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 comments have resulted in changes which certainly improved the paper.

Reviewer 1

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

   Solution: Thank you for your kind review. Current manuscript was submitted before announcement that TransAtomic is ceasing operations. It is removed from the list of MSR startups.

2. 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".

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

   Solution: That statement has been modified as requested.

4. 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].

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

Solution: The image has been replotted.

6. 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: Thanks for catching this. Following passage has been added:

Protactinium presents a challenge, since it has a large absorption cross section in the thermal energy spectrum. Moreover, ^{233}Pa left in the core would produce ^{234}Pa and ^{234}U, which both are not useful as fuel, and smaller amount of ^{233}Pa would decays into the fissile ^{233}U. Accordingly, ^{233}Pa is continuously removed from the fuel salt into a protactinium decay tank to allow ^{233}Pa to decay to ^{233}U without negative impact on neutronics.

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

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

8. 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.

9. 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. We tried use it before but these capabilities needs to be verified. Additional text to clarify this point has been added to solution for question 21.

10. 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.

11. 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 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. In fact, SaltProc fully removes all 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 cycle times (seminoble metals, volatile fluorides, Rb, Sr, Cs, Ba, Eu) using different approach. Only mass fraction (calculated separately for each reprocessing group) will be removed every depletion step (e.g. 3 days) instead of removing 100% of element atoms after cycle time.

12. 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.

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

> **Solution:** Explanation of this phenomena has been added as follows:
>
> Small dips in neptunium and plutonium number density every 16 years are caused by removing ^{237}Np, ^{242}Pu (included in Processing group "Higher nuclides", see Table 3) which decays into ^{235}Np, ^{239}Pu, respectively.

14. 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 accumulation in the core during reactor operation.

15. Fig 12: units on y-axis?

Solution: Thanks for catching this. Units $\frac{n}{cm^2 s}$ has been added.

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

Solution: Yes, we meant ^{233}U production. The typo has been fixed.

17. 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 the Table 5 was added information that reference column contains data for initial fuel salt composition. Details about uncertainties have been added:

Uncertainty for each temperature coefficient was obtain by propagating statistical error of effective multiplication factor calculation provided by SERPENT2 and also appears in Table 5. Other sources of uncertainty, such as cross section libraries uncertainty, error in salt and graphite density correlations, are not treated here.

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

Solution: Sentence has been extended as follows:

...the total temperature coefficient of reactivity is relatively large and negative during reactor operation (comparing with conventional PWR which temperature coefficient about -1.71 pcm/°F $\approx$ -3.08 pcm/K [5]), despite positive MTC, and affords excellent reactor stability and control.

19. 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. These factors and their statistical uncertainties have been calculated using SERPENT2 code for initial fuel salt composition (see Table 2) and for equilibrium salt composition which was obtained with SaltProc.

20. 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 and clarifying notes has been added.

Reviewer 2

21. 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 kind review. We tried to use these capabilities [6] but have had number of issues which are hard to resolve due to lack of documentation and publications involving this feature. Following paragraph has been added to clarify why these capabilities have not been used in current work:

We employed this extended SERPENT2 for a simplified unit-cell geometry of thermal spectrum thorium-fueled MSBR and have had following problems: (1) lack of documentation describing how to use this built-in SERPENT2 online reprocessing capabilities [1] (Discussion forum for SERPENT users is only useful source of information at the moment); (2) reactivity control module described in Aufiero *et al.* is not available in the latest SERPENT 2.1.30 release; (3) infinite multiplication factor behavior for simplified unit-cell model obtained using SERPENT2 built-in capabilities [6] does not match with exist MCNP6/Python-script results for the similar model by Jeong and Park[2] [7]; (4) only few publication [8, 9] using these capabilities are available which is indicative of lack of reproducibility. Nevertheless, truly continuous online reprocessing capabilities of SERPENT2 is desirable feature for our future research. However, these capabilities should be carefully verified against ChemTriton/SCALE or proposed SaltProc/SERPENT2 package, and could be employed for removal of fission products with shorter residence time (e.g., Xe, Kr) which have a strong negative impact on core lifetime and breeding efficiency which we intent to do in the nearest future.

22. 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.

23. 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 changed as follows:
>
> "Immediate advantages over traditional light water reactors include..."

24. 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 kind comment. The sentence has been removed.

25. "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. [14] 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 ERANOS transport code [10]. Aufiero *et al.* improved SERPENT2 using similar methodology by explicitly introducing continuous reprocessing in the system of Bateman equations by adding effective decay and transmutation terms for the different nuclides [8].

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

> **Solution:** The "cycle time" definition has been added in a first appearance in text.

27. 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. To clarify this following paragraph has been added:

Current version of SaltProc only allows separate out 100% of specific elements or group of elements (e.g. Processing Groups as described in Table 3 at the end of the specific cycle time. 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 time (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 (e.g. 3 days).

28. 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 are definitely will take your advice and improve the code in future releases. Following text has been added to clarify this issue:

In fact, SaltProc fully removes all 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 time (seminoble metals, volatile fluorides, Rb, Sr, Cs, Ba, Eu) using different approach. Only mass fraction (calculated separately for each reprocessing group) will be removed every depletion step (e.g. 3 days) instead of removing 100% of element atoms after cycle time.

29. Can the proposed tool adjust reactivity?

Solution: No, and we do not plan to add this capability.

30. "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.

31. 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:

Uncertainty for each temperature coefficient was obtain by propagating statistical error of effective multiplication factor calculation provided by SERPENT2 and also appears in Table 5. Other sources of uncertainty, such as cross section libraries uncertainty, error in salt and graphite density correlations, are not treated here.

32. 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 the displacements calculation it was assumed that the interface between graphite reflector and vessel did not move, and 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.

33. "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. Small positive effect of fuel density on reactivity is increasing from +1.21 pcm/K for the reactor startup to +1.66 pcm/K for equilibrium fuel composition which has negative effect on FTC magnitude during the reactor operation.

34. "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.

35. 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} cm^2/s the calculated poison fraction is less than 2% [11]. 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.

36. 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: ¡++¿

37. "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 has been removed completely.

Reviewer 3

38. 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 [7] are most similar to the work presented in this paper. However, few major differences worse to be mentioned: (1) Park *et al.* employed MCNP6 for depletion simulations while SERPENT2 was used in this work (SERPENT2 seems to be much faster in massively parallel simulations involving more than 1000 cores); (2) full-core reactor model herein is more detailed and has no major approximation comparing with Park *et al.* (detailed full-core models comparison might be found elsewhere [12]); (3) Park *et al.* and Jeong *et al.* both considered volatile gases, noble metals removal and ^{233}Pa separation while in the current work we are implemented detailed reprocessing scheme described in conceptual MSBR design [11]; (4) ^{232}Th neutron capture reaction rate has been investigated to prove advantages of two-region core design; (5) the effect of removing fission product removal from fuel salt has been studied.

39. 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.

40. 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.

41. 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 21).

42. In Page 10, "SERPENT generates the problem-dependent nuclear data library", how did SERPENT 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.

43. In Page 14, "depletioncalculations" should be "depletion calculations".

Solution: Thanks for catching this, fixed.

44. 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.

45. 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).

46. 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.

47. 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.

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

Solution: ¡++¿

References

[1] Benjamin R. Betzler, J. J. Powers, N. R. Brown, and B. T. Rearden. Implementation of Molten Salt Reactor Tools in SCALE. In *Proc. M&C 2017 - International Conference on Mathematics & Computational Methods Applied to Nuclear Science and Engineering*, Jeju, Korea, April 2017.

[2] J. J. Powers, T. J. Harrison, and J. C. Gehin. A new approach for modeling and analysis of molten salt reactors using SCALE. Sun Valley, ID, USA, July 2013. American Nuclear Society, 555 North Kensington Avenue, La Grange Park, IL 60526 (United States).

[3] Jinsu Park, Yongjin Jeong, Hyun Chul Lee, and Deokjung Lee. Whole core analysis of molten salt breeder reactor with online fuel reprocessing. *International Journal of Energy Research*, 39(12):1673–1680, October 2015.

[4] Benjamin R. Betzler, Jeffrey J. Powers, and Andrew Worrall. Molten salt reactor neutronics and fuel cycle modeling and simulation with SCALE. *Annals of Nuclear Energy*, 101(Supplement C):489–503, March 2017.

[5] Benoit Forget, Kord Smith, Shikhar Kumar, Miriam Rathbun, and Jingang Liang. Integral Full Core Multi-Physics PWR Benchmark with Measured Data. Technical report, Massachusetts Institute of Technology, 2018.

[6] Andrei Rykhlevskii, Alexander Lindsay, and Kathryn D. Huff. Online reprocessing simulation for thorium-fueled molten salt breeder reactor. In *Transactions of the American Nuclear Society*, Washington, DC, United States, November 2017. American Nuclear Society.

[7] Yongjin Jeong, Jinsu Park, Hyun Chul Lee, and Deokjung Lee. Equilibrium core design methods for molten salt breeder reactor based on two-cell model. *Journal of Nuclear Science and Technology*, 53(4):529–536, April 2016.

[8] M. Aufiero, A. Cammi, C. Fiorina, J. Leppnen, L. Luzzi, and M. E. Ricotti. An extended version of the SERPENT-2 code to investigate fuel burn-up and core material evolution of the Molten Salt Fast Reactor. *Journal of Nuclear Materials*, 441(13):473–486, October 2013.

[9] O. Ashraf, A. D. Smirnov, and G. V. Tikhomirov. Nuclear fuel optimization for molten salt fast reactor. In *Journal of Physics: Conference Series*, volume 1133, page 012026. IOP Publishing, 2018.

[10] Carlo Fiorina, Manuele Aufiero, Antonio Cammi, Fausto Franceschini, Jiri Krepel, Lelio Luzzi, Konstantin Mikityuk, and Marco Enrico Ricotti. Investigation of the MSFR core physics and fuel cycle characteristics. *Progress in Nuclear Energy*, 68:153–168, September 2013.

[11] R. C. Robertson. Conceptual Design Study of a Single-Fluid Molten-Salt Breeder Reactor. Technical Report ORNL–4541, comp.; Oak Ridge National Lab., Tenn., January 1971.

[12] Andrei Rykhlevskii, Alexander Lindsay, and Kathryn D. Huff. Full-core analysis of thorium-fueled Molten Salt Breeder Reactor using the SERPENT 2 Monte Carlo code. In *Transactions of the American Nuclear Society*, Washington, DC, United States, November 2017. American Nuclear Society.