Article

A HELIOS Based Dynamic Salt Clean-Up Study Analysing the Effects of a Plutonium Based Initial Core for IMAGINE

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Abstract: Nuclear technologies have a strong potential and a unique role to play in delivering reliable low carbon energy for a future net-zero society. However, to assure the sustainability required for the long-term success, nuclear will need to deliver innovative solutions as proposed in iMAGINE. One of the most attractive features, but also a key challenge for the envisaged highly integrated nuclear energy system is the need for a demand driven salt clean-up system. The work described provides an insight into the interplay between a potential salt clean-up system and the reactor operation in a plutonium started core in a dynamic approach. The results presented will help to optimize the parameters for the salt clean-up process as well as to understand the differences which appear between a core started with enriched Uranium and Plutonium as the fissile material. The integrated model is used to investigate the effects of the initial fissile material on core size, achievable burnup, and long term operation. Different approaches are tested to achieve a higher burnup in the significantly smaller Pu driven core. The effects of different clean-up system throughputs on the concentration of fission products in the reactor salt and its consequences are discussed for general molten salt reactor design. Finally, an investigation of how a plutonium loaded core could be used to provide fuel for future reactors through fuel salt splitting is presented with the outcome that one Pu started reactor of the same size as a uranium started core could deliver fuel for 1.5 new cores due to enhanced breeding. The results provide an essential understanding for the progress of iMAGINE as well as the basis for inter-disciplinary work required for optimizing iMAGINE.

Keywords: nuclear; nuclear energy; nuclear reactors; reactor physics; modelling and simulation; molten salt reactors; nuclear chemistry; fission products; salt clean-up; plutonium management

Introduction

Nuclear energy offers great potential to support a sustainable energy future, since it is currently the only available net-zero technology which can deliver 24/7 availability and adjustability while also producing the required massive amounts of on-demand, low carbon energy to make a net zero society viable. However, for large scale future deployment, it is vital to improve the sustainability of the technology, thus the fuel utilization, see [1]. The key to achieve improved fuel utilization is closed fuel cycle operation as has already been demonstrated in fast reactor technology, but this will only be possible when the operation is based on Plutonium as fissile material [2]. Fast reactors have always been planned to operate in a closed fuel cycle mode based on mixed oxide (MOX) fuel. But for a long time, even the only industrially operated fast reactor, BN-600, operated on pure uranium-based fuel. The follow up system, BN-800, has also been started on uranium oxide fuel, but it is now gradually being converted to MOX fuel-based operation [3]. However, for closing the fuel cycle with solid fueled reactors, a complex and investment intensive fuel reprocessing process has to be installed to separate and access Plutonium, often

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called 'Plutonium Economy'. To avoid these challenges, we have proposed the iMAGINE concept – a highly innovative nuclear system which aims at using molten salt fast reactors in conjunction with an integrated salt clean-up system. The system is proposed to operate on spent nuclear fuel without prior reprocessing and has the potential to deliver waste management [4] and power production [5] in a single unified process, significantly more efficient than the classical closed fuel cycle approaches [6]. In contrast to the above-mentioned solid fuel-based approaches and the recent approaches for transmutation in ADS [8], this novel integrated approach offers the prospect of a single holistic solution, starting with turning the spent fuel into the salt phase like it is done in the pre-processing stage of pyro reprocessing. The iMAGINE concept has the potential to deliver an innovative, resource efficient energy production system as well as a highly sustainable waste management process [6, 7], creating a solution to the often criticised negative aspects of nuclear [1]. This novel approach, which could be called "reverse" reprocessing, is completely different from other proposed molten salt reactor systems [9,10] and has been developed for iMAGINE with the aim to keep all actinides in the salt to avoid proliferation issues, while separating only selected elements which prevent the reactor from long term operation. Following the study of the enriched uranium based iMAGINE proposal in "A HELIOS based dynamic salt clean-up study for iMAGINE" [11], this paper focusses on the Pu started core since for the UK, with its massive amount of already separated civil plutonium [12], this approach could be the most interesting as a potential new strategy for most efficient utilization of plutonium.

This work will address the investigation of clean-up requirements for the Pu started core and it's comparison to the Uranium core as basis for planning the future application of iMAGINE. Some general knowledge on this is already available through historic experience. Replacing the Uranium based core with a Pu started core will improve the availability of neutrons for breeding due to the increased number of neutrons per fission in Pu. However, no significant difference in long term operation is to be expected since the large difference in the material composition at the beginning will disappear rapidly. This is because the breeding process in a Uranium based fast reactor core will anyway turn the fissile material composition to Pu. Nevertheless, it is essential to understand the differences to give consolidated advice in future discussions.

Code, Model and Methods

The HELIOS code system version: HELIOS 2.03, with the internal 173 group library [13] has been used for the simulations. The code is a 2D spectral code with wide unstructured mesh capabilities, a transport solver based on the collision probability method [14] and a newer extension based on the Method of Characteristics [15]. The general model is based on the EVOL benchmark configuration [16] which has been transformed into a volume corrected 2D HELIOS model (see Figure 1). The model has been adopted to reproduce the 3D structure and the relations between the different materials as closely as possible. Furthermore, the benchmark model has been extended to additionally represent the outer structures and the 16 heat exchanger pipe arrangement for a better representation of the real geometry. The leakage in the third dimension is introduced into the calculation through the insertion of a buckling correction available in HELIOS (BSQ: 0.00002). This value has been fixed by a comparison of 2D and 3D calculations within the EVOL benchmark exercises [17]. Using this setting, a k_{inf} of ~1.005 is required to achieve a pseudo 3D k_{eff} of 1.0. Leakage in the radial direction is directly modelled through vacuum boundary conditions.

The salt system chosen as reference system for iMAGINE is based on NaCl-UCl_3-UCl_4 with the eutectic composition 42.5%–17.0%–40.5%. More detailed discussion on the properties of the salt system and the rationale behind its choice is given in [18]. The blanket area is filled with sodium, while the protector (the component used to shield the sensitive components like pumps and heat excahgers) is based on B_4C. The starting reference model has a core size of model 287.5 cm radius and a U-235 enrichment of 11.06%. This

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will be used for the comparison with the Pu started core for which the dimension and the fissile loading has to be determined using the Pu vector as given in Table 1.

Table 1: Plutonium vector used for the study taken form [Error! Reference source not found.].

Nuclide	Content (weight %)
Pu-238	0.25
Pu-239	68.77
Pu-240	26.7
Pu-241	1.76
Pu-242	2.52

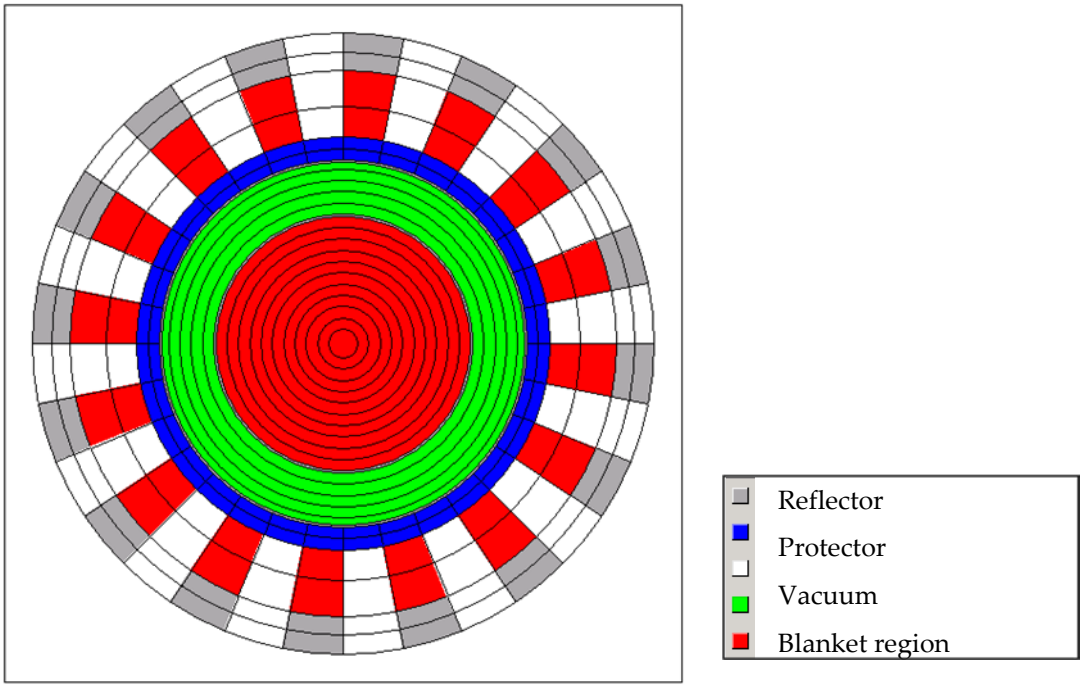

Figure 1: Volume corrected 2D HELIOS model of the molten salt reactor. .

The HELIOS code is an industrial standard software which is designed to perform the neutron transport and burn up calculations, and if requested the cross section preparation in the whole system or in defined calculation areas under the consideration of the boundary conditions set for the unit cell. Originally, HELIOS was developed to simulate solid structured fuel assemblies and, thus, the possibility of online refueling and reprocessing was not foreseen. To deal with these special molten salt reactor modelling requirements, a PYTHON script based on the special features of the HELIOS package has been developed [20]. All input data, which does not change during the whole reactor operation, is stored in a so-called expert input. The changing material configuration is fed into the system through a user input which is re-written in every cycle using a PYTHON script. Within each of these cycles, 5 burnup steps are calculated through HELIOS. The expert input and the updated user input are merged in the pre-processor AURORA [21], which creates the updated input for the HELIOS run used to determine the neutron flux distribution and material burnup. The results are finally evaluated at the end of each cycle in the post-processor ZENITH [22] where it is decided which elements are reduced or increased and to what extent, along with the isotopes to be fed back into the next user input which is created using the PYTHON script (see Figure 2). Theoretically, it would be possible to simulate a molten salt reactor precisely by using small time steps in this calculation loop. In a real MSR, however, two different time scales based on two different processes can be observed for the salt cleanup – the helium bubbling for gaseous and volatile fission

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products with a comparably short acting time, and the online salt cleanup for the dissolved fission products with a significantly longer acting time. To improve the modelling of both procedures, a new strategy has been developed based on the use of a reduced burnup per cycle (10 GWd/tHM using five burnup steps in HELIOS). The cycle length coincides with full removal of the gaseous and volatile fission products (the elements 18, 35, 36, 53, 54, 85) which are not carried forward through ZENITH. In contrast, the dissolved fission products can be removed within ZENITH based on a cleaning efficiency, providing the opportunity to set this efficiency elementwise for all considered elements.

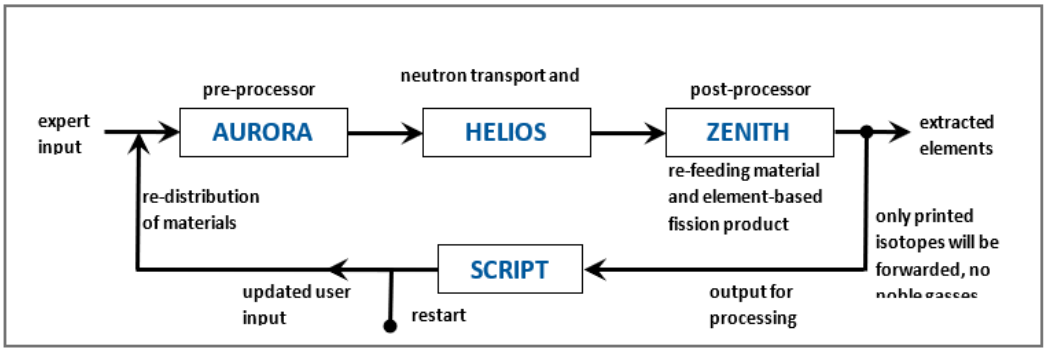

Figure 2: Description of the calculation cycle for the simulation of a MSR, based on the HELIOS package.

The use of the aforementioned process has already been validated and used in several peer-reviewed publications [20, 23, 24, 25]. However, the modelling and simulation quality will be significantly improved due to the new code version and the increased computational power which allows the use of the 173 energy group cross section set instead of the 47 group set used in the earlier publications.

However, due to the characteristics of HELIOS, some approximations still have to be accepted. The absence of fuel salt movement leads to an undesirable burnup distribution during each calculation cycle with the materials redistributed only on defining a new user input. HELIOS is an LWR code and an LWR spectrum is used for weighting the master libraries inside each energy group. However, this error will be significantly reduced compared to earlier publications, since the number of energy groups is tripled leading to a significant reduction in the width of each energy group. Comparisons with other codes in the EVOL benchmark [17], in a fast reactor isotope accumulation test against SERPENT [26], as well as comparisons with SCALE/POLARIS [18,27] have shown good agreement. This is what is currently available in terms of modelling techniques and solvers; therefore, to judge the reliability of the results, a real reactor physics experiment for molten salt reactors would be required as discussed in [27].

The approximations and the use of the HELIOS code package seem to be adequate for the approximation level required for this kind of long-term investigation of isotope accumulation to support the development of a clean-up system. The results of the influence of different elements on the system criticality has been evaluated against earlier publications [19,28].

The accumulation of isotopes is evaluated through a series of calculations of the system using the dynamic calculation scheme described above.

This is followed by a more sophisticated evaluation of the effect of the partial removal of different fission product element combinations on the potential increase in criticality compared to the reference case as well as the increase in the final burnup achievable through the removal of specific element combinations. The same approach is used to investigate the effect of initiating the fission product clean-up at different burnup and the effect of different separation efficiencies, representing different salt throughputs through the clean-up system. In the next step the fission product removal is applied in a staggered

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way, where the clean-up of one additional fission product is initiated every time the criticality of the test system approaches unity. For some of the cases, the fission product accumulation of specific elements will be analyzed in addition to get a deeper insight into the influence of the clean-up system on the fission product accumulation and a potential asymptotic limit of accumulation. The study will be finished with the investigation of the opportunity of controlling the system by splitting a share of fuel salt from the operational system and replacing it with pure fertile salt. This could be a promising approach for creating the fuel required for a next of a kind system which would help to avoid the transient system adaption at the beginning of the operation.

Results and Discussion

In the first step of comparison, the Pu content in the core is adopted to achieve the same starting criticality value for the core with 287.5 cm radius. The calculated Pu content is 10.4% with the same core dimension to achieve the same starting value for criticality, see Figure 3. The burnup curve shows that using the same reactor dimensions, the Pu loaded core provides substantially more efficient breeding than the Uranium based start-up core leading to an almost 4000 pcm higher peak criticality. Also, the achievable burnup before the clean-up would have to be initiated is roughly 50% higher. The original approach for the determination of the core size in the U-ref case was to keep the criticality between ± 500 pcm to allow using fuel temperature control as promised in [**Error! Reference source not found.**]. To satisfy these limits, obviously, the core size has to be significantly reduced. In addition, it has to be mentioned that the final burnup for a case with such high criticality will not be completely correct since a considerable number of neutrons will get lost due to normalization and will thus not be available for breeding. A rough estimation of the effect showed that the breeding is under estimated by ~2.3% due to the normalization. Based on this first outcome, the next step will be to determine a reactor core size which will fulfil the criterion of deviating within ± 500 pcm from criticality throughout the observed operational period.

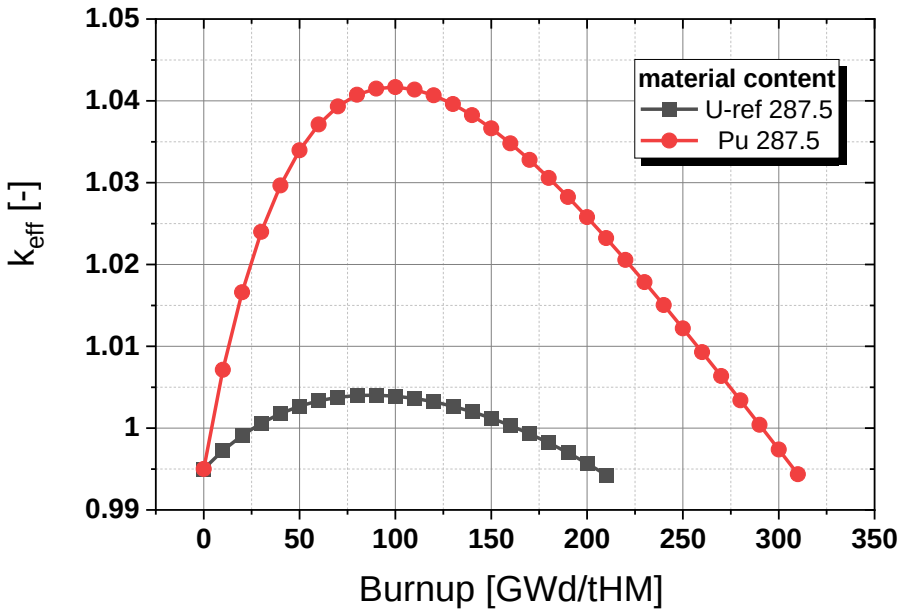

Figure 3. Evolution of criticality over burnup for a MSFR with clean-up of only gaseous and volatile fission products for identical reactor size operating on different fissile materials

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The considerably reduced core size, which allows to stay within the defined limit, is 187 cm in radius with 11.67% Pu content in the core and leads to a breeding curve delivering a comparable criticality change as in the U reference case but with a reduction in achievable burnup, see Figure 4. This is equivalent to a 34 reduction in diameter, or more importantly a 58% reduction in core volume and thus, in fuel salt required for the first core loading. The Plutonium balance indicates roughly a 52% reduction in the Pu demand for the start-up of the system. However, this comes with a drawback; the energy which can be extracted from the fuel before demanding the start of the salt clean-up system is 34% lower compared to the Uranium reference fuel due to the reduced achievable burnup (see Figure 4). The achievable burnup without initiation of the clean-up system is ~130 GWd/tHM compared to more than 200 GWd/tHM, a decrease by ~35%. Combined with the reduced salt mass or volume in the core the energy which can be harvested form the core is reduced to 35%. The major reason for the different behaviour of the Pu and U based cores is the change in the fissile material in the U based core which turns slowly from a U-235 driven core to a Pu-239 driven core. This change leads to a substantial increase of the number of neutrons per fission which enhances the breeding process considerably.

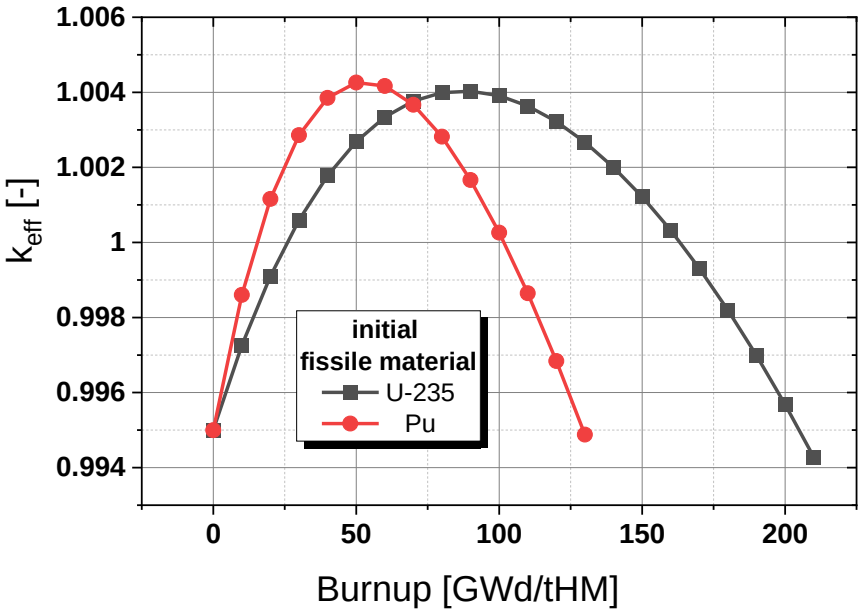

Figure 4: Evolution of criticality over burnup for a MSFR with clean-up of only gaseous and volatile fission products matching the criticality maximum when operating on different fissile materials

Adding different levels of salt clean-up (20% and 40%) to the operational schedule will help to extend the achievable burnup of the core, see Figure 5 and Figure 7. The clean-up leads to a significant increase in the maximum criticality from +426 pcm in the reference case to ~530 pcm after initiating the Ru clean-up, to slightly more than 700 pcm after the initiation of the Ru and Mo clean-up, to 1220 pcm if Pd is integrated to the clean-up and finally to ~1620 pcm when Ru, Mo, Pd, and Cs clean-up is initiated at 50 GWd/tHM.

The following increase in achievable burnup from the Pu reference case is observed with start of the clean-up system at a burnup of 50 GWd/tHM: ~40 GWd/tHM for only Ru, +~40 GWd/tHM integrating the clean-up of Mo, +~100 GWd/tHM integrating Pd to the clean-up, and +~100 GWd/tHM integrating Cs to the clean-up. The overall increase of the burnup when Ru, Mo, Pd, and Cs are integrated into the clean-up is from 130 GWd/tHM to almost 410 GWd/tHM. It gets obvious here that for the Pu started case, we would need

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to initiate already the clean-up of Ru and Mo to achieve a burnup comparable to the U-ref case.

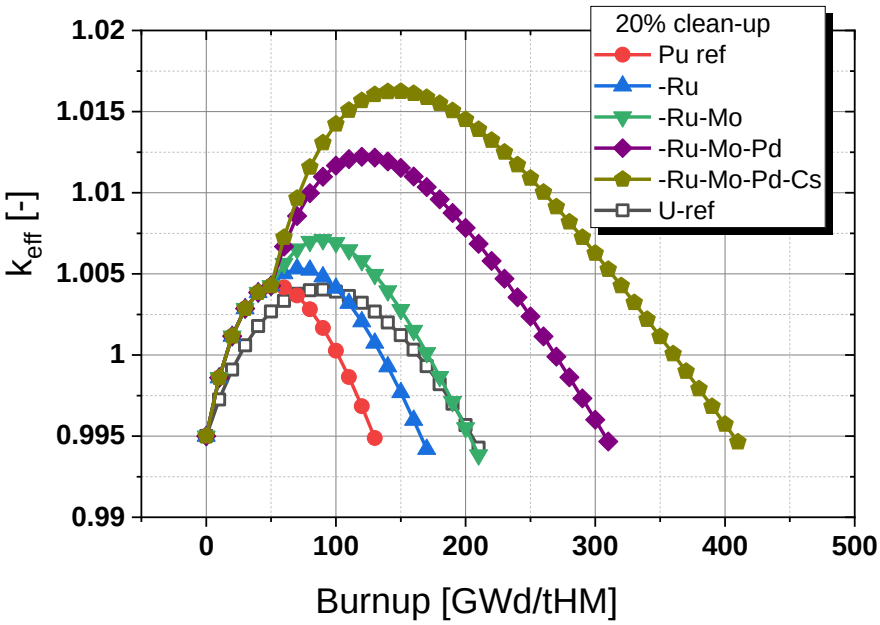

Figure 1: Evolution of criticality over burnup for a MSFR with clean-up of gaseous and volatile fission products and clean-up of different soluble fission products from the salt

In comparison to the uranium started case, Figure 6 shows that the achievable burnup for the Pu started case is not only significantly lower in the reference case, where the difference was 35%, but the effect becomes even more pronounced when considering the salt clean-up in the Uranium and Plutonium started case. Here the reduction is about 50%, which would become even higher if the k_{eff} normalization effect is considered since this normalization becomes more pronounced with higher maximum criticality. In general, the curves are comparable in their form, but the effect of the change in the fissile material in the Uranium started case leads to this significant difference in the achievable burnup which is still normalized on the fuel volume. Thus, the effect on the energy which can be extracted in a real reactor would have to be multiplied with the difference in core volume as given in the initial analysis.

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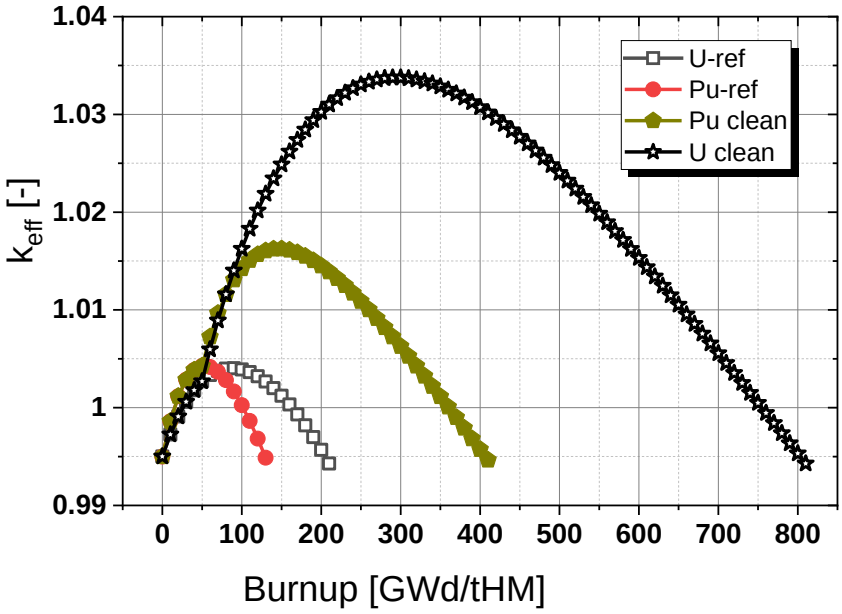

Figure 6. Evolution of criticality over burnup for a MSFR with clean-up of gaseous and volatile fission highlighting the difference between the Uranium and the Plutonium started case with and without salt clean-up

Next, the effect of doubling the clean-up rate to 40% is investigated, see Figure 7. The increased clean-up leads to a significant increase in the maximum criticality from +426 pcm in the reference case to ~690 pcm (vs 530 pcm for the standard 20% clean-up rate) after initiating Ru clean-up, to slightly more than 1000 pcm (vs 710 pcm) after the initiation of the Ru and Mo clean-up, to almost 1800 pcm (vs 1220 pcm) if Pd is integrated to the clean-up and finally to ~2300 pcm(vs 1624 pcm) when Ru, Mo, Pd, and Cs clean-up is initiated at 50 GWd/tHM. Doubling of the clean-up rate to 40% also helps to extend the achievable burnup of the core, see Figure 7. This is reflected in the higher burnup where the following increases are achieved: +~50 GWd/tHM (vs ~ 40 GWd/tHM) only Ru, +~50 GWd/tHM (vs ~40 GWd/tHM) integrating the clean-up of Mo, +~120 GWd/tHM (vs ~100 GWd/tHM) integrating Pd to the clean-up, and +130 GWd/tHM (vs ~100 GWd/tHM) integrating Cs to the clean-up. Thus, the overall increase in achievable burnup when Ru, Mo, Pd, and Cs are integrated into the clean-up is from 130 GWd/tHM to almost 480 GWd/tHM instead of 410 GWd/tHM. In other words, doubling the amount of the clean-up rate from 20% to 40% leads to less than 20% increase in the final achievable burnup, which shows that the effect is self-limiting.

A comparison of the average long term fission product concentrations which will develop as a result of the increased throughput of the clean-up system is shown in Figure 8. The main take away message from this figure is that the doubling of the throughput will be disadvantageous for the chemical engineering. The clean-up system would have to work on roughly half as large concentrations which will have a clear drawback on the efficiency of the chemical processes [**Error! Reference source not found.**]. Considering the associated cost of doubling the clean-up efforts and the increased challenges in the clean-up due to the lower concentrations of the elements to be separated, the effect is clearly not as strong as desired which raises the question regarding the optimal value.

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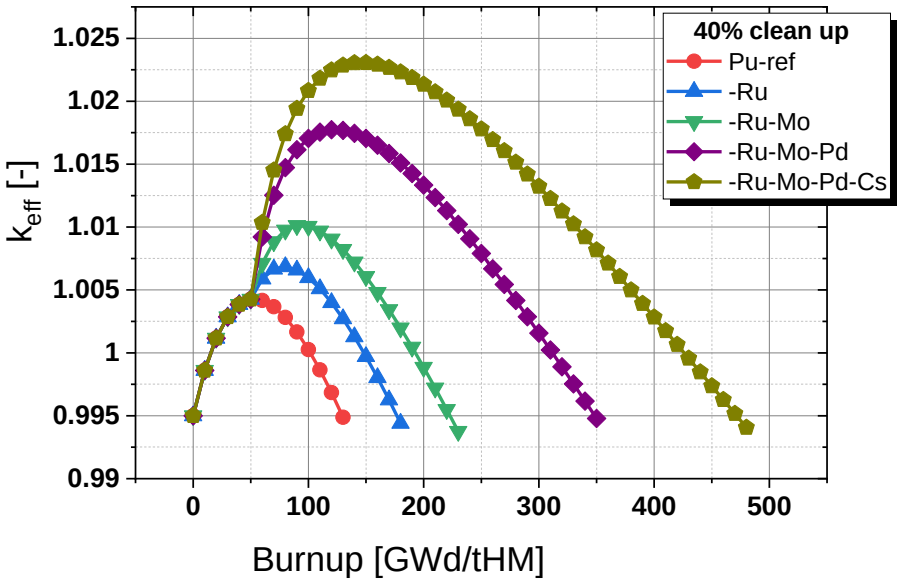

Figure 7: Evolution of criticality over burnup for a MSFR with clean-up of gaseous and volatile fission products with doubled clean-up system throughput

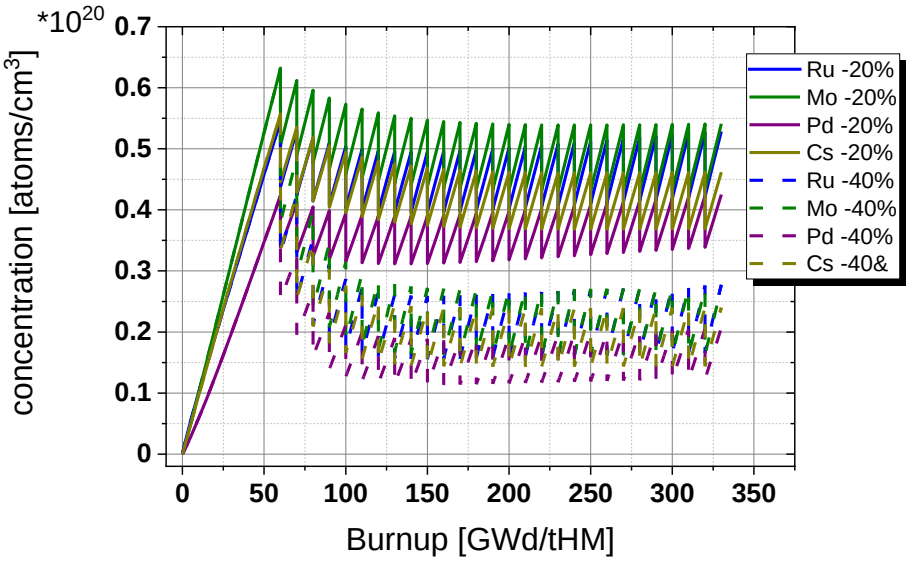

Figure 8: Evolution of fission product concentrations over burnup for a MSFR with 20% and 40% throughput of the clean-up system

From this discovery, some important outlook can be developed regarding different potential reactor concepts, their efficiency and their breeding potential. Large Uranium-Plutonium based systems like those investigated, are characterized by a comparatively high number of neutrons per fission >~2.7 due to the hard neutron spectrum and the high number of neutrons per fission produced in Pu-239. This offers a robust number of excess neutrons for breeding, while fast reactor is typically relatively robust against fission product poisoning due to the low absorption cross sections of the fission products in the fast spectrum. Two other important factors have to be considered in addition: a typical fast reactor core requires a significantly higher amount of fuel and the neutron leakage from

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the core is higher in a fast spectrum. From this we could have an outlook to thermal breeding which is typically delivered in the Thorium-Uranium cycle. U-233 releases ~2.3 neutrons per fission in the thermal breeder systems which is significantly lower than in the fast reactors and thus, leads to a lower breeding potential. In addition, the absorption cross section of several fission products is much higher in the thermal spectrum. The only advantage in the thermal core will be the reduced neutron leakage compared to fast reactor cores and thus, lowers the amount of fuel required in the core. Based on these physical and technological observations, it can be postulated that the demand on the salt clean-up will be significantly higher for a thermal core, due to the reduced breeding potential, the lower amount of fuel in the core, and the higher risk of fission product poisoning.

Coming back to the steps of the study, the detailed comparison between the Uranium started system with 20% clean-up and the Pu started system with doubled throughput, see Figure 9, indicates that the maximum of criticality of the Pu-based core is increased from ~1.016 to 1.023 (compare Figure 9 with Figure 6) by increasing the clean-up throughput which leads to a lower content of fission products, as shown in Figure 8. However, from this figure it becomes clear that doubling the throughput of the clean-up system leads to a limited gain of about 20% in the potential operational time. This is significantly lower than the potential operational time achievable in the Uranium started system due to the substantially larger system dimension and the resulting higher salt volume. This is due to the progressive change in fissile material composition of the Uranium started system from primarily U-235 to Pu which produces a higher number of neutrons per fission.

Looking onto the wider picture, some conclusions should be drawn here to complete the view. Taking the difference in the volume into account, the energy amount which can be produced in the Pu started core will be only about a quarter of the amount in the U started core. This needs some discussion. The Pu started core is more attractive based on the amount of initially invested fissile material, since less than 50% of the initial salt is required as compared to the Uranium case. However, considering the economics of the overall system, the system cost will not differ significantly since the Pu based core, although slightly smaller, would have a larger clean-up demand and finally in the long term operation both systems will be fed using spent fuel or tailings which are low in cost.

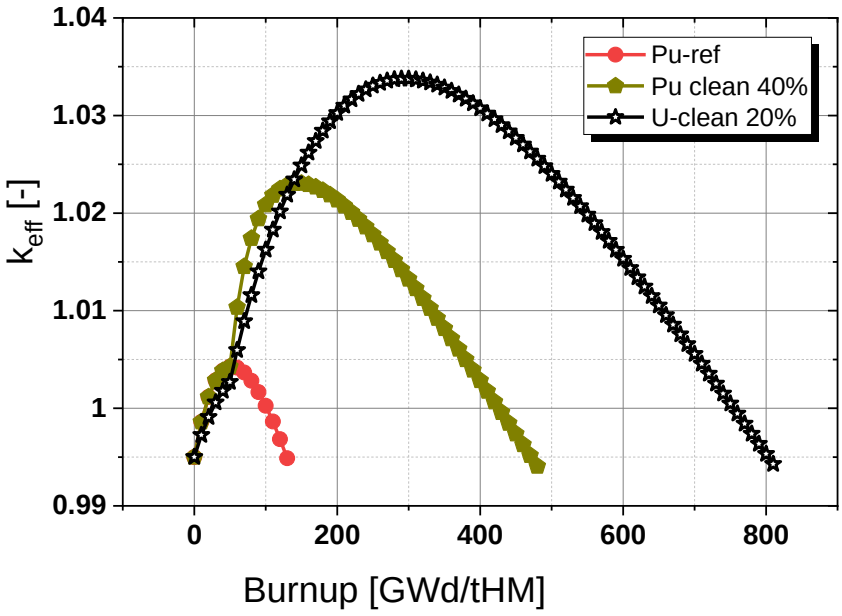

Figure 2: Evolution of criticality over burnup for a MSFR with clean-up of gaseous and volatile fission products highlighting the difference between the increased clean-up in the Pu stated system versus the Uranium started system

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Following the investigation of the increased throughput through the salt clean-up system which shows an insufficient increase of the core lifetime, the next investigation will be to look into the effect of adding a new clean-up step for the element Tc and the potential for achieving a higher burnup [11]. Also, the clean-up process with 20% throughput is now staged in a staggered manner such that, the clean-up for an additional element is initiated at the step prior to the system criticality going below unity, see Figure 10. The advantage of this approach is that the system achieves a criticality maximum of less than 1.005 leading to a more reliable result for the efficiency of the breeding process by minimizing the influence of k_{eff} normalization. A staggered initiation of additional elements in the clean-up process will allow a final burnup of almost 500 GWd/tHM and the additional clean-up step delivers a burnup increase of ~ 80 GWd/tHM. This approach shows that it would not be necessary to have the technology for clean-up of all the elements at the same time. Progressive addition of clean-up approaches for elements lower on the clean-up list delivered in [14] will allow to extend the core lifetime even if it is not initiated at an early stage of the core life. This opens promising opportunities for the long-term operation of the system with a stepwise increase in sophistication of the salt treatment technology.

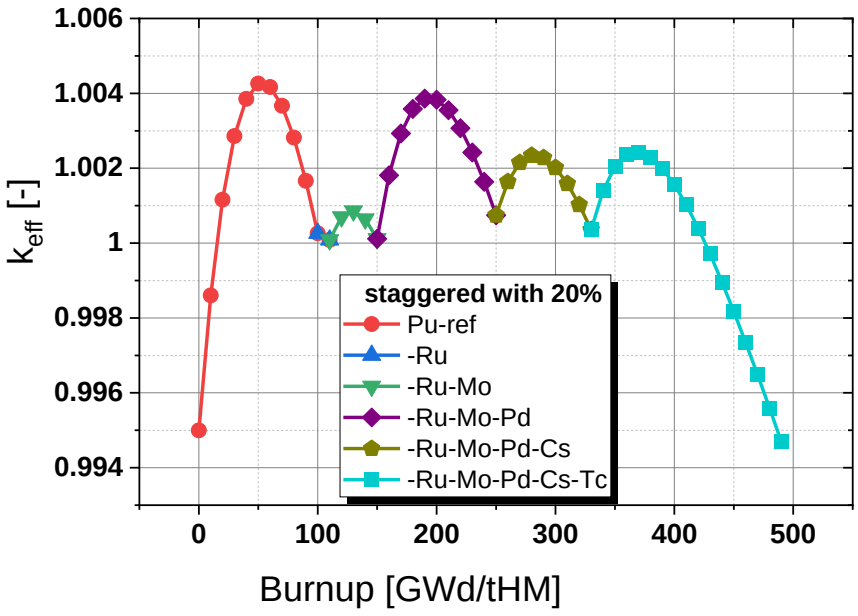

Figure 10. Evolution of criticality over burnup for a MSFR with clean-up of gaseous and volatile fission products applying a staggered introduction of the clean-up of different soluble fission products

However, comparing the Plutonium started system to the Uranium started system with staggered clean-up approach again shows that the Uranium started system will work for longer and will thus be more preferable, see Figure 11. This choice becomes even more evident when considering that not all countries will have access to separated Plutonium from operating a reprocessing facility like it is the case in the UK. These results further show that it is possible and more attractive to start iMAGINE with enriched Uranium based fuel and accept the increased system size required for this system.

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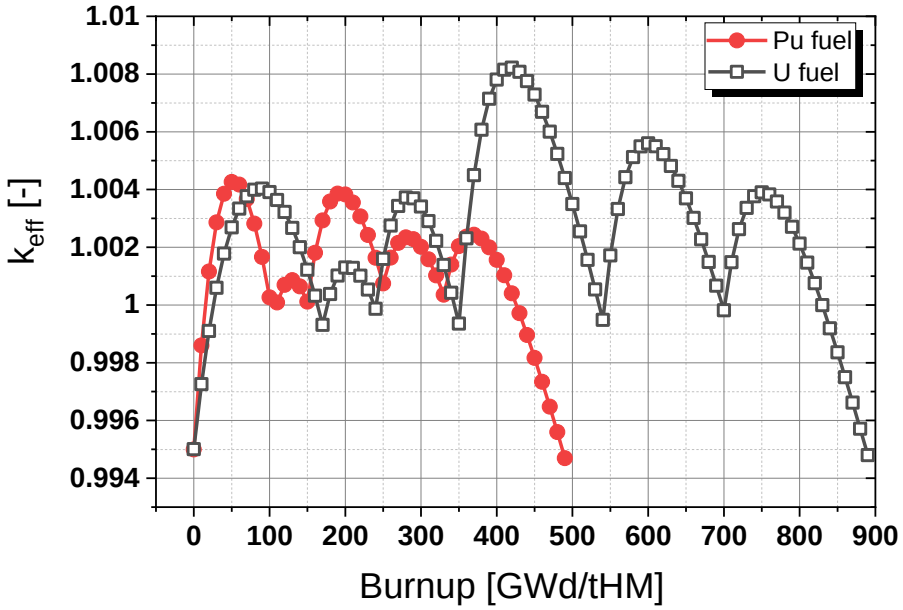

Figure 3: Evolution of criticality over burnup for a MSFR with clean-up of gaseous and volatile fission products comparing the effect of the Uranium and the Plutonium started core

Evaluating the approaches from a technical point of view, it might be questionable to develop a new system with different sizes in parallel; a smaller one for the Plutonium started system and a larger one for the Uranium started system. This guides us to the question; how to design a Plutonium started system with the required criticality control using the same size as determined for the Uranium started system. One potential approach could be to use salt splitting as the operational reactivity control mechanism where the salt amount split away from the system is replaced by UCl based fuel manufactured from depleted Uranium (tailings)- fertile salt. In this approach the split away salt could be used for feeding a next reactor in a chain avoiding the Uranium enrichment process. Figure 12 indicates the criticality curve for the 287.5 cm radius system filled with the required amount of Plutonium to achieve the initial starting condition of k_{eff} = 0.995. The system without any additional clean-up would rise to a maximum criticality of ~1.04, before the effect of the fission product poisoning reduces the criticality, leading to a final achievable burnup of ~370 GWd/tHM. Starting the clean-up system after 50 GWd/tHM and separating Ru, Mo, Pd, and Cs with a throughput of 20% after every 10 GWd/tHM burnup would lead to a maximum criticality of ~1.075 and a final burnup more than 1200 GWd/tHM. The salt splitting is applied by replacing a certain amount of the fuel by pure fertile salt, while the amount split away at each step is progressively reduced from initially 2% to zero in steps of 0.5%. The burnup periods with the amounts of salt (in %) split away from the core are given at the bottom of Figure 12. The red curve indicates the efficiency of the core reactivity control through this process, even if it gets clear that in a real reactor operation the steps would need to be smaller. However, the approach does not only help to control excess core reactivity, it additionally increases the final burnup from ~1200 GWd/tHM to ~1600 GWd/tHM, and the split away salt amount would be sufficient to reproduce ~ 1.5 times the core. Thus, the salt would be sufficient to start one additional reactor and could feed another to 50%. The achievable burnup increase can be explained here with two mechanisms: a) the inaccuracy of the breeding calculation due to the k_{eff} normalization (this can be estimated to ~4% here); b) the withdrawal of a significant amount of fission products due to the salt splitting while fresh, clean breeding material is provided to the core. In general, the process seems to be highly promising since the reactivity control, anyway needed, would create the basis for the supply of fuel for future reactors. Concluding

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from these results, and the discussion above Figure 9 it seems to be much more promising to rely on a larger core and use the salt splitting as a potential excess reactivity control instrument. This will, on the one hand, allow the clean-up to work with higher fission product concentrations while on the other hand, the improved breeding will help to provide new fuel as well as to keep the core critical for a substantially longer time. Thus, the core size may not be limited by maximum energy production but instead by the radioactive source term as a safety related parameter [18].

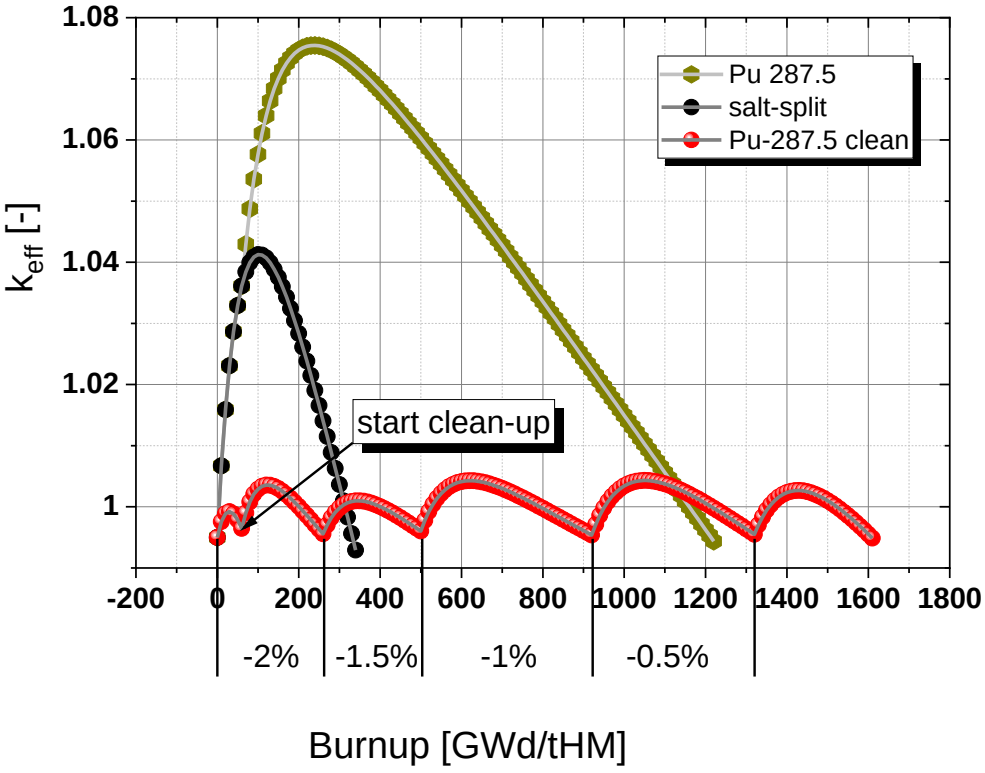

Figure 4: Evolution of criticality over burnup for a MSFR with clean-up of gaseous and volatile fission products investigating the opportunities of salt splitting as reactivity control approach

Conclusions

Nuclear energy has the potential to play a very unique role in a sustainable energy future, since it is the only currently available net-zero technology which can assure 24/7 availability and controllability, and can also deliver massive amounts of low carbon energy on demand to support a net zero society. However, to deliver on the expected large-scale demand, substantial innovation would be required in new reactor development, in improving the handling of the fuel within the fuel cycle, as well as in the waste management. The iMAGINE concept delivers exactly on these challenges [30]. However, the core challenges to deliver on are self-sustained breeding and the development of a strictly demand driven salt clean-up process based on reverse reprocessing. To develop and optimize this process a substantial input from reactor physics is required which has been delivered in [11]. The present work extends this insight by investigating a Plutonium started core in comparison with the previously investigated Uranium started core. The start-up of the reactor using Pu as a fissile material is an essential view for the UK due to the Pu stockpile and the demand for a positive management [31].

The results provided help to optimize the parameters for the salt clean-up process by creating a deeper understanding of the differences which appear between a core started

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with enriched Uranium and one started with Plutonium as fissile material. The integrated model is used to investigate the effects of the initial fissile material on core size, achievable burnup, and long term operation. Obviously, a Pu started core can be much smaller, but with the consequence of a much lower target burnup, before salt clean-up becomes necessary. Due to the significantly smaller fissile inventory the power delivered before initiation of the clean-up is significantly lower. Different approaches are tested to achieve a higher burnup in the considerably smaller Pu driven core. The effects of different throughputs through the clean-up system on the concentration of fission products in the reactor salt and its consequences are discussed for general molten salt reactor design. The evaluations are supported by a first estimate of the effect or error caused by the k_{eff} normalization for cases where higher criticality is achieved which, in a future molten salt reactor, could potentially be controlled with methods other than simply absorbing neutrons like in solid fuelled reactors. Finally, an analysis is presented how a larger plutonium loaded core could be used to provide fuel for future reactors through fuel salt splitting as a method for controlling the excess core reactivity. The outcome is that one Pu started reactor of the same size as a uranium started core could deliver critical molten salt fuel for 1.5 new cores through the enhanced breeding as a positive side effect of this criticality control approach.

In general, the work proves the requirement for dynamic simulation of the interaction between the clean-up system and the reactor operation as a basis for creating deeper understanding of the operational behavior of molten salt reactors. Only in such an interconnected modelling and simulation approach, it is possible to investigate the detailed effects of the clean-up on the reactor operation to get to an optimal solution. For the longer term, it would be essential to develop a toolset for coupled operational analysis including heat removal which would allow investigation of the role of feedback effects on the operation of a molten salt reactor at constant criticality and integrated salt clean-up, ideally with an automatic determination of the feeding streams.

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