

The impact of xenon-135 poisoning on Load Following Transatomic Power Molten Salt Reactor

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INTRODUCTION

First-of-kind civil Molten Salt Reactor (MSR) was developed, built and operated in Oak Ridge National Laboratory (ORNL) in 1960s. It was called Molten Salt Reactor Experiment (MSRE) and at the moment is the only one operated MSR worldwide. Based on experience from this experiment first commercial Molten Salt Breeder Reactor (MSBR) was designed in early 1970s. In the thermal spectrum MSR, fluorides of fissile and/or fertile materials (i.e. UF_4 , PuF_3 and/or ThF_4) are mixed with carrier salts (i.e. LiF) to form a liquid fuel which is circulated in a loop-type primary circuit which lead to following advantages over traditional solid-fueled reactors: (1) relatively low pressure in the primary loop; (2) strong negative thermal feedback; (3) passive decay heat cooling; (4) reduced fuel fabrication costs; (5) online refueling and reprocessing [1].

Nevertheless, cost-competitiveness of such innovative designs in the current domestic energy market may only be feasible with load-following operation. Load following operation has the potential to dramatically increase the commercial competitiveness of nuclear power. Due to increasing penetration of renewables on the electric grid, base-load operation carries the risk of correspondingly frequent negative electric energy pricing. Thus, responsiveness to net electricity demand is essential to market relevance for new designs [2].

The main physical effect that limit the possibilities of power variations in a conventional Light Water Reactor (LWR) is fission product poisoning, especially the xenon effect (few hours after the change in the reactor power level). The ^{135}Xe is the most powerful known neutron absorber (average cross section for thermal neutrons approximately 10^6 barns) with half-life $\tau_{1/2} = 9.17h$ and yield for ^{235}U fission about 6.3%. The vast majority of xenon-135 (6.1%) is produced from another fission product - ^{135}I ($\tau_{1/2} = 6.6h$) [3]. Under normal operating conditions, the ^{135}Xe is burned in the reactor core as it is produced, so while it has a negative impact on the neutron economy, balancing the reactor controls can compensate for its effect. The difficulty comes when the reactor power is reduced and there are fewer neutrons to burn out the ^{135}Xe , so its concentration increases and further suppresses reactor power. In this case, the core takes some time to recover from the power reduction impact of ^{135}Xe . This response to changing power levels, particularly from higher to lower power levels, dramatically slows the reactor's response to power demands. Potentially, ^{135}Xe removal during reactor operation would allow more flexibly vary power levels to follow power demands, typically referred as 'load following'.

The Transatomic Power (TAP) MSR design is a 1250 MW_{th} liquid-fueled reactor has been selected as a prototype of modern MSR. The TAP is an intermediate spectrum reactor

designed to be started with high-assay low-enriched uranium (LEU) uranium as initial fissile load. This work presents modeling and simulation of load following power transient operation of the TAP MSR. We compared these results with well-studied Pressurized Water Reactor (PWR) behavior. This study focuses on the $^{135}\text{Xe}/^{135}\text{I}$ balance in the TAP core and its effect on the reactor performance. Another feature of the MSR, its circulating liquid fuel and corresponding delayed neutron precursor drift, is not treated here.

Much of the analysis herein used a full-core 3-D model of the TAP developed using the continuous-energy Serpent 2 Monte Carlo reactor physics software. The PWR transient analysis has been done for single-assembly model with burnable poison (gadolinium) provided with Serpent [4]. All calculations presented in this paper were performed using the Serpent 2 code version 2.1.31 with JEFF-3.1.2 nuclear data [5].

DESCRIPTION OF THE ACTUAL WORK

Transatomic Power reactor design description

The TAP design is very similar to original MSRE design developed by ORNL [1] but has two major innovations: the fuel salt composition and the moderator. The MSRE's $\text{LiF-BeF}_2\text{-ZrF}_4\text{-UF}_4$ salt has been substituted with LiF-UF_4 salt which allows for an increase in the uranium concentration within the fuel salt from 0.9 to 27.5% while maintaining a relatively low melting point (490°C compared with 434°C for the original MSRE's salt) [6]. The graphite has a very high thermal scattering cross section which makes it a perfect moderator but has a few major drawbacks: (1) the low lethargy gain per collision requires a large volume of moderator to be present to reach criticality, which leads to a larger core and obstructs the core power density; (2) even special reactor-grade graphite has relatively high porosity, consequently, it holds gaseous Fission Products (FPs) (e.g., tritium, xenon) in pores; (3) the reactor graphite lifespan in a commercial reactor is about 10 years [7]. To resolve these issues, the TAP concept uses another moderator, namely, zirconium hydride, allowing for a more compact core and a significant increase in power density. These two innovative design choices, together with a configurable moderator (the moderator-to-fuel ratio can be changed during regular maintenance shutdown), facilitate the commercial deployment of this conceptual design in the current commercially available 5% LEU fuel cycle.

The TAP MSR primary loop contains the reactor core volume (including the zirconium hydride moderator rods with silicone carbide cladding), pumps, and primary heat exchanger. Pumps circulate the LiF-(Act)F_4 fuel salt through the primary loop. The pumps, vessels, tanks, and piping are made of a

nickel-based alloy (similar to Hastelloy-N¹), which is highly resistant to corrosion in various molten salt environments. Inside the reactor vessel, in close proximity to the zirconium hydride moderator rods, the fuel salt is in a critical configuration and generates heat. Table I contains details of the TAP system design which are taken from technical white paper [8] and a neutronics overview [9] as well as ORNL analysis of the TAP design [6, 10].

TABLE I: Summary of principal data for the TAP MSR (reproduced from [8, 10]).

Thermal power	1250 MW _{th}
Electric power	520 MW _e
Gross thermal efficiency	44%
Outlet temperature	620°C
Fuel salt components	LiF-UF ₄
Fuel salt composition	72.5-27.5 mole%
Uranium enrichment	5% ²³⁵ U
Moderator	Zirconium Hydride (ZrH _{1.66}) rods (with silicon carbide cladding)
Neutron spectrum	thermal/epithermal

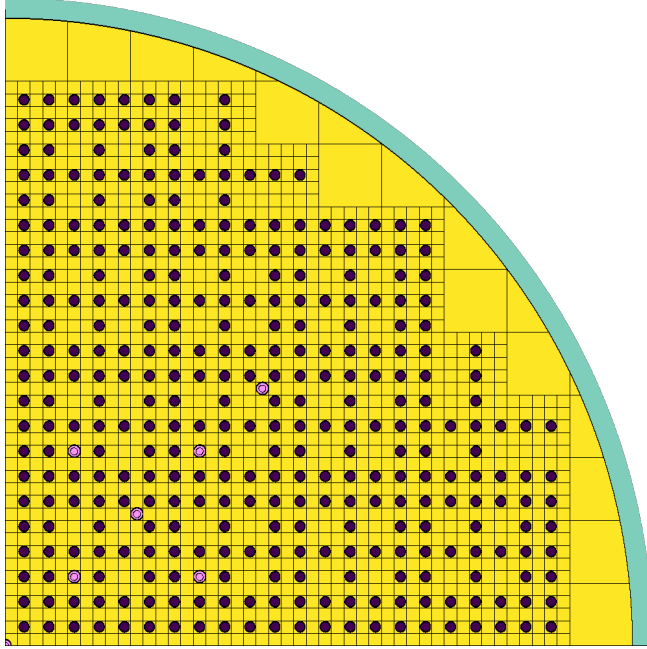


Fig. 1: The TAP MSR schematic core view showing moderator rods (figure reproduced from ORNL/TM-2017/475 [10]).

RESULTS AND ANALYSIS

Using the methodology described previously, the MSBR unit cell depletion analysis was performed to find equilibrium core conditions. Calculation results reported in this section include multiplication factor, neutron flux energy spectrum,

¹ Hastelloy-N is very common in reactors now but have been studied and developed at ORNL in a program that started in 1950s.

and atomic density of major isotopes.

Equilibrium state analysis

This analysis models a single representative unit cell rather than the whole MSBR core. Consequently, it does not take into consideration different fuel-moderator volume ratios for Zone I, Zone II, the annulus, and the reflector. The initial multiplication factor during depletion calculation is selected for a state with fully withdrawn control rods, which gives considerable excess reactivity in the beginning of the cycle (approximately 5000 pcm). The standard deviation for these calculations is approximately 100 pcm. Figure 4 shows the infinite multiplication factor for 1200-days reprocessing cycle calculated by Serpent 2 with ENDF/B-VII.0 nuclear data. A significant standard deviation causes the multiplication factor fluctuations visible in this plot.

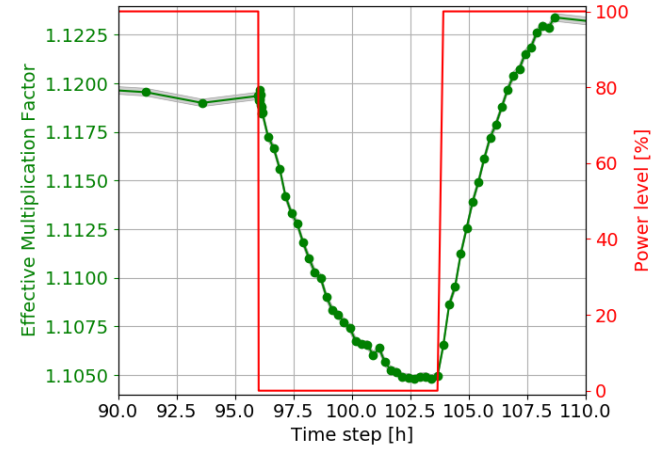


Fig. 2: The TAP MSR schematic core view showing moderator rods (figure reproduced from ORNL/TM-2017/475 [10]).

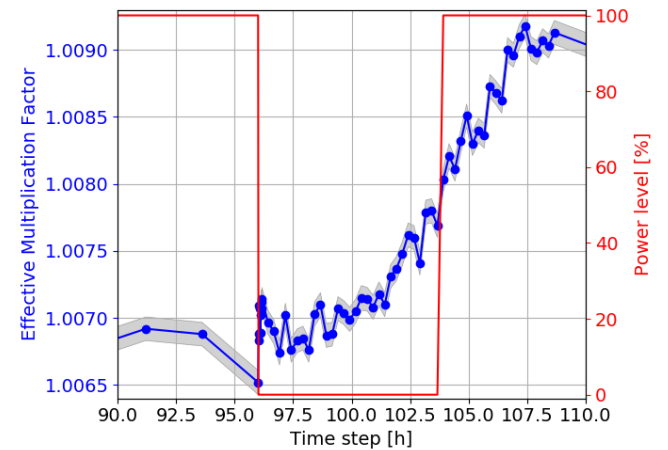


Fig. 3: The TAP MSR schematic core view showing moderator rods (figure reproduced from ORNL/TM-2017/475 [10]).

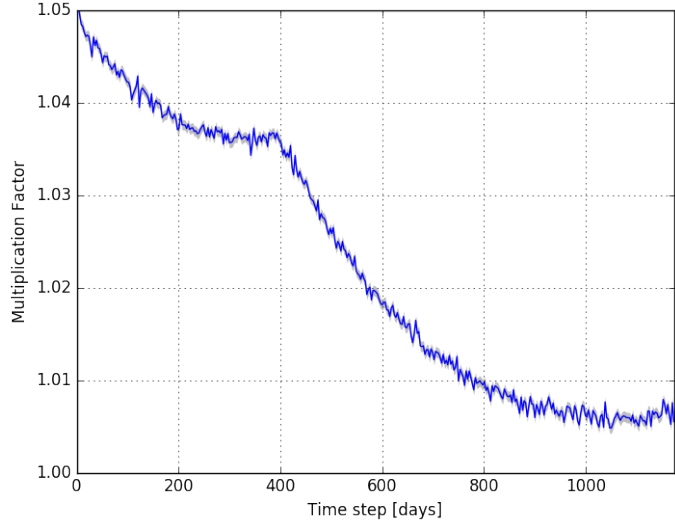


Fig. 4: Infinite multiplication factor during a 1200 days depletion simulation. The confidence interval $\pm\sigma$ is shaded.

Protactinium-233 is continuously removing into the tank for protactinium decay. ^{233}Pa has a half-life of 27 days and beta decays into ^{233}U which as fresh fuel goes back to the reactor core. The infinite multiplication factor decreases first 400 days of depletion due to strong absorbers (e.g. ^{233}Th , ^{234}U) accumulation which causes relatively high fuel (^{233}U) refill inflow to keep reactor critical. During reactor operation producing fissile materials other than ^{233}U in the core (e.g. ^{235}U , ^{239}Pu) which makes it possible to decrease fresh fuel refill rate after 1 year of operation.

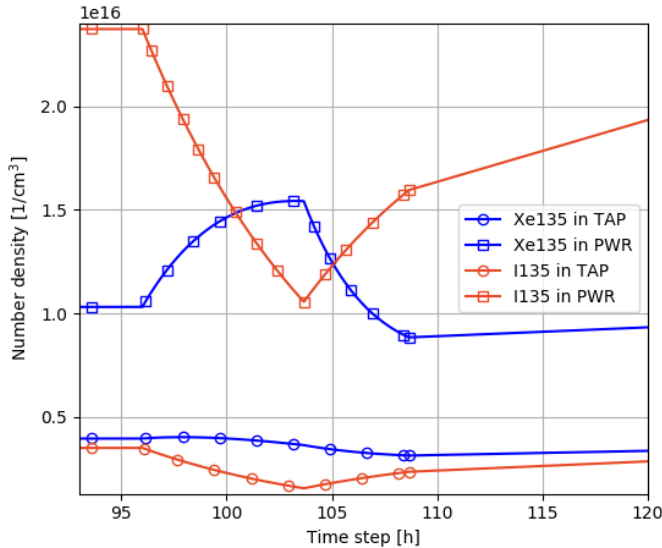


Fig. 5: Normalized number density of major isotopes during 1200 days of depletion.

The analysis of the fuel salt composition variation gives clearer information about the equilibrium core state. Figure 5 shows the normalized number density of isotopes influential to core neutronics at the beginning of each depletion time

interval. The number density of protactinium is very low (less than 10^{16} 1/cm^3) but some small amount of it is produced during the 3-day reprocessing period. In this assessment, the multiplication factor stabilizes after approximately 950 days. Figure ?? represents the rates of online reprocessing material flows over the 4-year depletion calculation. In Figure ?? we can see that, to keep the reactor critical, a higher ^{233}U flow rate from the protactinium decay tank was required for first 400 days of operation. After that, the ^{233}U flow rate can be reduced. The ^{232}Th rate slightly decreases over 4 years of operation due to other than ^{233}U fissile materials accumulation.

As shown in Figure ??, the tank for protactinium decay accumulates ^{233}Pa for approximately 200 days. Fresh ^{233}U fuel flow is also established after 200 days. Uranium produced in the tank by protactinium decay is separated by circulation of the salt through a fluorinator. The fully processed molten salt, on its way back to the primary loop, has uranium added at the rate required to maintain or adjust the fissile material concentration and, hence, the reactivity, in the reactor core as desired.

Neutron spectrum

Figure 6 represents the neutron flux per lethargy energy distribution of the initial and the equilibrium core compositions. The spectrum for the equilibrium state is harder than the initial state due to heavy fission products.

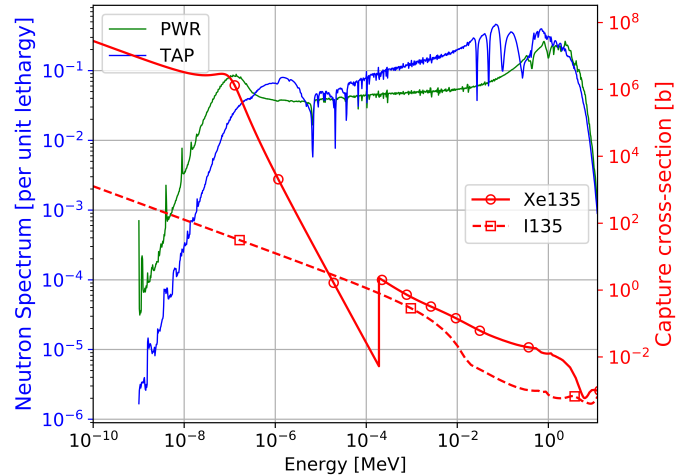


Fig. 6: Neutron spectra for PWR and TAP vs ^{135}Xe and ^{135}I capture cross-section.

In this work, only the Zone I unit cell, where the fuel salt volume fraction is 13.2%, was considered. The neutron spectrum for Zone II cell, where the fuel salt volume fraction is 37% is expected to be harder and the peak in thermal energy region is predicted to be much lower. To obtain a high-fidelity neutron energy spectrum, a full-core MSBR analysis is required.

CONCLUSIONS

The depletion calculation of the MSBR unit cell model for finding the equilibrium states was performed using the

Serpent 2 Monte Carlo code to simulate simplified case of the online reprocessing and refueling to find equilibrium material composition. When running depletion calculation, the fission products are removed and fertile/fissile materials are added to fuel salt every 3 days. The important MSR feature, online reprocessing & refueling is implemented in the Serpent 2 material burnup routine. The results of this study indicate that from the depletion calculation the multiplication factor slowly decreases and reaches to the equilibrium state. The most obvious finding to emerge from the analysis of initial and equilibrium materials composition is that neutron energy spectrum is harder for equilibrium state because significant amount of heavy fission products were accumulated in the MSBR core.

These results are contrary with those of Jeong and Park (2016) who suggested two different unit cell models and uses Monte Carlo N-Particle code (MCNP) with Python-script to simulate MSBR online reprocessing to find equilibrium composition. This inconsistency may be due to different fuel fraction in the unit cell (Jeong and Park selected 20.6% salt fraction). To obtain better results for this online reprocessing simulation, many future efforts are planned. First, a depletion simulation will be performed using a full-core, three-dimensional, high-fidelity model of MSBR that has been developed in Serpent 2. In this case, different fuel-moderator volume ratios for the reactor Zone I, Zone II, annulus, and reflectors will be taken into account to find accurate multiplication factor, neutron spectrum and, hence, depleted composition. Secondly, an additional Serpent 2 flow control system subroutine should be developed to simulate adjusting material flows (e.g. rate of removing ^{233}Pa from the salt and adding fissile ^{233}U from the tank for protactinium decay) depending upon the instantaneous reactivity value, which is a more promising reactivity control method than moving control rods. Finally, the temperature effect of reactivity for both fuel salt and graphite should be calculated to find optimal effective multiplication factor range.

REFERENCES

1. P. N. HAUBENREICH and J. R. ENGEL, "Experience with the Molten-Salt Reactor Experiment," *Nuclear Technology*, **8**, 2, 118–136 (Feb. 1970).
2. ENERGY INFORMATION ADMINISTRATION, "U.S. Electric System Operating Data," (2016).
3. A. LOKHOV, "Technical and economic aspects of load following with nuclear power plants," *NEA, OECD, Paris, France* (2011).
4. J. LEPPANEN, M. PUSA, T. VIITANEN, V. VALTAVIRTA, and T. KALTIAISENAHO, "The Serpent Monte Carlo code: Status, development and applications in 2013," *Annals of Nuclear Energy*, **82**, 142–150 (Aug. 2015).
5. O. D. BANK, "The JEFF-3.1.2 Nuclear Data Library," Tech. Rep. JEFF Report 24, OECD/NEA Data Bank (2014).
6. B. R. BETZLER, J. J. POWERS, A. WORRALL, S. ROBERTSON, L. DEWAN, and M. MASSIE, "Two-Dimensional Neutronic and Fuel Cycle Analysis of the Transatomic Power Molten Salt Reactor," Tech. Rep. ORNL/TM-2016/742, Oak Ridge National Lab.(ORNL) (2016).
7. R. C. ROBERTSON, "Conceptual Design Study of a Single-Fluid Molten-Salt Breeder Reactor." Tech. Rep. ORNL-4541, comp.; Oak Ridge National Lab., Tenn. (Jan. 1971).
8. TRANSATOMIC POWER CORPORATION, "Technical White Paper," White Paper 2.1, Transatomic Power Corporation, Cambridge, MA, United States (Nov. 2016).
9. TRANSATOMIC POWER CORPORATION, "Neutronics Overview," White Paper 1.1, Transatomic Power Corporation, Cambridge, MA, United States (Nov. 2016).
10. B. R. BETZLER, S. ROBERTSON, T. E. E. DAVIDSON, J. J. POWERS, and A. WORRALL, "Assessment of the Neutronic and Fuel Cycle Performance of the Transatomic Power Molten Salt Reactor Design," Tech. Rep. ORNL/TM-2017/475, Oak Ridge National Lab. (ORNL), Oak Ridge, TN (United States) (2017).