

# MOLTEN SALT CRITICAL REACTORS FOR THE TRANSMUTATION OF TRANSURANICS AND FISSION PRODUCTS

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## ABSTRACT

The primary goal of this work is to study the feasibility of designing molten salt reactors to be critical when fed with, and only with the transuranium isotopes (TUs) and long lived fission products produced in LWRS. Another goal is to estimate the transmutation characteristics which might be attainable from such a Molten Salt Transmuting Reactor (MSTR). Both moderated and unmoderated MSTR systems are examined. It is found that MSTRS can be designed to be critical and to have potentially attractive transmutation characteristics. However, the minimum required TUS concentration, and the transmutation characteristics of the MSTR relative to the ATW and LMRs need yet to be quantified. Implications of MSTR on the nuclear energy strategy are discussed.

## INTRODUCTION

The transmutation of transuranium isotopes (TUs) and long-lived fission products (FPs) into short lived and stable isotopes has long been suggested as a means for treating the radioactive waste disposed from nuclear reactors. A large number of different conceptual and technical approaches to waste transmutation have been proposed throughout the years. In recent years, the attention is focused on a couple of approaches: the use of liquid metal reactors (LMR)<sup>(1-4)</sup> and the use of an accelerator-driven thermal subcritical blankets -the accelerator transmute of waste (ATW) proposed by Los Alamos National Laboratory<sup>(5-8)</sup>. Two classes of blankets are being considered for the ATW - a molten salt (MS) blanket in which the TUS are in solution in the MS<sup>(5,8)</sup>, and an aqueous blanket in which the TUS are incorporated as slurries in heavy water<sup>(6,7)</sup>.

LMRs specially designed for TU transmutation can transmute on the order of 10%<sup>(9)</sup> of their inventory per year. Owing to their hard neutron spectrum, they are not considered suitable for the transmutation of FPs. The ATW offers a fractional TU transmutation which is at least an order of magnitude higher than the LMR while, at the same time, can also transmute the long

lived FPs. On the other hand, the LMR does not need a high energy accelerator and can be designed to convert fission to electrical energy at a higher efficiency.

In early 1991 one of the coauthors (E. G.) proposed that it might be possible to achieve the attractive transmutation characteristics of the ATW by using a MS transmuting reactor (MSTR) that is designed to be critical, rather than to be driven by an accelerator neutron source. Shortly thereafter, a very preliminary study indicated that it might be possible to design a MSTR to be critical, when fed with the TUS and long lived FPs which can be extracted from LWR spent fuel. Consequently, a more thorough study of the feasibility of the MSTR approach to transmutation was recommended.

The present paper summarizes the first part of this more thorough feasibility study which was sponsored in 1992 by the Los Alamos National Laboratory (LANL).

## STUDY GOALS AND APPROACH

The primary goal of the study was to find whether or not it is possible to design a molten salt (MS) reactor to be critical, when fueled with, and only with the TUS and long lived FPs from LWRS spent fuel. Additional goals were to find the following: 1. The minimum critical concentration of TUS in the MS, and 2. The transmutation rate the MSTR can be designed to have.

As the purpose of this feasibility study was to establish lower or upper bounds, we considered simplified MSTR models and used other simplifying assumptions for the analysis. Consequently, the reader should not assume that the specific systems analyzed are being proposed for practical implementation.

## MSTR CONCEPT AND MODE OF OPERATION

Two types of MSTR cores were considered in this work: moderated and unmoderated. Both use MS for their fuel and coolant. The moderated MSTR core is assumed to be similar in geometry to a CANDU type

HWR in which the D<sub>2</sub>O coolant and fuel rod cluster are replaced by MS. The pressure and calandria tubes are taken to be zircaloy (Zy), both 0.1 cm in thickness, separated by a 1.06 cm gap. The unmoderated MSTR is assumed to be made of a double wall tank filled with MS which is surrounded by a D<sub>2</sub>O reflector. The core tank is also taken to be made of Zy. It consists of two 0.3 cm thick walls separated by a 3.2 cm thick gap. It ought to be realized that Zy is not likely to be compatible with MS at the contemplated operating temperatures -- up to 7500C at the MS outlet. It has been selected for this study in order to establish upper/lower bound estimates. The D<sub>2</sub>O moderator and reflector are assumed to be maintained at a relatively low temperature of few dozens degrees Centigrade, so that the entire MSTR system operates at a low pressure. It is due to the low pressure that the Zy tubes are assumed to have the small thickness they have.

Nine TUS from LWRS (<sup>237</sup>Np; <sup>238</sup><sub>1</sub>, <sup>239</sup><sub>240</sub>, <sup>241</sup>&<sup>242</sup>Pu; <sup>241</sup> & <sup>243</sup>Am; <sup>244</sup>Cm) are continuously fed into the MS and are assumed to be in solution with the MS. The FPs from the LWRS (<sup>79</sup>Se; <sup>90</sup>Sr; <sup>99</sup>Tc; <sup>126</sup>Sn; <sup>129</sup>I; <sup>135</sup>Cs; <sup>137</sup>Cs; <sup>151</sup>Sm) are continuously fed into the D<sub>2</sub>O and are assumed to be in the form of slurries. The relative number of atoms of these TUS and FPs are taken from Ref. 5; they correspond to the composition of the fuel discharged with 33 GWD/T from a 3 GWth PWR and cooled for 10 years (except for the Sr and Cs isotopes which are cooled for 30 years). The compound used for the slurries, as well as their volatility limit, were also taken from Ref. 5. A side stream of the MS is continuously reprocessed to remove the short lived and stable isotopes resulting from the transmutation. The long-lived FPs generated in the MS are extracted and then fed into the D<sub>2</sub>O. Following are additional assumptions used in the analysis:

(1) The short lived and stable transmutation and activation products are continuously extracted with an effective "half-life" of one day (Dr. Uri Gat from ORNL is suggesting<sup>(11)</sup> that 10 hours might be attainable). The effective half life for Xe is taken to be one hour. The Cs generated from the extracted Xe decay is kept out of core for 30 years, so only half of the extracted <sup>137</sup>Xe and all of the extracted <sup>135</sup>Xe are returned to the MSTR as, respectively, <sup>137</sup>Cs and <sup>135</sup>Cs. Also continuously removed from the core are activation products of the MS constituents, and these constituents are being fed in to make up for their depletion.

(2) The maximum MS flow velocity is 10 m/s. For a core 1 m in height and a MS temperature increase across the core of 300°C, this implies a maximum permissible average power density of 10 kW/cm<sup>3</sup>. It ought to be mentioned that a number of studies of MS reactors performed in the sixties and seventies called for power densities in the range between 10 kW/cm<sup>3</sup> to 20 kW/cm<sup>3</sup> (See Ref. 12 and references in Ref. 13).

At the initiation of the study we assumed that the MS for the MSTR will be the low melting temperature eutectic of LiF (47 mole %) and BeF<sub>2</sub> (53 mole %). Consequently, most of the calculations we did so far pertain to Flibe. In the course of the study we learned from Prof. Furukawa<sup>(14)</sup> that the volatility limit of Pu in Flibe (LiF-BeF<sub>2</sub>) at the temperature range of interest to the MSTR is approximately 1 mole %. This implies that if the critical concentration of Pu need be higher than 1%, it will either have to be introduced into the core in an immiscible form, or a MS other than Flibe will have to be used. Subsequently we have identified the following alternative salts which can accommodate, in a liquid phase, much higher concentration of TUS: (a) (TU)Cl<sub>3-x</sub>NaCl<sup>(12)</sup>, (b) (TU)F<sub>3</sub>-NaF-ZrF<sub>4</sub><sup>(12)</sup> and (c) (TU)F<sub>3-x</sub>KF-yNaF-zBeF<sub>2</sub><sup>(15)</sup>. Limited number of calculations have been done, so far, with alternative "b" only, as the cross sections for Cl and K were not included in the 27 group neutronic library.

The study was organized in three parts. The first part included the establishment of the computational tool and methodology, developing data analysis and display procedures, identifying deficiencies in data, and carrying out a preliminary scoping study. Two types of MS systems were studied: (a) Infinite medium of unmoderated MS, and (b) Infinite D<sub>2</sub>O moderated lattices. The second part included improving the data base, improving the modeling of the MSTR systems and checking the effect of these improvements on the results obtained in the first part. Both parts used Flibe for the MS, and ignored the volatility constraints.

The third part considered a D<sub>2</sub>O reflected finite, one dimensional cylindrical core. Its goal was to establish an upper bound on the TU concentration which is needed for obtaining criticality, when accounting for neutron leakage and reflector effects, and while being free from any constraint on the volatility of the TUS in the MS; (TU)F<sub>3</sub>-3.5NaF-3.4ZrF<sub>4</sub> was used for the MS. Due to paper size limitations, we shall present here detailed results from only Part 1 of the study.

The primary design variables considered in this study are the equilibrium plutonium concentration and the average power density in the MS. The flux amplitude is derived from the equilibrium plutonium (actually, TU) concentration and the average core power density. The D<sub>2</sub>O volume in the moderated MSTR lattices was another design variable.

## COMPUTATIONAL METHODOLOGY

The computational tool used for the study is based on the SCALE-4.1 code system (Released during the Spring of 1992) which was developed and thoroughly tested by ORNL for the NRC<sup>(16)</sup>. A special subset of modules of the SCALE-4.1 system was put together to simulate the MSTR neutronic/isotopic behavior; it consists of the BONAMI, NITAWL, XSDRN, COUPLE and ORIGIN modules, and of the following data

libraries: Bondarenko factors (for BONAMI); Resonance parameters (for NITAWL); 27 group neutronic cross-sections (for XSDRN); Transmutation cross-sections (to be collapsed in XSDRN for ORIGEN); FP yield library and decay constants library (for ORIGEN). This special subset of modules was set up to calculate the equilibrium core composition under continuous feed-in and extraction conditions. It enables carrying out consistent neutron and isotope balance calculations, taking into account spectral effects, spatial effects (1-D), and transmutation effects.

The above described computational system was checked by applying it to relevant sample problems provided in the SCALE-4.1 code package, as well as to PWR burnup benchmark problems reported in the literature<sup>(17)</sup>. Good agreement was obtained in all the benchmark problems considered. In addition, our computational system was compared against WIMS<sup>(18)</sup> for a couple of FP free MS lattices, one moderated and the other unmoderated, which were calculated in Ref. 10. The infinite multiplication constant calculated with SCALE was within 0.014% and 3.2% of the WIMS results. The SCALE values were the more conservative.

The calculation of the equilibrium composition and the corresponding performance parameters was done in an iterative manner, in the following sequence: (a) Assume the concentration of all the major system constituents and run the BONAMI-NITAWL-XSDRN (BNX) codes to generate spectrum and spatial averaged group constants. (b) Run COUPLE to implant these 3-group constants into the ORIGEN library of activation cross-sections. (c) Set the  $^{239}\text{Pu}$  feed-in rate (all the other TUS and FPs feed-in rate is proportional to that of the  $^{239}\text{Pu}$ ) and the thermal flux amplitude and run ORIGEN to a limited burnup. (d) If the power density is lower/higher than desirable, increase/decrease the feed-in rate and repeat "c". (e) If after a relatively short burnup the concentration of any important constituent varies by more than 10%, use the new concentration and go to "a". Also adjust the thermal flux level to provide the desirable power density. (f) If after long (3000 full power days) burnup the concentration of any of the constituents which has non-negligible contribution to either the reactivity or long term radioactivity  $\gamma$  varies by more than 10%, adjust the equilibrium concentration and the thermal flux level and go to "a". Otherwise convergence is assumed.

It turns out that the nuclear data libraries of SCALE-4.1 are missing data of relevance to the MSTR. The 27 group library for XSDRN contains data for only the following TUs and FPs of relevance to our system (the cross-sections for the other isotopes are available in the ORIGEN 3-group library; they are not updated according to the system spectrum):  $^{237}\text{Np}$ ;  $^{238,239,240,241}$  &  $^{242}\text{Pu}$ ;  $^{242}$  &  $^{243}\text{Am}$ ;  $^{244}\text{Cm}$ ;  $^{90}\text{Sr}$ ;  $^{99}\text{Tc}$ ;  $^{126}\text{Sn}$ ;  $^{129}\text{I}$ ;  $^{135}$  &  $^{137}\text{Cs}$ ;  $^{105}\text{Rh}$ ;  $^{135}\text{Xe}$  and  $^{150,151}$  &  $^{152}\text{Sm}$ . Fortunately, these isotopes account for more than 90% of the total neutron absorption in the system, so that the BNX calculated

neutron spectrum is expected to well represent the system spectrum.

Following Part 1 of the study we made the following improvements in the SCALE-4.1 data base (1) Addition of fission and removal cross sections for the short lived TUs  $^{238}\text{Np}$ ,  $^{243}\text{Pu}$ ,  $^{242}\text{Am}$ ,  $^{244}\text{Am}$ ,  $^{253}\text{Cf}$  and  $^{250}\text{Bk}$ . Most of these TUS had only thermal group fission cross sections. The resonance group fission cross section was deduced by assuming<sup>(19)</sup> that its ratio to the thermal group cross section is the same ratio as for  $^{242}\text{Am}$ . Similarly, the fast group fission cross section was assumed to be related to the thermal group cross section as the ratio of the fast to thermal cross sections for  $^{242}\text{Am}$ , the capture to fission cross section ratio in the resonance and fast groups was taken to be the same as for the thermal group. (2) "Nu" values were added to 31 TUS in from one up to three of the energy groups. When missing from the ORIGEN data base, the thermal group "Nu" values were borrowed from a recent IAEA compilation. The resonance and fast group "Nu"s were extrapolated from the corresponding thermal "Nu" value. (3) The FP yield data of  $^{239}\text{Pu}$  was assigned to 12 TUs for which our ORIGEN data base had no yield data, including  $^{237,238}\text{Np}$ ,  $^{238,240,242,243}\text{Pu}$ ,  $^{242,242m,243}\text{Am}$ , and  $^{244,245}\text{Cm}$ .

Vast amount of information on the equilibrium system characteristics (as well as on the rate of approach to equilibrium) is obtained from our code system. This information includes the multiplication constant, fractional neutron absorption in each of the system constituents as well as the concentration of these constituents (which include 26 TUS, over 800 FPs and several dozens other isotopes associated with the MS,  $\text{D}_2\text{O}$  and structural materials); and the flux and power density distribution across the system (either unit cell or reflected core).

## RESULTS - PART 1

Figs. 1 through 8 pertain to an infinite medium of unmoderated, structure free lattices of Flibe containing 1 mole %  $^{239}\text{Pu}$ . All the TUs and FPs are assumed to be in elemental form, uniformly distributed in the MS. Hence, the results are just to give indication on orders of magnitude and trends, rather than to represent the characteristics to be expected from a realistic design.

Fig. 1 shows the infinite multiplication constant,  $K_{\text{inf}}$ , with (bottom line) and without (top line) the transmutation of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ . The varying slope of  $K_{\text{inf}}$  is a result of the following trends related to increasing power density: (a) Increase in the FP parasitic neutron capture; (b) Increase in the fission probability of the short lived TUS; and (c) Reduction in the equilibrium concentration of some of the short lived TUS due to a reduction in the equilibrium concentration of their short lived mothers (Responsible for the decline in  $K_{\text{inf}}$  following the "hump" around 2

kW/cm<sup>3</sup>).

Fig. 2 shows the equilibrium concentration of the primary long lived FPs. The molar fraction of all these FPs (relative to FLiBe) varies from approximately 0.1 at very low power densities, to nearly 0.4 around 10 kW/cm<sup>3</sup>. The concentration dependence on the power density comes, primarily, from the relatively short lived FPs <sup>137</sup>Cs and <sup>90</sup>Sr. The equilibrium concentration of the TUS is an order of magnitude smaller.

Figs. 3 and 4 show the effective half-life of the important TUS and FPs versus the physical half-life (Displayed at the zero power density line). The effective half-life of all the TUS is reduced to below one year as the power density exceeds few tenth of a kW/cm<sup>3</sup>. More difficult is to reduce the effective half life of the FPs. Thus, at a power density of 10 kW/cm<sup>3</sup> the effective half life of <sup>126</sup>Sn, <sup>90</sup>Sr, <sup>137</sup>Cs and <sup>79</sup>Se is, approximately, 30, 9, 4 and 2 years.

Fig. 5 shows the net mass of TUs consumed per year (assuming a 85% capacity factor) per unit mass of TUs residing in the MSTR core. This indicator of the TU transmutation ability of the MSTR is seen to be strongly sensitive to the equilibrium Pu concentration [Pu] in the MS, and to the power density. At [Pu]=1 % and 10 kW/cm<sup>3</sup>, the MSTR (if could be designed to be critical and to operate at such a high power density) could transmute in one year nearly 14 times its in core TU inventory.

Fig. 6 shows the MSTR transmutation ability of the long lived FPs excluding <sup>137</sup>Cs and <sup>90</sup>Sr. At [Pu] = 1% and 10 kW/cm<sup>3</sup>, the MSTR could transmute approximately 60% of its in core FP inventory per year. If <sup>137</sup>Cs and <sup>90</sup>Sr were to be transmuted as well, the fraction of the in core inventory which could be transmuted per year would have been lower by a factor of about 4.

Figs. 7 and 8 show the effect of the equilibrium Pu concentration on K<sub>inf</sub> and on the neutron balance. These results are not corrected to account for the effect of the fissions of the short lived TUS, hence, K<sub>inf</sub> is underestimated. Notice that the neutron balance is, practically, independent of the [Pu], and very slightly depends on the power density. Typically 80% of the neutrons are absorbed in the TUS, 18% in the FPs, and 2% in the MS.

Figs. 9 through 12 pertain to the reference D<sub>2</sub>O moderated lattices. The TUS are being fed into the MS as fluorides, whereas the FPs are being fed into the D<sub>2</sub>O as soluble salts. The power density is assumed to be 10 kW/cm<sup>3</sup>. The increase in K<sub>inf</sub> with a reduction in [Pu] below about 0.02 (Fig. 9) is due to enhancement in the fission probability of the short lived TUS (as a result of an increase in the flux amplitude). The increase in K<sub>inf</sub> with an increase in [Pu] above about 0.02 is due to spectrum hardening effect (leading to an increase in

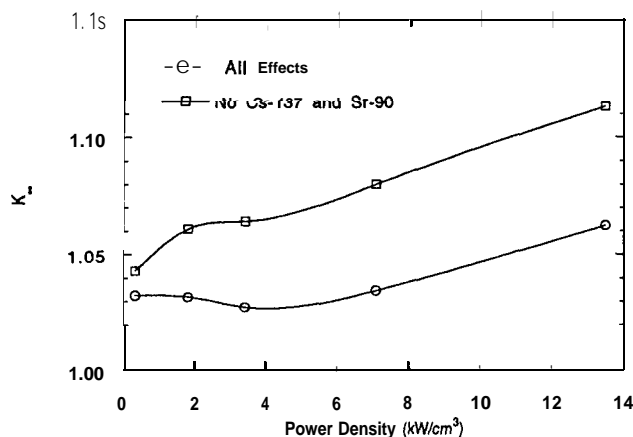


Fig. 1 Power density effect on K<sub>inf</sub>. Unmoderated MS with [Pu]=1%.

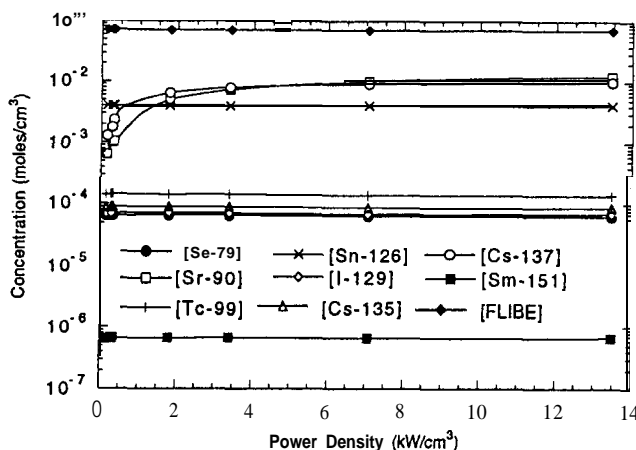


Fig. 2 Equilibrium FP concentration in unmoderated MS systems. [Pu]=1%.

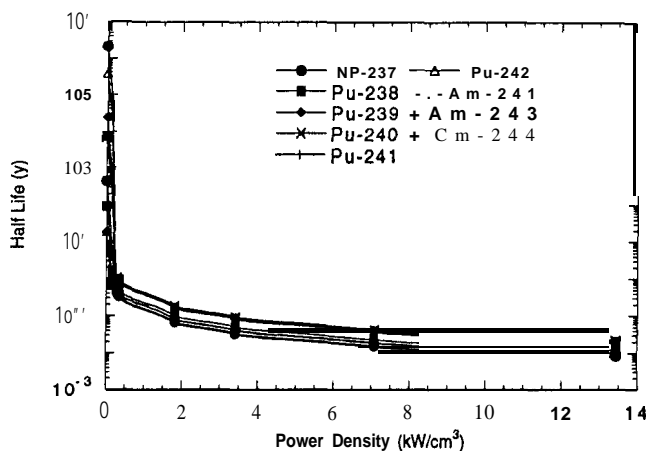


Fig. 3 TU effective half-life in unmoderated MS systems. [Pu]=17%.

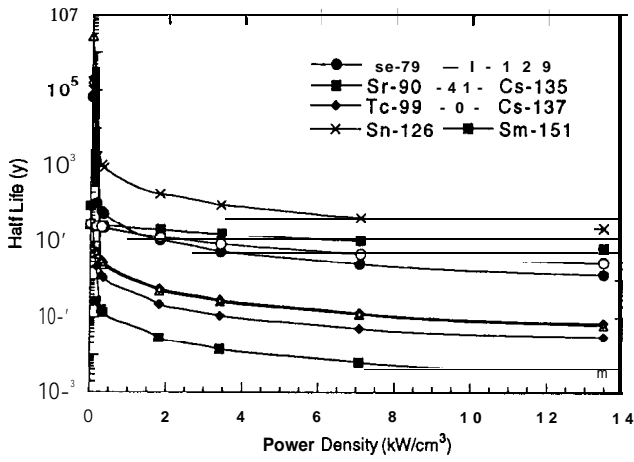


Fig. 4 FP effective half-life in unmoderated MS systems. [Pu]=1%

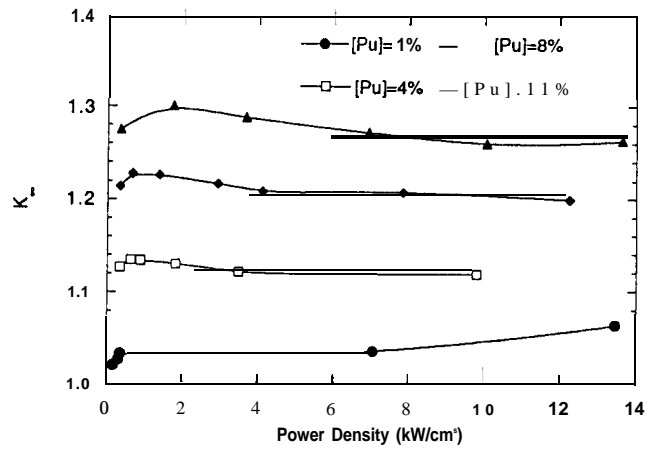


Fig. 7 Effect of [Pu] and power density on  $K_{inf}$ . Unmoderated MS systems.

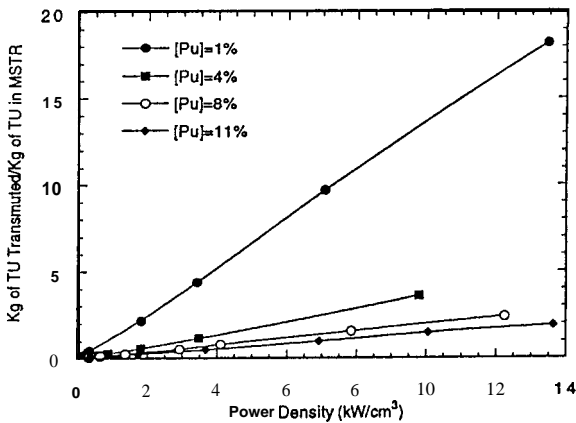


Fig. 5 Fraction of TU in-core inventory transmuted per year in unmoderated MS systems. Capacity factor = 0.85

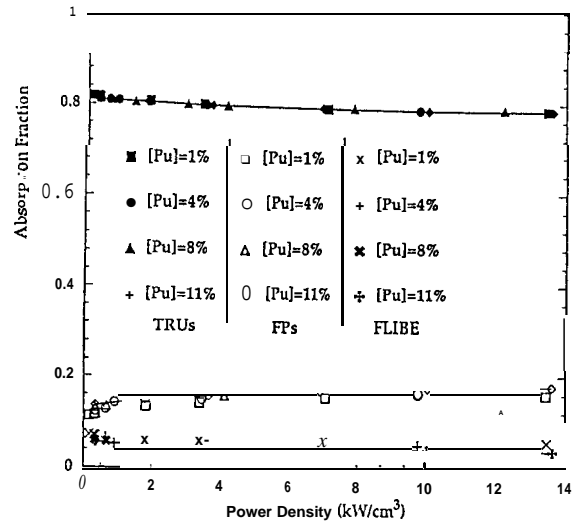


Fig. 8 Neutron balance in unmoderated MS systems.

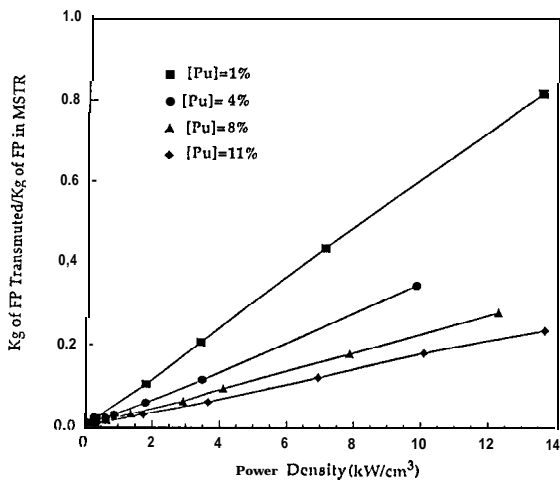


Fig. 6 Fraction of FP in-core inventory transmuted per year in unmoderated MS systems. Capacity factor = 0.85

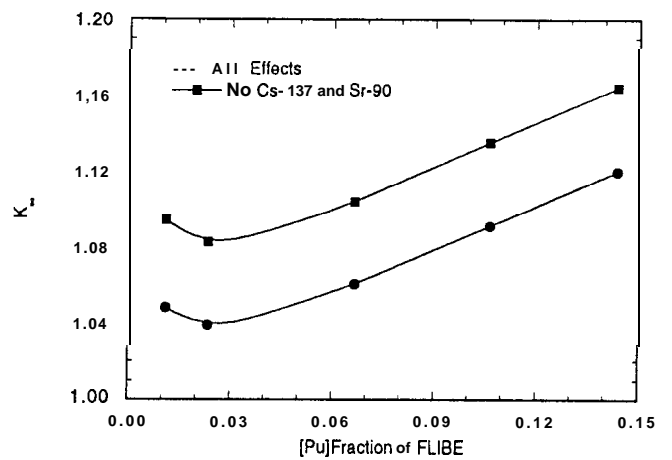


Fig. 9  $K_{inf}$  attainable in moderated MSTR lattices. Power density = 10 kW/cm<sup>3</sup>

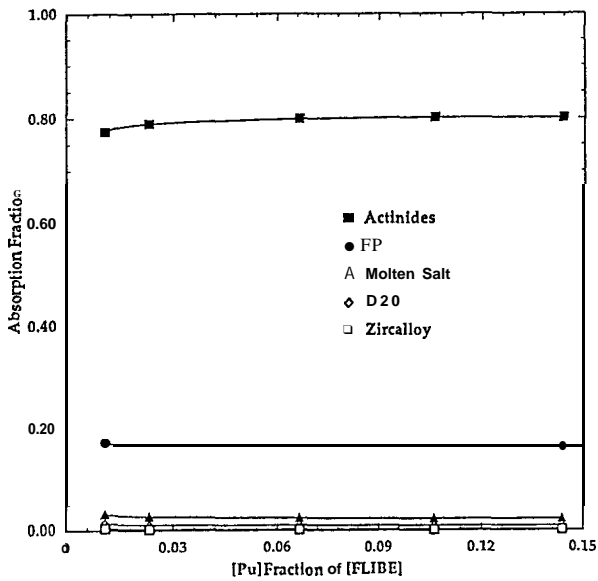


Fig. 10 Neutron balance in moderated MSTR systems. Power density = 10 kW/cm<sup>3</sup>.

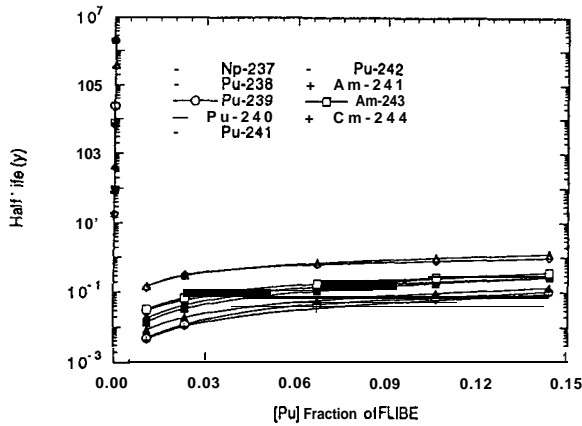


Fig. 11 TU effective half-life in moderated MSTR systems. Power density = 10 kW/cm<sup>3</sup>.

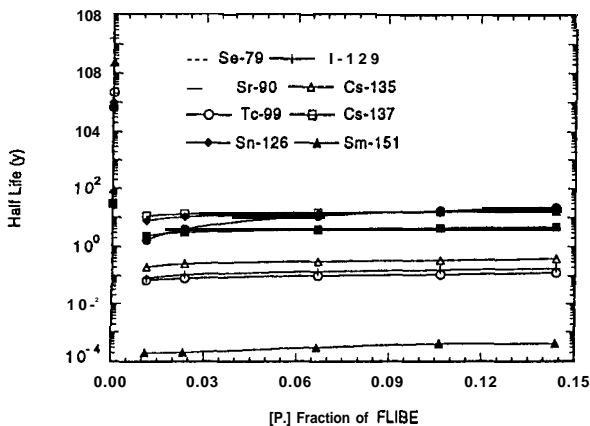


Fig. 12 FP effective half-life in moderated MSTR systems. Power density = 10 kW/cm<sup>3</sup>.

fission-to-capture ratio). The neutron balance in the moderated lattices (Fig. 10) is quite similar to that of the unmoderated lattices (Fig. 8).

The effective half life of the TUs in the moderated lattices (Fig. 11) is also below 1 year, although somewhat higher than for the unmoderated lattices having the same Pu concentration [Pu] and power density. On the other hand, the effective half life of the FPs (Fig. 12) is somewhat lower than in the unmoderated lattices having similar [Pu] and power density. At 1 mole % Pu and 10 kW/cm<sup>3</sup>, the effective half life of <sup>137</sup>Cs, <sup>126</sup>Sn, <sup>90</sup>Sr and <sup>78</sup>Se is, approximately, 12, 7, 2 and 1.6 years.

The similarity in the transmutation ability of the unmoderated and the moderated MS systems may surprise many of the readers. It is due to the fact that although the spectrum averaged transmutation cross sections tend to decline as the spectrum hardens, the ratio of transmutation to fission cross sections is only slightly sensitive to the spectrum variation. As the spectrum hardens, the flux amplitude needed to provide a given power density increases. Thus, whereas in the moderated lattice the average thermal and epithermal flux in the MS is, approximately, 8x10<sup>15</sup> and 1.5x10<sup>16</sup> n/cm<sup>2</sup>/s, in the unmoderated lattice these fluxes are, respectively, 3x10<sup>12</sup> and 2x10<sup>17</sup> n/cm<sup>2</sup>/s. The above flux values pertain to 1 mole % Pu and 10 kW/cm<sup>3</sup>.

## RESULTS - PART 2

Repeating the calculation of representative unmoderated lattices using the corrected and upgraded data base we found that (a) the addition of fission cross section and "Nu" values increases  $K_{inf}$  by, typically, 1.5%, but (b) the accounting of FP yield from all the TUS leads to a comparable reduction in  $K_{inf}$ . It is concluded that due to compensating effects, the  $K_{inf}$  values presented in Part 1 are representative of the values to be expected from more refined representation of the MSTR core.

More significant was the difference in the value of the equilibrium concentration of certain of the FPs; the newly calculated equilibrium concentration of the FPs of relevance is higher by 10% to 20% than the concentration calculated in Part 1 of the work. The equilibrium concentration of most of the TUS is found to be insensitive to the data change. An exception are few of the radioactive decay daughters of certain short lived actinides; the concentration of these daughters is now smaller than obtained in Part 1.

Overall, the improved data base did not change the conclusions which could be drawn from Part 1 of the study.

## RESULTS - PART 3

The finite core was modeled similarly to the

moderated infinite lattice except for three modifications: (a) The zones radial thickness was enlarged, (b) The outer boundary condition was changed from reflective to vacuum, and (c) The multiplication constant was corrected to account for the axial neutron leakage. An estimate of the axial leakage was obtained by applying the BNX calculations to a slab geometry core the thickness of which is the height of the actual core. Most of the neutrons which leak radially from the core are being captured in the long-lived FPs which are present in the radial reflector. In reality, FPs could be introduced also in the axial reflector, thus reducing the axial leakage loss.

The critical plutonium concentration in a 150 cm diameter, 150 cm high MSTR was calculated to be 5%. 79.3% of the neutrons are absorbed in the TUS, 17.3% in the FPs, and 3.4% in the MS, D<sub>0</sub>, and Zy core vessel (0.8%). Had the <sup>90</sup>Sr and <sup>137</sup>Cs not been transmuted, there would have been an excess neutrons of 4.2% and the critical TU concentration could have been reduced (by approximately 2%).

Examination of the flux and power density distribution across the core shows that, even though the epithermal flux is orders of magnitude higher than the thermal flux over most of the core volume, the majority of the fissions are induced by neutrons which are thermalized in the reflector. Correspondingly, there is a very steep power spike over the outer 5 cm or so of the core. Thus, a large fraction of the fissions in this supposedly unmoderated MSTR are, in fact, thermal fissions. No attempt was made to find the MSTR design that will minimize the critical TU concentration.

## DISCUSSION

At this point we can not reliably conclude about the lowest practical Pu (or TU) concentration MSTRS can be designed to have. Most of the parasitic neutron captures which are not accounted for in the infinite lattice calculations take place in the reactor vessel. Let us assume that this vessel will absorb 1.5 % of the neutrons and allow for 6.5% of the neutrons to be lost in the axial direction. Then a  $K_{inf}$  of 1.08 should be sufficient to assure criticality. Figs. 1 and 6 show that such a  $K_{inf}$  might be obtained with both the unmoderated and the moderated lattices even with as low a Pu concentration as 1 % provided the MSTR is not to transmute the <sup>90</sup>Sr and <sup>137</sup>Cs.

Suppose that the MSTR could, indeed, be designed to be critical with a 1 mole % Pu concentration, and have a power density of 10 kW/cm<sup>3</sup>. Also suppose that the out-of-core MS inventory (needed for transferring the fission power to the thermodynamic working fluid through out-of-core heat exchangers, as well as for continuous processing) is ten times the in-core inventory. Then the annual TU transmutation rate of the MSTR is calculated to be 1.3 times its TU inventory. This is an order of magnitude higher than attainable

with a LMR designed to maximize the fractional transmutation<sup>(9)</sup>. This MSTR will also transmute its in-core inventory of long lived FPs (excluding <sup>90</sup>Sr and <sup>137</sup>Cs) within approximately 1 year.

It is expected that a MS blanket for an ATW could be designed to have a lower TU concentration (and inventory) than a MSTR. Assuming that the ATW blanket and the MSTR core could be designed to have the same power density and the same relative out-of-core (or blanket) TU (and FP) inventory, the ATW is expected to offer a higher fractional transmutation than the MSTR. However, the present study does not enable us to quantify the differences in the transmutation characteristics of the two type of systems. Hence, we can not yet evaluate whether the expected improved transmutation characteristics of the ATW will justify the use of an accelerator and target systems for generating the spallation neutron source.

One MSTR should be able to transmute the TUs and long lived FPs generated by LWRS the total thermal capacity of which is 3.4 times that of the MSTR. As the net energy conversion efficiency of the MSTR is expected to be at least 30% higher than that of LWRS, the MSTR can increase the electrical energy generated from a given mass of natural uranium ore by 40% over that generated by conventional LWRS. A "nuclear island" consisting of the proper synergism of LWRS, a MSTR and a reprocessing plant could conceivably generate 4070 more electricity from a given natural uranium resource, while incinerating most of the TUS and long-lived FPs that would otherwise accumulate in the LWRS spent fuel. Trace amounts of TUS and long lived FPs will find their way with the short half life and non radioactive waste to be extracted in the MSTR reprocessing plant. Nevertheless, it is possible that the waste from this nuclear island will not have to be stored in a deep geologic repository.

In addition to nuclear waste from LWRS, MSTRS could be very attractive for transmuting the excess Pu from military programs. As the technology development required for transmuting the military Pu is expected to be less demanding than that required for transmuting LWR waste, the development of the MSTR technology can proceed in phases; the first phase being the development of MS reactor technology for utilizing the military Pu (A proposal for such a reactor can be found in Ref. 21). The experience to be gained from this development will be most valuable for the development of the MSTR for transmuting the LWR waste.

## CONCLUDING REMARKS

Based on the preliminary results obtained so far it appears that a molten salt reactor can be designed to be critical when fueled with, and only with the TUS and long lived FPs extractable from the spent fuel from LWRS. The resulting MSTR appears to offer several of

the unique features of the ATW, including the ability to transmute FPs, to fission short lived actinides, and to have a significantly higher fractional transmutation than LMRs and other solid fuel fission reactors. At this point it is not clear to us how much higher need be the critical TU concentration in MSTRS relative to the ATW and, hence, it is not possible to reliably compare the two design approaches.

Nevertheless, in view of the attractive upper bound estimates of the expected performance of MSTRS and of the interesting possibilities they open for the development of the nuclear energy economy, it is recommended to undertake a more thorough study of MSTRS. This study should consider more realistic designs, and account for out-of-core inventories and for relevant design constraints.

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#### REFERENCES

1. J.E. Quinn et al. "Balancing the Fuel Equation with the Advanced Liquid Metal Reactor System (ALMRS)," Proc. Int. Conf. Emerging Nucl. Energy Systems, Monterey, CA June 16-21, 1991. Fusion Technology, 20, No. 4, Part 2, p. 605 (1991).
2. J-S Choi and T.H. Pigford, *Trans. Amer. Nucl. Soc.* 62,104 (1990).
3. R.N. Hill et al., "Physics Studies of Higher Actinide Consumption in an LMR," *Proc. Int. Conf. on the Physics of Fast Reactors*, Marseille, France, April 1990.
4. R.N. Hill, "LMR Design Concepts for Transuranic Management in Low Sodium Void Worth Cores," *Proc. Int. Conf. on Fast Reactors and Related Fuel Cycles*, Kyoto, Japan, Oct. 28- Nov. 1, 1992.
5. C.D. Bowman et al., Nuclear Energy Generation and Waste Transmutation Using an Accelerator-Driven Intense Thermal Neutron Source, " *Nuclear Instrumentation and Methods in Physics Research (Sec. A)*, **A320, 336**,(1992). Also as LA-UR-91-2601 (1991).
6. B.J. Krohn et al., "ATW Neutronic Analysis and Parameter Variation Studies: 800-MeV Protons and a

Molten Lead Target," **LA-UR-92-3469**, Los Alamos National Laboratory (1992).

7. J. Sapir and B.J. Krohn, *Trans. Amer. Nucl. Soc.*, 65, 451, (1992).

8. F. Venneri, C. Bowman and R. Jameson, "Accelerator-Driven Transmutation of Waste (ATW)- A New Method for Reducing the Long-Term Radioactivity of Commercial Nuclear Waste," LA-UR-93-752, Los Alamos National Laboratory (1993).

9. R. N. Hill, Reactor Analysis Div., ANL, Personal Communication to E.G. (1992).

10. E. Greenspan, "Molten Salt Reactors for the Transmutation of Transuranics and Fission Products," Internal Memo, University of California Dept. of Nuclear Engineering, July 15, 1991.

11. Dr. Uri Gat, Oak Ridge National Laboratory, Personal communication to E. G., 1992.

12. M. Taube, *Nucl. Sci. Eng.*, 61,212 (1976).

13. E.H. Ottewitte, "Configuration of a Molten Chloride Fast Reactor on a Thorium Fuel Cycle to Current Nuclear Fuel Cycle Concerns," Ph. D., UCLA (1982).

14. Prof. K. Furukawa, Tokai University, Personal Communication to E.G. (1992).

15. V.D. Kazaritsky et al., "Practical Treatment of Minor Actinides by Single-Fluid Molten Saly Reactor Concepts," *Proc. Int. Conf. Design & Safety of Advanced Nuclear Power Plants*, Tokyo, Japan, 25-29 Oct. 1992.

16. C. V. Parks et al., "SCALE-4 - A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation," CCC-545, ORNL Radiation Shielding Information Center (1990).

17. S.P. Cerne et al., "Reactivity and Isotopic Composition of Spent PWR Fuel as a Function of Initial Enrichment, Burnup, and Cooling Time," ORNL/CSD/TM-244, Oak Ridge National Laboratory (1987). See also O.W. Hermann et al., *Tran. Am. Nucl. Soc.*, 64,147 (1991).

18. J.P. Askew, F.J. Fayers and P.B. Kemshall, *J. British Nuclear Energy Soc.*, **5,564** (1966).

19. Charles Bowman, Personal Communication to E. G., 1992.

20. M. Lammer and O. Schwerer, "Handbook of Nuclear Data for Safeguards," **INDC(NDS)-248**, International Nuclear Data Committee (1991).

21. U. Gat, R.J. Engel and H.L. Dodds, *Nuclear Technology*, 100, Dec. 1992.