Modeling and Simulation of Online Reprocessing in the Thorium-Fueled Molten Salt Breeder Reactor Response to Review Comments

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Review General Response

We would like to thank the reviewers for their detailed assessment of this paper. Your suggestions, clarifications, and comments have resulted in changes which certainly improved the paper.

Reviewer 1

1. Overall, a well written article in a topic area of interest to a number of audiences. Technically very good, well written in the description and analysis. Well done authors!

A few minor comments/suggestions/clarifications as follows:

Solution: Thank you for these kind comments. Your suggestions have certainly improved the paper.

2. Abstract: Given the recent announcement about TransAtomic Power, you may want to remove them from your list?

Solution: Thank you for your kind review. The current manuscript was submitted before announcement that TransAtomic is ceasing operations. It is removed from the list of MSR startups (indeed, the whole list was removed based on a comment by another reviewer).

3. Table 1, page 3: SCALE/TRITON is fast as well as thermal reactor capable.

Solution: Thank you for the recommendation. In Table 1 "thermal" has been changed to "thermal/fast".

4. Page 4, Line 48: Should read "...and refill using a single or multiple unit cell"

Solution: That statement has been modified as requested.

5. Page 4, Line 55. Also worth noting that the latest SCALE release will have the same functionality using continuous removal (B. R. Betzler, J. J. Powers, N. R. Brown, and B. T. Rearden, "Molten Salt Reactor Neutronics Tools in SCALE," Proc. M&C 2017 - International Conference on Mathematics & Computational Methods Applied to Nuclear Science and Engineering, Jeju, Korea, Apr. 16-20 (2017).)

Solution: Thank you for the update. This sentence has been added:

"The latest SCALE release will also have the same functionality using truly continuous removals [1]."

6. Page 8, Fig 2: Appears the image is cut off at the top?

Solution: The image has been replotted.

7. Page 13, line 209: The description of the Pa removal, although correct, isn't quite fully correct. The reason the Pa is removed from the core and hence flux is to then enable the Pa to decay to U233. If it was left in the core, it would transmute further and hence not be able to produce the U233 that is necessary for this breeding cycle to work.

Solution: "Protactinium presents a challenge, since it has a large absorption cross section in the thermal energy spectrum. Moreover, ²³³Pa left in the core would produce ²³⁴Pa and ²³⁴U, neither of which are useful as fuel, would produce a smaller amount of ²³³Pa which decays into the fissile ²³³U. Accordingly, ²³³Pa is continuously removed from the fuel salt into a protactinium decay tank to allow ²³³Pa to decay to ²³³U without the corresponding negative neutronic impact."

8. Table 3: "cycle time" is not defined in the paper. Please add.

Solution: The "cycle time" definition has been added at the first appearance in text.

9. Page 14, line 224. The 3 day time step as the "optimum" for Th fuel cycles in an MSR was first described and concluded by Powers et al. Please add a reference to their initial work.

Solution: The reference to [2] has been added.

10. Page 14, line 234 onwards: Doesn't SERPENT already have an MSR removal capability? If so, what is different about using SaltProc with SERPENT?

Solution: It does. Indeed, much of the inspiration for SaltProc was the lack of documentation, difficulty of use, and various inexplicable results we acquired when we tried use this built-in capability. This capability has not performed well and needs to be better documented and more robustly verified before we can use it. We fully intend to contribute to its improvement if it can be made to agree with expectations. Additional text to clarify this point has been add to solution for question 23.

11. Page 18, Figure 7: The figure is hard to interpret or see clearly what is going on. Could an additional figure or a zoomed in portion be added to show what the swing in k is over a much shorter time interval? k seems to be swinging dramatically but over what time period and how would this be controlled in reality? The graph almost suggests that the core is unstable??

Solution: Zoomed portion for 150 EFPD interval has been added. We also added notes on a plot to explain swing in multiplication factor.

12. Page 18, line 327: Are those elements removed every 3435 days, or is it that the entire salt core is discharged?

Solution: 100 % of those elements atoms are removed every 3435 days. Full salt discard as it mentioned in Table 3 has not been implemented. More detailed explanation of this has been added:

"Additionally, the presence of rubidium, strontium, cesium, and barium in the core are disadvantageous to reactor physics. Overall, the effective multiplication factor gradually decreases from 1.075 to ≈ 1.02 at equilibrium after approximately 6 years of irradiation.

In fact, SaltProc fully removes all of these elements every 3435 days (not a small mass fraction every 3 days) which causes the multiplication factor to jump by approximately 450 pcm, and limits using the batch approach for online reprocessing simulations. In future versions of SaltProc this drawback will be eliminated by removing elements with longer residence times (seminoble metals, volatile fluorides, Rb, Sr, Cs, Ba, Eu). In that approach, chemistry models will inform separation efficiencies for each reprocessing group and removal will optionally be spread more evenly across the cycle time."

13. Page 19, Figure 8 (and same for Fig 9): y-axis in grams/kgs or mass units would be better for the reader.

Solution: Thank you for the recommendation. Atom density was chosen for publication-to-publication comparison (e.g. Park *et al.* and Betzler *et al.* [3, 4]). Although mass would certainly be more understandable for the reader and will be added in a future releases.

14. Page 20, Fig 9: What are the wiggles and dips , especially seen for Np235?

Solution: Explanation of this phenomenon has been added as follows:

"Small dips in neptunium and plutonium number density every 16 years are caused by removing 237 Np and 242 Pu (included in Processing group "Higher nuclides", see Table 3 which decays into 235 Np and 239 Pu, respectively."

15. Page 20, line 351: It is more than just the Pu isotopes that makes the spectrum harder? What about the other MAs etc?

Solution: Thank you for the excellent point. The corrected sentence reads thus:

"The neutron energy spectrum at equilibrium is harder than at startup due to plutonium and other strong absorbers accumulating in the core during reactor operation."

16. Fig 12: units on y-axis?

Solution: Thanks for catching this. The units $\frac{n}{cm^2s}$ have been added.

17. Page 24, line 389: Should that be "233U" and not "233Th"?

Solution: Yes, we meant 233 U production. The typo has been fixed. Thanks so much for catching it!

18. Table 5: Please provide some comments on the uncertainties - where do they come from? Also, the "reference" results need to state whether "initial" or "equilibrium"

Solution: In Table 5, we added information in the reference column containing data for initial fuel salt composition. Details about uncertainties have been added:

"Table 5 summarizes temperature effects on reactivity calculated in this work for both initial and equilibrium fuel compositions, compared with the original Oak Ridge National Laboratory (ORNL) report data [5]. By propagating the k_{eff} statistical error provided by SERPENT2, uncertainty for each temperature coefficient was obtained and appears in Table 5. Other sources of uncertainty are neglected, such as cross section measurement error and approximations inherent in the equations of state providing both the salt and graphite density dependence on temperature."

19. Page 26, line 425: "Relatively large" compared with what? Perhaps results for an LWR case would be a good basis for comparison?

Solution: The sentence has been extended as follows:

The moderator temperature coefficient (MTC) is positive for the startup composition and decreases during reactor operation because of spectrum hardening with fuel depletion. Finally, the total temperature coefficient of reactivity is negative for both cases, but decreases during reactor operation due to spectral shift. In summary, even after 20 years of operation the total temperature coefficient of reactivity is relatively large and negative during reactor operation (comparing with conventional PWR which has temperature coefficient about -1.71 pcm/°F \approx -3.08 pcm/K [6]), despite positive MTC, and affords excellent reactor stability and control.

20. Page 27, section 3.8: It needs to be made more clear that these results were calculated, and that they are taken from the code output.

Solution: This has now been clarified in the text:

"Table 7 summarizes the six factors for both initial and equilibrium fuel salt composition. Using SERPENT2 and SaltProc, these factors and their statistical uncertainties have been calculated for both initial and equilibrium fuel salt composition (see Table 2)."

21. Page 29, Figure 15: Similar comment to above regarding Fig 7 - the results are difficult to see and interpret with such notable swings.

Solution: Zoomed portion for 150 EFPD interval has been added along with clarifying details.

Reviewer 2

22. The paper presents a python script (SaltProc) that can complement the Serpent 2 code capabilities in terms of fuel cycle analysis of MSRs. The paper is well written and the tool might be useful for the nuclear engineering community. However, before publishing the paper, I recommend some revisions.

Solution: Thanks very much for these comments. We appreciate your detailed review, which has certainly improved the paper.

23. The most critical point of the work is that the built-in capabilities for online reprocessing of Serpent 2 have not been used. Their use is mentioned in the future work, but it is not clear why these capabilities have not been used in the current work. To the author's knowledge, they have been available in Serpent 2 since quite a while. The authors should clarify this point at the beginning of the paper, and not only in the "Future work" section. Even though the technical

work was done without using these capabilities, they should highlight what SaltProc adds to the built-in Serpent capabilities, and they should at least try to extrapolate on the potential advantages of combining SaltProc and Serpent capabilities. Based on this, they should slightly restructure the paper in order to prove the claimed advantages over Serpent 2.

Solution: Thank you for your helpful review. We tried to use these capabilities [7] but have had a number of issues which are hard to resolve due to lack of documentation and published verifications of this feature. The following paragraph has been added to clarify why these capabilities have not been used in current work:

"Aufiero *et al.* added an undocumented feature to SERPENT2 using a similar methodology by explicitly introducing continuous reprocessing in the system of Bateman equations and adding effective decay and transmutation terms for each nuclide [8]. This was employed to study the material isotopic evolution of the Molten Salt Fast Reactor (MSFR)[8]. The developed extension directly accounts for the effects of online fuel reprocessing on depletion calculations and features a reactivity control algorithm. The extended version of SERPENT2 was assessed against a dedicated version of the deterministic ERANOS-based EQL3D procedure in [9, 10] and adopted to analyze the MSFR fuel salt isotopic evolution.

We employed this built-in SERPENT2 feature for a simplified unit-cell geometry of the thermal spectrum thorium-fueled Molten Salt Breeder Reactor (MSBR) and found it unusable¹. Primarily, it is undocumented, and the discussion forum for SERPENT users is the only useful source of information at the moment. Additionally, the reactivity control module described in Aufiero *et al.* is not available in the latest SERPENT 2.1.30 release. Third, the infinite multiplication factor behavior for simplified unit-cell model obtained using SERPENT2 built-in capabilities [7] does not match with exist MCNP6/Python-script results for the similar model by Jeong and Park² [11]. Finally, only two publications [8, 12] using these capabilities are available, reflecting the reproducibility challenge inherent in this feature.

If these challenges can be overcome through verification against ChemTriton/SCALE as well as this work (the SaltProc/SERPENT2 package), we hope to employ this SERPENT2 feature for removal of fission products with shorter residence time (e.g., Xe, Kr), since these have a strong negative impact on core lifetime and breeding efficiency."

24. In addition to this major point, I would suggest a few other revisions. Considering the scope of the journal, mentioning the names of companies and start-up is not appropriate. I would suggest removing them.

Solution: The names of companies have been removed.

25. The sentence "Immediate advantages over traditional, solid-fueled, reactors include near-atmospheric pressure in the 15 primary loop, relatively high coolant temperature, outstanding neutron economy, improved safety parameters," may suggest improved neutron economy vs solid-fuel fast reactors. This is rarely the case, especially vs Pu-based SFRs. I would suggest reformulating.

Solution: Thank you for the exceptional recommendation. The sentence has been split into two, as follows:

Immediate advantages over traditional commercial reactors include near-atmospheric pressure in the primary loop, relatively high coolant temperature, outstanding neutron economy, and improved safety parameters. Advantages over solid-fueled reactors in general include reduced fuel preprocessing and the ability to continuously remove fission products and add fissile and/or fertile elements [13].

26. The sentence "With regard to the nuclear fuel cycle, the thorium cycle produces a reduced quantity of plutonium and minor actinides (MAs) compared to the traditional uranium fuel cycle" is correct, but the fact that this is an advantage is questionable. The pros&cons of thorium cycle have been long debated and there is no consensus on its advantage in terms of exposure of workers, exposure of public, geological repository, etc. I would suggest removing the sentence.

Solution: Thanks for your insightful comment. The sentence has been removed.

27. "Methods listed in references [14, 17, 24, 25, 28, 29, 30] as well as the current work also employ a batch-wise approach". As a matter of fact, the work in [14] allows for continuous reprocessing via introduction of "reprocessing" time constants. The work from Aufiero (mentioned in the following paragraph) actually used the methodology previously developed in [14] for verification purposes.

Solution: Thank you for the information. That reference has been removed from the sentence, and following paragraph has been modified to read:

Accounting for continuous removal or addition presents a greater challenge since it requires adding a term to the Bateman equations. Fiorina *et al.* simulated MSFR depletion with continuous fuel salt reprocessing via introducing "reprocessing" time constants into the ER-ANOS transport code [10]. The latest SCALE release will also have the same functionality using truly continuous removals [1]. A similar approach is adopted to model true continuous feeds and removals using the MCNP transport code listed in references [14, 15, 16].

28. Table 3. It is not clear what "effective cycle times" are. Please clarify.

Solution: The "cycle time" definition has been added to its first appearance in text.

29. The removal of fission products is made batch wise in the described algorithms. However it is not clear how the fission products with the longest "effective cycle times" are removed. Part of them at every batch? Or all of them at the end of the "effective cycle times". Please clarify. And please clarify the relation between "effective cycle times", batches and the average time spent by a fission product in the reactor.

Solution: All of them at the end of the cycle time, and we agree that it was not best solution. Feature improvements in SaltProc are underway to enable more realistic handling times in the processing heuristics. To clarify this following paragraph has been added:

The current version of SaltProc only allows 100% separation efficiency for either specific elements or groups of elements (e.g. Processing Groups as described in Table ??) at the end of the specific cycle time. This simplification neglects the reality that the salt spends appreciable time out of the core, in the primary loop pipes and the heat exchanger.

This approach works well for fast-removing elements (gases, noble metals, protactinium) which should be removed each depletion step. Unfortunately, for the elements with longer cycle times (i.e. rare earths should be removed every 50 days) this simplified approach leads to oscillatory behavior of all major parameters. In future releases of SaltProc, this drawback will be eliminated by removing elements with longer cycle times using different method: only mass fraction (calculated separately for each reprocessing group) will be removed each depletion step or batch (e.g. 3 days in the current work).

30. The oscillatory behavior shown in Fig. 7 on the time scale of months/years is hard to explain. Is this because the fission product with longer residence time are batch-wise removed at their "effective cycle time"? In case, why not to remove part of them at every depletion step?

Solution: Yes, the oscillation happened because the fission products with longer residence time are removed at the end of cycle time. We definitely will take your advice and improve the code in future releases. The following text has been added to clarify this issue:

In fact, SaltProc fully removes all of these elements every 3435 days (not a small mass fraction every 3 days) which causes the multiplication factor to jump by approximately 450 pcm, and limits using the batch approach for online reprocessing simulations. In future versions of SaltProc this drawback will be eliminated by removing elements with longer residence times (seminoble metals, volatile fluorides, Rb, Sr, Cs, Ba, Eu). In that approach, chemistry models will inform separation efficiencies for each reprocessing group and removal will optionally be spread more evenly accross the cycle time.

31. Can the proposed tool adjust reactivity?

Solution: No, and we may someday add this capability, but we expect this will be a challenge outside of the depletion code.

32. "The main physical principle underlying the reactor temperature feedback is an expansion of material that is heated". This sentence would deserve some support data. Can you please calculate separately the effect of temperature (Doppler in fuel and spectral shift in graphite) and density?

Solution: Thank you for the excellent recommendation. Effects of temperature and density have been calculated separately and added in Table 5. Moreover, during these simulations we have discovered mistake in fuel salt and graphite density correlations and completely recalculated temperature coefficients of reactivity with lower statistical error. Now initial total temperature coefficient is closer to the reference and statistical uncertainty was reduced from 0.046 to 0.038 pcm/K.

33. How uncertainties have been calculated in Table 5? Is this just statistical uncertainty from Serpent calculations? Please clarify

Solution: Yes, uncertainties were determined from statistical error from SERPENT output. Following passage has been added to clarify this point:

By propagating the k_{eff} statistical error provided by SERPENT2, uncertainty for each temperature coefficient was obtained and appears in Table ??. Other sources of uncertainty are neglected, such as cross section measurement error and approximations inherent in the equations of state providing both the salt and graphite density dependence on temperature.

34. For calculating the coefficients in table 5, are you only changing densities, or also dimensions? Please clarify and provide a justification.

Solution: We have changed both densities and dimensions. This passage has been modified to read:

A new geometry input for SERPENT2, which takes into account displacement of graphite surfaces, was created based on this information. For calculation of displacement, it was assumed that the interface between the graphite reflector and vessel did not move, and that the vessel temperature did not change. This is the most reasonable assumption for the short-term reactivity effects because inlet salt is cooling graphite reflector and inner surface of the vessel.

35. "The fuel temperature coefficient (FTC) is negative for both initial and equilibrium fuel compositions due to thermal Doppler broadening of the resonance capture cross sections in the thorium." What is the effect of density?

Solution: This passage has been modified to illuminate the effect of density:

The fuel temperature coefficient (FTC) is negative for both initial and equilibrium fuel compositions due to thermal Doppler broadening of the resonance capture cross sections in the thorium. A small positive effect of fuel density on reactivity increases from +1.21 pcm/K at reactor startup to +1.66 pcm/K for equilibrium fuel composition which has a negative effect on FTC magnitude during the reactor operation.

36. "This thorium consumption rate is in good agreement with a recent online reprocessing study by ORNL [29]." Please notice that in a reactor with only Th as feed, and near equilibrium, the Th consumption rate is exclusively determined by the reactor power and by the energy released per fission.

Solution: Thank you for the recommendation. Following sentence has been added:

It must be noted that for the reactor with only thorium feed, at near equilibrium state, the thorium consumption rate is determined by the reactor power, the energy released per fission, and neutron energy spectrum.

37. Are you considering the effect of the gradual poisoning of the graphite with fission products? If not, it would be worthwhile briefly discussing its effect.

Solution: This passage has been added:

 135 Xe is a strong poison to the reactor, and some fraction of this gas is absorbed by graphite during MSBR operation. ORNL calculations shown that for unsealed commercial graphite with helium permeability 10^{-5} cm²/s the calculated poison fraction is less than 2% [5]. This parameter can be improved by using experimental graphites or by applying sealing technology. The effect of the gradual poisoning of the core graphite with xenon is not treated here.

38. In the manuscript it is not always clear when the authors refer to numerical approximations of physical situations. For instance, the authors write "Figure 16 demonstrates that batch-wise removal of strong absorbers every 3 days did not necessarily leads to fluctuation in results but rare earth elements 480 removal every 50 days causes an approximately 600 pcm jump in reactivity." These 600 pcm are an effect of a numerical approximation, but the way things are presented can be confusing to the reader. Please try to explicitly separate physical and numerical effects. And try to related numerical effects to physical effects. For instance, how does these numerical "jumps" affect the results? In this sense, why the batch-wise removal of strong absorbers every 3 days was not done? And why a fraction of rare earths is not removed every three days?

Solution: This sentence was quite unclear and we have rewritten it to address your concerns. Effectively these simulations exactly simulated the batch-wise refuelling detailed in the Robertson design document, while a more realistic approach would give smooth behavior. A description of the changes to the text appears in solution 30.

39. "The current work results show that a true equilibrium composition cannot exist but balance between strong absorber accumulation and new fissile material production can be achieved to keep the reactor critical." Not clear. Do you mean that the equilibrium composition cannot be achieved in a lifetime of the reactor? Please clarify **Solution:** This statement, being unclear and misleading, has been removed completely. The intention was to communicate that for a relaxed definition of equillibrium, it can be acheived.

Reviewer 3

40. This paper presents a SaltProc-Serpent2 coupled code system to simulate the depletion and online reprocessing process by directly changing the isotropic composition of fuel salt. The Molten Salt Breeder Reactor (MSBR) was analyzed to demonstrate the simulation capability of the developed code system. However, large number of similar works have been done and published on this topic. In particular, this work is similar with that done by Park et al. with the title of "Whole core analysis of molten salt breeder reactor with online fuel reprocessing" published on "International Journal of Energy Research". The authors need to prove the uniqueness and innovation within their work.

Solution: We appreciate this comment and have made clarifying improvements to indicate the impact of this work throughout the paper. More detail is described regarding these changes in the specific comment responses below.

41. What are the main differences of this work with the previous works, especially with the work published by Park at al. in "Whole core analysis of molten salt breeder reactor with online fuel reprocessing"?

Solution: Thank you for your question. The new paragraph has been added:

The works described in [3] and [11] are most similar to the work presented in this paper. However, a few major differences follow: (1) Park *et al.* employed MCNP6 for depletion simulations while this work used SERPENT2; (2) the full-core reactor geometry herein is more detailed [17]; (3) Park *et al.* and Jeong *et al.* both only considered volatile gas removal, noble metal removal, and ²³³Pa separation while the current work implemented the more detailed reprocessing scheme specified in the conceptual MSBR design [5]; (4) the ²³²Th neutron capture reaction rate has been investigated to prove advantages of two-region core design; (5) the current work explicitly examines the independent impacts of removing specific fission product groups.

42. How did the authors verify the coupled SaltProc and Serpent code system?

Solution: We have compared few parameters (multiplication factor, Th refill rate, neutron energy spectrum) with Betzler *et al.* [4] and mentioned it in the Result section. We also compared neutron energy spectrum and temperature coefficients for equilibrium composition with Park *et al.* [3]. In a future SaltProc release suite of unit tests will be added to make sure that results are consistent with our test cases.

43. In Page 3, the title of "Table 1" should not only contain the "fast spectrum system". The work published by Zhou and Yang et al. with the title of "Fuel cycle analysis of molten salt reactors based on coupled neutronics and thermal-hydraulics calculations" needs to be included in Table 1 for completeness.

Solution: Thank you for your excellent recommendations. The table has been enriched as requested.

44. In Page 5, the following sentence needs to be explained. "We employed this extended SERPENT 2 for a simplified unit-cell geometry of thermal spectrum thorium-fueled MSBR and obtained results which contradict existing MSBR depletion simulations."

Solution: This statement has been significantly extended (see solution 23).

45. In Page 10, "SERPENT generates the problem-dependent nuclear data library", how did SER-PENT generate the problem-dependent nuclear data library? What kind of nuclear data library did SERPENT generate?

Solution: Thank you for your kind review. The statement has been removed entirely and we emphasized that temperature of each material is assumed to be constant over 60 years:

The specific temperature was fixed for each material and did not change during the reactor operation.

46. In Page 14, "depletion calculations" should be "depletion calculations".

Solution: Thanks for catching this, fixed.

47. In Page 21, it looks like the neutron spectrum is not normalized in Figure 10. It is recommended to normalize the neutron spectrum for comparison.

Solution: Thank you for the comment. Neutron energy spectrum in Figure 10 and 11 are normalized per unit lethargy and the area under the curve is normalized to 1. The caption has been edited to make this more clear.

48. In Page 22, "Figure 13 reflects the normalized power distribution of the MSBR quarter core, which is the same at both the initial and equilibrium states" contradicts the following statement of "The spectral shift during reactor operation results in different power fractions at startup and equilibrium".

Solution: Thanks for catching this. The difference between power fraction is very small that could be seen from Table 4. It is impossible to see the difference in a contour plot that is why we left only equilibrium composition on Figures 13 and 14. The paragraph has been modified as follows:

Table 4 shows the power fraction in each zone for initial and equilibrium fuel compositions. Figure 13 reflects the normalized power distribution of the MSBR quarter core for equilibrium fuel salt composition. For both the initial and equilibrium compositions, fission primarily occurs in the center of the core, namely zone I. The spectral shift during reactor operation results in slightly different power fractions at startup and equilibrium, but most of the power is still generated in zone I at equilibrium (table 4).

49. In Page 24, it is hard to agree with the statement that "the majority of 233Th is produced in zone II.". How did the authors draw this conclusion?

Solution: The statement has been removed entirely.

50. In Page 24 and 25, why did the normalized power density distribution and the 232Th neutron capture reaction rate distribution share the same figure for both initial and equilibrium fuel salt compositions?

Solution: Thanks for catching this, it is a typo. Figure 13 and 14 are plotted for equilibrium composition only.

51. Too much background information was contained in the abstract, which deteriorates the readability of the abstract.

Solution: The abstract has been shortened.

References

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