisture simply by sparging $H_2$-HF gas into the molten salt.

The front-end process recovering most of uranium for further use in LWR, and preparing the fuel salt to be fed to the Tier-1 IMSR is the first to come. A combination of «Fluoride Volatility Method» based on a pilot-plant experience known as «FREGAT» and «Pyro-electrochemistry» seems to be appropriate[16,17]. A particular feature of the once-through IMSR system is that the out-put of a reactor is not «spent fuels,» but can be used as the initial charge to the sister reactors. This in return needs a special engineering issue to operate the beginning reactor until the fuel salt reaches equilibrium isotopic composition. As the proposed fuel carrier salt in the Tier-1 IMSR is $LiF$-$BeF_2$ mixture, fission product separation will be straightforward, and be selected from the wide variety of proposed processes[17]. That in the Tier-2 IMSR may contain $ZrF_4$ or $ThF_4$, however, as there will be no need to feed back the PMA to the reactor, Zr or Th would be utilized to synthesize phosphate host minerals to immobilize residual PMA for final geological disposal[18,19,20].

## 6- IMPACTS OF IMSR-TPMA

Table IV shows the significant advantages of the IMSR-TPMA over the LWR-MOX as a competitive alternative. Burning 639 MT (580MTPu+59MTMA) of the LWR grade PMA will support 20GW(e) × 42 years operation of IMSR at the cumulative remnant ratio less than 5%. This will make up 32% of the total decreased capacity of LWR for 42 years. Assuming the unit capacity is arbitrary 1-GW(e), 17 sets of the front-end process/Tier-1 IMSR will be deployed during 2030~2060, and 3 sets of the intermediate process/Tier-2 IMSR/back-end process during 2060~2072 in order to decrease Japanese inventory of PMA down to at least 5% of the feed by the end of 2120, even if the system inventory after decommissioning is taking into account. 30 years time lag behind the Tier-1 IMSR, the Tier-2 IMSR and associated chemical processing technologies will have significant allowance of time for development and demonstration. The operation of IMSR should produce HAW equivalent to 56% of entire power production by LWR. TRU concentration in HAW will be virtually negligible comparing with that from the current PUREX process. About 51,500 MT of slightly enriched uranium will be recovered. This can contribute to 16% (425 GW(e)-y) of power generation by re-enrichment during a later half of the LWR era. The IMSR will offer not only a reasonable risk hedge for delayed deployment of FBRs, but also offer an appropriate technological bases for the thorium utilization using a molten salt reactor technology complying with the future potential demands.

*Table IV. Comparison IMSR-TPMA with a Competitive Alternative (LWR-MOX).*

| Items | LWR-MOX | IMSR-TPMA |
|---|---|---|
| LWR Spent Fuels | 54,230 MTU | |
| Fissile Amount | Pu 580MT | Pu 580MT+MA 59MT=639MT |
| UO$_2$ Fuel Reprocessing Plant<br>MOX Fuel Fabrication Plant | 800MT/y × 2 unit × 40y<br>150 MT/y × 2 unit × 40y | -<br>- |
| Power Plant Construction | 100%-MOX: 38.3 GW(t)× 42y | IMSR: 47.6 GW(t)× 42y<br>Front-end Plant: 17 units<br>Intermediate Plant: 3 units<br>Back-end Plant 3 units |
| Power Generation | 12.8 GW(e) × 42y (33.5% eff.) | 20.0 GW(e) × 42y (42% eff.) |
| Spent MOX Fuels | 12,400 MT | - |
| Residual PMA | 464 MT (in MOX Fuels)<br>59 MT (MA in HAW) | 32 MT (Tier-2 IMSR out-put)<br>Negligible MA in HAW |

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# 7- CONCLUSIONS

The life extension of LWRs is economically verified and not only can make up for the possible delay of practical deployment of FBRs, but also it tends pushing away the starting gate of FBR era. In due course, unexpectedly large amount of plutonium contained in the spent fuel would have been produced. We require a new strategy in the fuel cycle not to simply reserve plutonium for an energy resource crisis, but to manage plutonium while utilizing its energy effectively by the end of LWR era. Molten salt reactor technology combined with fluoride chemistry seems to be one of the best solution. Should the accelerator-driven molten salt blanket transmutation system (ADS) be feasible, a reactor version of the ADS could be feasible, provided that the concentration of tri-fluorides should be within the solubility limit. Once-through mode without on-line processing is particularly favorable to decrease the cost of system, and therefore would make the system realizable and viable, despite some amount of residual PMA should be managed. Prior to set forth the R&D program to realize the demonstration plant of the IMSR-TPMA by 2030, preliminary but fundamental technological feasibility of the IMSR should be established focusing on the experimental confirmation of solubility behavior, on the engineering verification of selected carrier salt, and on the burn-up and reactivity calculations.

## ACKNOWLEDGEMENT

## REFERENCES

[1]  M. BENEDICT, et al., *Nuclear Chemical Engineering*, 2nd Ed., McGraw-Hill, Book Co., 1981.

[2]  C. D. BOWMAN, «Once-Through Thermal-Spectrum Accelerator-Driven LWR Waste Destruction without Reprocessing,» Nuclear Technology, 132, 66 (2000).

[3]  C. D. BOWMAN, «Once-Through Thermal-Spectrum Molten-Salt Accelerator-Driven Waste Transmutation: Performance Calculations,» Report ADNA/01-03, The ADNA Corporation, Los Alamos, NM 87544, Jan. 18, 2001.

[4]  V. PRUSAKOV, et al., «Concept of the Demonstration Molten Salt Unit for the Transuranium Elements Transmutations,» ADTTA '99, June 6-11, 1999, Praha, Czech Republic, We-I-14.

[5]  J. C. MAILEN, et al., «Solubility of $PuF_3$ in Molten $2LiF-BeF_2$,» J. of Chem. and Eng. Data., Vol. 16, No. 1, pp. 68-69, 1971.

[6]  W. T. WARD, et al., «Solubility Relation among Rare-Earth Fluorides in Selected Molten Fluoride Solvents,» ORNL-2749, October 1959.

[7]  W. T. WARD, et al., «Rare Earth and Yttrium Fluorides, Solubility Relations in Various Molten $NaF-ZrF_4$ and $NaF-ZrF_4-UF_4$ Solvents,» J. of Chemical and Engineering Data, Vol. 5, No. 2, pp. 137-142, April 1960.

[8]  C. J. BARTON, «Solubility of Plutonium Trifluoride in Fused-Alkali Fluoride-Beryllium Fluoride Mixtures,» J. of the American Chemical Society, Vol. 64, pp. 306-309, March 1960.

[9]  M. W. ROSENTHAL, et al., «The Development Status of Molten-Salt Breeder Reactors,» ORNL-4812, August 1972.

[10]  L. O. GILPATRICK, et al., «The Disproportionation Equilibrium of $UF_3$ Solutions in Graphite,» MSR Program, Semiannual Progress Report for Period Ending August 31, 1970, ORNL-4832, pp. 56.

[11]  J. H. DeVAN, et al., «Materials Considerations for Molten Salt Accelerator-based Plutonium Conversion Systems,» International Conf. of Accelerator-Driven Transmutation Technologies and Applications, Las Vegas, July 1994.

[12]  O. MEPLAN, «Technical Report on the Bowman Reactor,» August 2000, private communication.

[13]  J. H. BULTMAN, «Molten Salt Reactor: Optimal Transuranics Burner,» Global '95, September 11-14, 1995, Versailles, pp. 1616-1623.

[14]  C. T. MAYS, et al., «Distribution and Behavior of Tritium in the Coolant-Salt Technology Facility,» ORNL/TM-5759, April 1977.

[15]  R. E. THOMA, «Chemical Feasibility of Fueling Molten Salt Reactors with $PuF_3$,» ORNL-TM-2256, October 1968.

[16]  V. MACHACEK, et al., «Uranium Branch for FBR Fuel Reprocessing by the Fluoride Volatility Method,» ENS-3/ANS-40 Conf., April 26-30, 1982, Brussels, pp. 121-122.

[17]  I. PEKA, «Fluoride Processing Chemistry for ADTT Molten Salt Systems,» ADTTA '99, June 6-11, 1999, Praha, Tu-O-G2.

[18]  Y. HIROSE, et al., «Alternative Conditioning Method for the Waste from LWR Fuel Reprocessing,» Global '97, pp. 1181-1186, 1997.

[19] A. I. ORLOVA, et al., «Elaboration of Phosphate Ceramics as a Safe Form for Halide Salt Waste Immobilization of 'Dry' Fuel Reprocessing,» Global '97, pp. 1253-1258, 1997.

[20] V. BRANDEL, et al., «Thorium Phosphates as Matrices for Radionuclides Immobilization,» NATO Workshop on Global Solutions to Disarmament Involving Management of Radionuclides, Cracow, 9-13 November 1998.