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New Waste Management Options Created by iMAGINE through Direct Operation on Spent Nuclear Fuel Feed

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Abstract: The demand for improving the nuclear waste management has since long been identified as one of the major hurdles for widespread use of nuclear energy. Nuclear waste management, through partitioning and transmutation (P&T), has been researched since the 1990s with partitioning being a prerequisite for the process. Recently, an innovative approach of reactors directly operating on spent, or nowadays often called used nuclear fuel, iMAGINE has been proposed which could deliver on the aims of P&T as a side effect to more efficient and sustainable nuclear energy production in the future. A HELIOS model of the core has been used to analyze the long-term operation of a molten salt reactor including the investigation of the minor actinide accumulation over the entire burnup period. The results shown here confirm that long-term reactor operation is possible, even with higher amounts of vitrified waste loaded. Thus, it is possible to achieve the aims of P&T without prior partitioning, but it is certainly less efficient since the high concentration of minor actinides (MAs), required for efficient burning, is impossible to obtain in a short operational time. On this basis, the proposed nuclear waste management approach will be a long-term effort when it is accomplished without partitioning/separation technologies. However, none of the analyses contradicts this effort. The key points are: (a) when the technology for treating the waste is possible and reliable, the time horizon will not be a major concern; (b) the waste management is now intrinsically linked with energy production instead of requiring dedicated costly facilities, delivering a promising economic basis; (c) the waste management is now associated with long-term energy production and massively improved resource utilization. The study of feedback effects has shown that the modeled system has a strong negative feedback effect of ~−6 pcm/K, and even with spent nuclear fuel feed reduces to ~−3.8 pcm/K, ensuring the basis for a safe operation. In summary, it has been demonstrated that the objectives of P&T are achievable without prior partitioning, an approach which was never even discussed in the past. These ground-breaking results and the new insights will allow or even require rethinking the nuclear waste management of the future.

Keywords: nuclear; nuclear energy; nuclear waste management; partitioning and transmutation; reactor physics; modeling and simulation; molten salt reactors; salt cleanup; plutonium management; minor actinides

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1. Introduction

"One of the greatest challenges in the use of nuclear energy is the highly radioactive waste which is generated during power production. It must be dealt with safely and effectively. While technical solutions exist, including deep geological repositories, progress in the disposal of radioactive waste has been influenced, and in many cases delayed, by public perceptions about the safety of the technology. One of the primary reasons for this is the long life of many of the radioisotopes generated from fission, with half-lives of the

order of 100,000 to a million years. Problems of perception could be reduced to an essential degree if there were a way to burn or destroy the most toxic long-lived radioactive wastes during the production of energy." [1].

This excerpt taken from Victor Arkhipov, a consultant in the IAEA division of nuclear power and fuel cycle, highlights the importance of technological approaches for nuclear waste management, not only from a technical point of view, but also from the view of the public perception of nuclear technologies. This statement has not lost its validity although it was published in the *IAEA Bulletin* number 39 back in 1997. A very recent study [2] has again highlighted that an improved technical solution for waste management is what people hope for and expect to be delivered from the nuclear community to make nuclear energy more attractive, in addition to the fear around misuse of nuclear materials, and environmental pollution through releases or accidents [2]. Here, the new research and innovation work created through the iMAGINE approach [3,4] provides new, interesting opportunities for significantly improved solutions for a future final disposal [5]. The use of reverse reprocessing has the potential to significantly reduce the final disposal challenge due to element-wise separation of fission products and the related opportunities to either avoid their carryover in a mix of elements into the final disposal or the chance to use significantly more efficient conditioning processes. Both have the potential to reduce or at least slow down the radionuclide migration into the environment [5].

Partitioning and transmutation (P&T) is the waste management strategy which has been developed to answer this expectation. P&T attracted much attention in the research in the 1990s and the following decade. In Europe, it led to the large integrated research program EUROTRANS [6] and its follow-ups. These projects delivered the demonstrations for the technological steps required for transmutation of transuranics and were supported by research on partitioning in the programs NEWPART (FP-4), PARTNEW (FP-5), and EUROPART (FP-6) [7]. These projects successfully delivered the lab-scale demonstration of P&T technologies, but the follow-up challenge is now to establish P&T technologies on an industrial scale for successful delivery [8]. Currently, nuclear waste management research on an industrial scale is only being undertaken in Russia, mainly using its fast reactor fleet [9], and is largely based on the principles proposed through EUROTRANS. Another, significantly more far-reaching future approach, called iMAGINE [3,4], could be based on the operation of an innovative, fast molten salt reactor directly on SNF without prior reprocessing [10]. This leads to the first research question: "Are the aims and objectives of P&T achievable without prior partitioning"?

The second challenge after direct SNF-based operation is the integration of already reprocessed material into the waste management strategy. The re-dissolution of vitrified waste was previously discussed in the acatech study on P&T in Germany from 2012 to 2014 [11,12], but it was not followed up in detail due to the limited effect of only removing Am and Cm with the remaining material reconditioned in borosilicate glass. The situation has now changed due to the possibility of reverse reprocessing in the iMAGINE system, which allows new approaches of dealing with waste [5]. This leads to the second research question: "What could be done if reprocessing has already taken place and 'the soup' still contains minor actinides, either as liquid or already conditioned in borosilicate glass? Can this situation be improved?"

The problem will be tackled through long-term operational analysis based on the modeling approach and the staggered start-up of the cleanup, as developed and described in [13]. The calculations will be used to investigate and understand the effect of different feed streams with increased fission product content on reactor operation and criticality. Finally, the accumulation of transuranic elements inside the core will be analyzed.

2. Code, Model, and Methods

The codes and methods description has already been provided in several publications [13–15]. However, it is adapted to the specifics of this study and it is essential for a general understanding. The HELIOS code system HELIOS 2.03 with the internal

173 group library [16] was used for these simulations. HELIOS is a 2D spectral code with wide unstructured mesh capabilities and a transport solver based on the collision probability method [17] and the Method of Characteristics [18]. The general model is based on the EVOL benchmark configuration [19], which is transferred to a volume-corrected 2D HELIOS model (see Figure 1). The model has been adopted to reproduce the 3D structure and the relations between 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. Leakage in the third dimension is introduced into the calculation through the insertion of a buckling correction available in HELIOS (buckling, BSQ: 0.00002). The chosen buckling value has been fixed by a comparison of 2D HELIOS and 3D Monte-Carlo calculations within the EVOL benchmark exercises [20]. Using this BSQ setting, for a pseudo 3D k_{eff} of 1.0, an equivalent multiplication factor (k) of ~1.005 is required in the 2D system. The leakage in radial direction is directly modeled through vacuum boundary conditions.

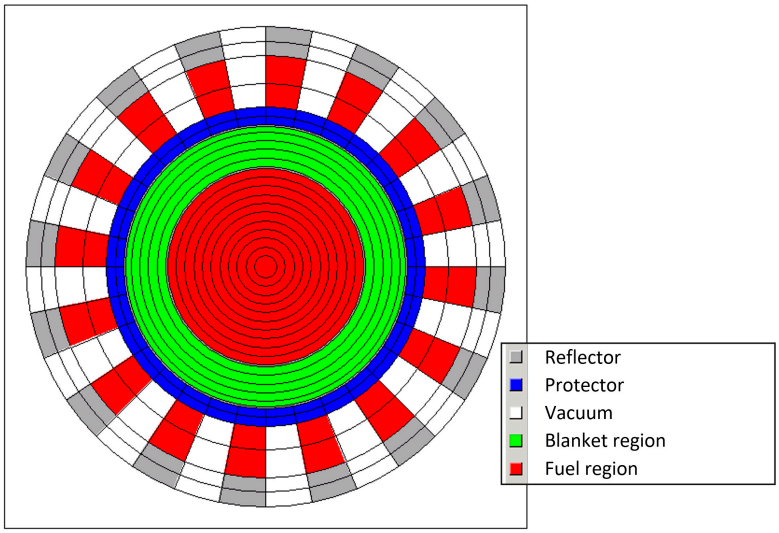


Figure 1. Volume-corrected 2D HELIOS model of the molten salt reactor.

The composition of the salt system chosen for iMAGINE is based on NaCl-UCl_3-UCl_4 with the eutectic formed at 42.5%–17.0%–40.5%. A detailed discussion on the data of the salt system and the rationale behind the choice is given in [21]. The blanket area is filled with sodium, while the protector is based on B_4C. The reference model has a core of radius 287.5 cm and a U-235 enrichment of 11.06%.

The composition for the SNF feed as well as for the additional feed of vitrified waste material is calculated, based on light water reactor fuel with an average burnup of 50 GWd/tHM (Giga-Watt days per ton of initial Heavy Metal), using HELIOS. The model is based on the 4.5% enriched fuel configuration of the NEA MOX benchmark [22].

The HELIOS code is an industrial standard software which is designed to perform the neutron transport calculations, the burnup calculations, and if requested, the cross-section preparation for core simulators. Originally, the HELIOS code was written for the simulation of solid structured fuel assemblies; thus, the possibility of online refueling and online reprocessing was not foreseen. To deal with these special features required for the simulation of molten salt reactor operation, a PYTHON script has been developed [14], which is based on the special features of the HELIOS package.

All input data, which do not change during the whole reactor operation, are stored in a so-called expert input. The changing material configuration is fed into the system through a user input which is rewritten in every cycle using a PYTHON script. Within each of the cycles, five burnup steps are calculated through HELIOS. The expert input and the updated user input are merged in the pre-processor AURORA [23], which creates the updated input

for the HELIOS run used for the determination of the neutron flux distribution and based on this the burnup of different materials. The results are finally evaluated at the end of each cycle in the post-processor ZENITH [24]. On the one hand, here it is decided which elements are reduced or increased and to what extent. On the other hand, new user input is also created in ZENITH using the information about the isotopes to be fed back into the next cycle. These are finally used to create the input for the next cycle with the help of 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.

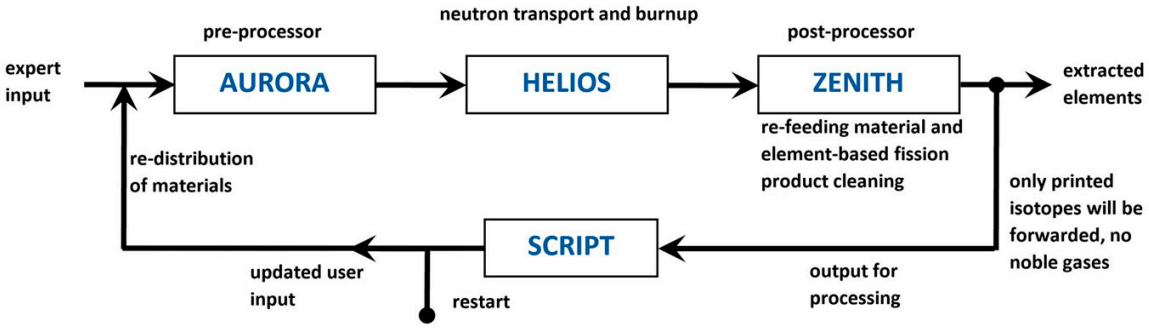


Figure 2. Description of the calculation cycle for the simulation of an MSR, based on the HELIOS package.

In a real MSR, two different time scales for the salt cleanup can be observed based on two different processes, the helium bubbling for gaseous and volatile fission 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 modeling of both procedures, a new strategy has been developed based on the use of a burnup of 10 GWd/tHM per cycle (using five burnup steps in HELIOS), coinciding with a full removal of gaseous and volatile fission products (the elements 18, 35, 36, 53, 54, 85 are not carried forward through ZENITH) after each initiation of a Python cycle. The dissolved fission products can be removed based on a variable cleaning efficiency, providing the opportunity to set this efficiency or the share of salt to be cleaned element-wise for all considered elements.

The use of the described process has already been validated and used in several peer-reviewed publications [14,15,25,26]. Meanwhile, the modeling and simulation quality will be significantly improved due to the new code version and the increased computational power, which allow the use of the 173 energy group cross-section set instead of the 47 group set in the earlier publications.

However, due to the characteristics of HELIOS, some approximations still have to be accepted. There is no fuel salt movement; thus, an undesired burnup distribution arises during each of the calculation cycles, while the materials are only redistributed when a new user input is defined. HELIOS is a light water reactor (LWR) code and an LWR spectrum is used for the weighting of the master libraries inside each energy group. However, this error will be significantly reduced compared to those in earlier publications, since the number of energy groups is tripled; thus, the width of each energy group has significantly reduced. Comparisons with other codes in the EVOL benchmark [20], in a fast reactor isotope accumulation test against SERPENT [27] as well as comparisons with SCALE/POLARIS [21,28], have shown good agreement. This is what is currently available in terms of modeling techniques and solvers; therefore, to verify the reliability of the results, a real reactor physics experiment for molten salt reactors would be required, as discussed in [28].

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 cleanup system. The results of the influence of different elements on the system criticality have been evaluated in earlier publications [29,30] and a detailed comparison to SCALE/Polaris is given in [13]. However, recent studies

have shown a significant difference between SCALE/Polaris and HELIOS in studies on the effects of Cl-37 enrichment [31].

This study investigated the use of the iMAGINE approach (see Figure 3) for a technology-based management of nuclear waste following the aims and objectives of P&T. For this, a long-term operation based on a staggered initiation of the cleanup system, as developed in [13], was used as the reference case and the evolution of transuranic elements Pu, Am, and Cm will be discussed for this reference case and for different scenarios which will lead to higher insertion of transuranic elements.

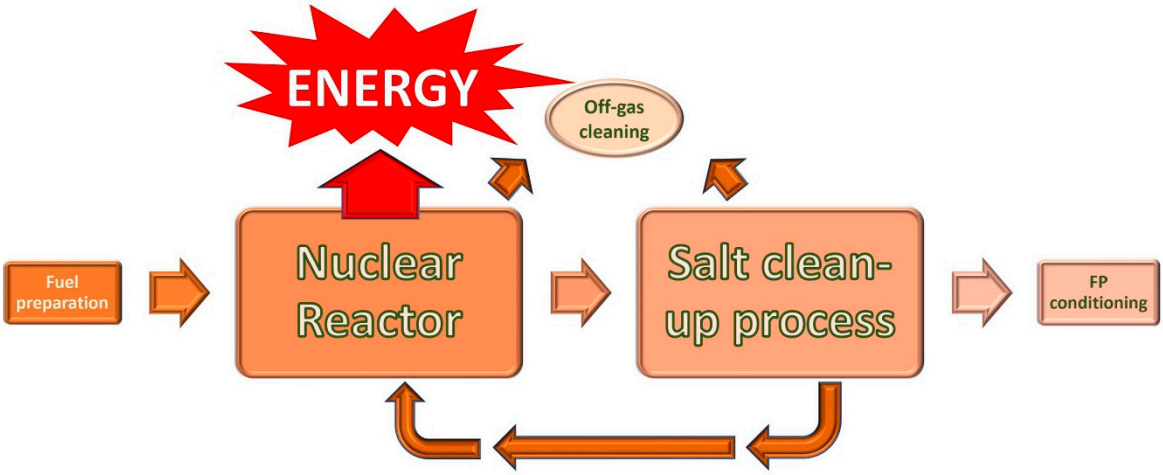


Figure 3. Schematic of the iMAGINE approach based on a molten-salt reactor with reverse reprocessing.

3. Results

3.1. Setup

As already mentioned, long-term operational analysis was based on the modeling approach and the staggered startup of the cleanup as developed and analyzed in [13]. The aim of the investigation was to provide an understanding of whether the iMAGINE approach would be able to fulfil the expectations of delivering a highly innovative waste management strategy without the demand for prior reprocessing. The two main points are: (a) the direct operation on SNF from light water reactors; and (b) use of the system to treat already vitrified wastes from reprocessing to burn the minor actinides and of the reverse reprocessing approach to allow improved handling of the fission products.

The basis for all investigations provided here is the analysis of different potential feed streams:

- Clean, depleted uranium tailings (tailings);
- Used/spent nuclear fuel without reprocessing (SNF);
- An increased amount of fission products that would be in the reactor system when the already separated fission products would be added to perform a kind of P&T operation on already vitrified leftovers of previous reprocessing approaches (SNF + FP).

These streams were fed into the reactor model after the start-up to understand the influence of the feed on the long-term evolution of the core. The simulations were used to gain an initial understanding of the effects of an increased fission product feeding into a reactor during operation by investigating core criticality and the concentrations of the elements to be transmuted, Pu, Am, and Cm, as well as Tc as a representative fission product where the accumulation and the effect of the salt cleanup can be observed. The results were used to draw conclusions on a potential optimization for a potential future operational scheme.

3.2. The Reference Case Study

The evolution of the system criticality over burnup is given in Figure 4 for the staggered initiation of the cleanup of different fission product elements, as shown at the bottom of the figure. The aim of the scheme was to balance the reactivity of the core for an extended period only through the cleanup initiation without relying on active control measures, like control rods, as well as to limit the effect of the k_{eff} normalization [13] on the results of breeding in a molten-salt system. The idea behind the staggered cleanup approach is to allow studies on the long-term behavior of a potential self-sustained iso-breeding molten salt reactor system with different feed streams, as described above (tailings, SNF, SNF + FP), and gain a deeper understanding of the effect of feeding very small material amounts over a long time on the long-term changes in composition and criticality.

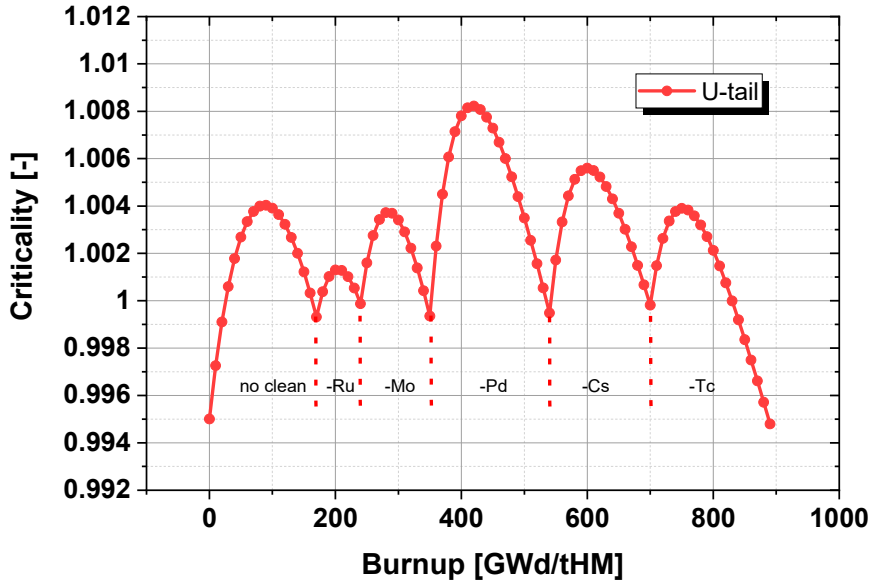


Figure 4. Evolution of criticality over burnup for the uranium started system with successive increase in the number of elements separated. A depleted uranium feeding stream is used to replace the U-238, which is consumed through breeding for the self-sustained long-term operation.

The feed stream with depleted uranium tailings was chosen as the reference case for this study (see Figure 4). It is one of the easiest envisaged operational modes in which U-238, already transmuted into fissile Pu-239, is replaced after every burnup cycle using the U-238 from clean tailings in the feed stream. The feeding stream is optimized to ensure a constant amount of U-238 within the core within a reasonable accuracy after each 10 GWd/tHm cycle. This is essential to balance the amount of breeding material available as basis for the iso-breeding. The cleanup of a certain additional element is activated every time when the system criticality falls below 1. In this case, 20% of the whole salt amount was cleaned after every 10 GWd/tHM cycle and the choice of the elements to be separated followed the priority list as previously developed [13]. Applying this staggered approach allows reactor operation until ~900 GWd/tHM is extracted, related to the initial load, compared to ~200 GWd/tHM without cleanup.

For a more detailed analysis of the changes of the isotopic contents of TRU materials, the contents in the core were analyzed and are shown in the following Figures 5–7. As already described, the initial core was based on enriched uranium. The initial U-235 content decreased rapidly due to reactor operation (see Figure 5) and was replaced by Pu-239 as the main fissile material at a burnup of ~75 GWd/tHM. The amount of U-235 continued to decrease beyond this point and became almost negligible at a burnup of ~500 GWd/tHM. At this time, the amount of Pu-239 in the system also attained a steady state with an equilibrium between breeding and burning being approached. It is important to highlight here that only a negligible amount of fissile material, besides the U-235 required

for reactor start-up, was inserted into the system. All fissile material was produced in-situ through internal breeding within the system. This process was supported by continuous replenishment of U-238 through the feed stream to maintain a constant amount of fertile material in the system. The creation of the higher Pu isotopes started with some time delay, since these isotopes are formed out of material which has to be bred first. At the end of the observation period of ~900 GWd/tHM, the Pu-240 production slowed down significantly along with the total Pu content, but an asymptotic value had not yet been achieved. The production of Pu-241 and Pu-242 was on a very small level. The final Pu vector was Pu-239, 67%; Pu-240, 28%; Pu-241, 3%; and Pu-242, 2% at 900 GWd/tHM.

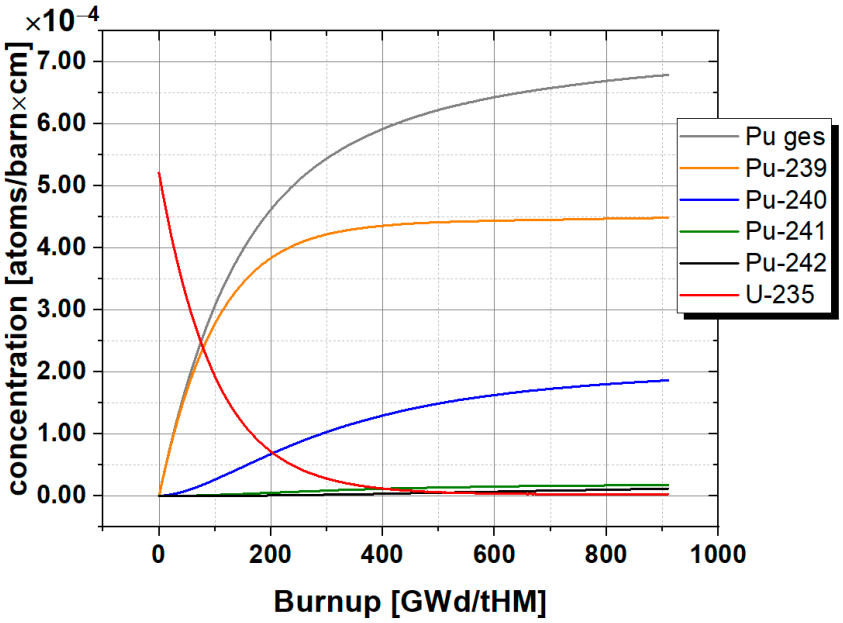


Figure 5. Investigation of Pu production and vector over fuel burnup for the case with tailings feed in the uranium started system.

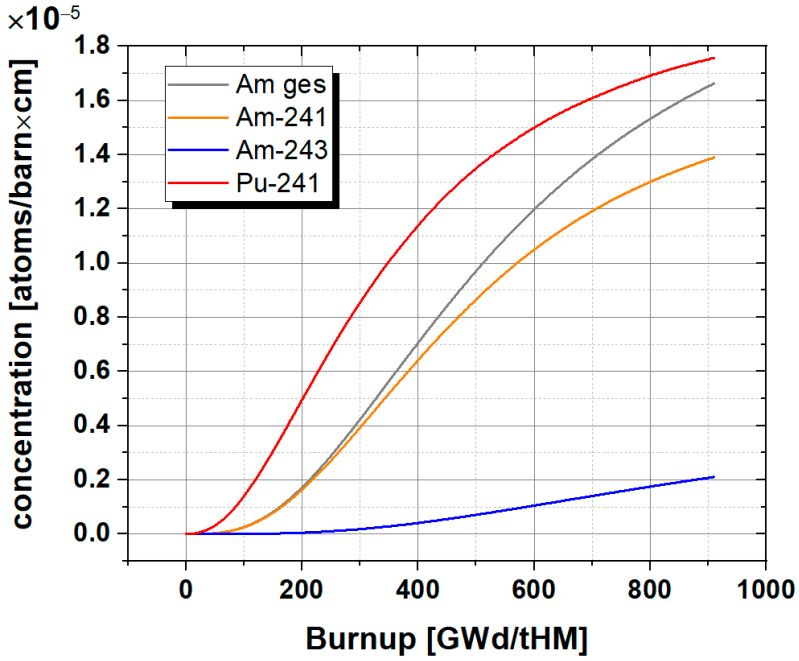


Figure 6. Investigation of Am production and vector over fuel burnup for the case with tailings feed in the uranium started system.

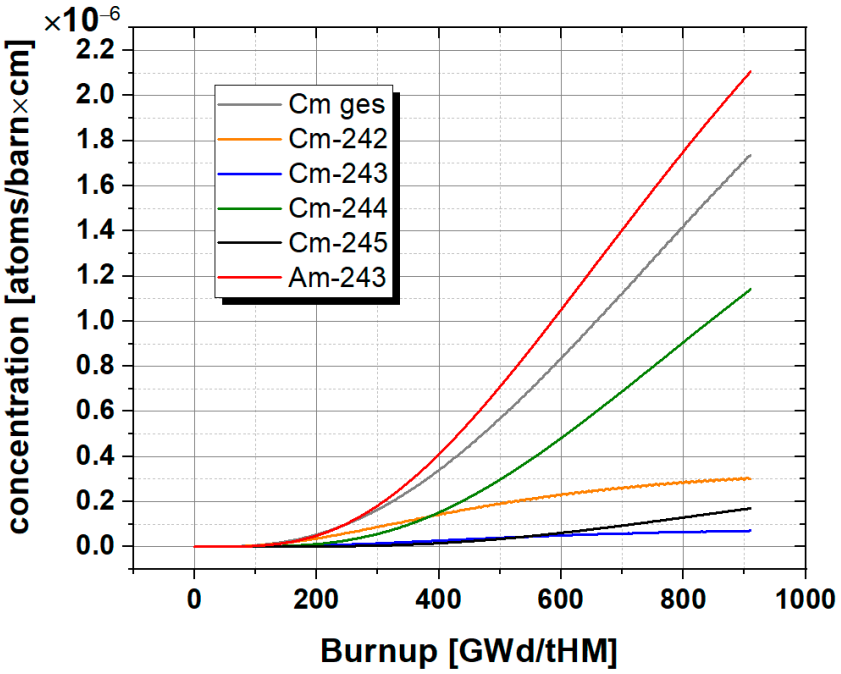


Figure 7. Investigation of Cm production and vector over fuel burnup for the case with tailings feed in the uranium started system.

The americium production (see Figure 6) followed a similar long-term behavior as the higher Pu build-up, but with even longer time delays since a substantial amount of the precursor isotope had to be bred first. The overall americium quantity at the end of the observation period was less than 3% of the total Pu content in the fuel salt. Am-241 is mainly formed though the decay of Pu-241; thus, the amount is slightly dependent on reactor power since there is the competition between Pu-241 fission and β-decay with a half-life of ~14 years. The delay in Am-241 production from the precursor Pu-241 is clearly observed in Figure 6. The small amount of Am-243 is formed mainly through the rapid beta decay of Pu-243 (half-life of 5 h), which is itself formed through neutron capture in Pu-242. Am-243 may also be formed due to neutron capture in Am-241 and Am242-m, but with a significantly lower contribution. Total Am production reaches a point of inflection at ~600 GWd/tHM, indicating that an asymptotic value will be achieved, although this would require a much longer operational time.

The curium formation in the core was 10 times lower than the americium formation (see Figure 7). The leading isotopes were Cm-244 and Cm-242. Cm-242 is mainly formed through neutron capture in Am-241, creating Am-242 (16 h half-life), followed by a beta decay to either Cm-242 or through electron capture to Pu-242 (marginal path). Cm-244 is mainly formed through neutron capture in Am-243 and the following decay of Am-244 with a small half-life of only 10 h. The delay in the formation of the leading isotope Cm-244 compared to the precursor isotope Am-243 was clearly observable. Cm-242 production had already reached a clear point of inflection at about 500 GWd/tHM and was almost asymptotic, while the Cm-244 still accumulated following the trend of Am-243. In general, the Cm-production had not reached a point of inflection during the observation period, mainly due to the still increasing amounts of Cm-244 and Cm-245. To observe the long-term behavior and to assess potential asymptotic behavior, a significantly longer observation period would be required.

However, an important fact is that due to the reverse reprocessing, all these TRU isotopes will stay in the core and will not require handling like in traditional P&T systems with solid fuel and external reprocessing. This new approach will help to reduce the radiation exposure to workers, unlike the multi recycling scheme required in the manufacturing of fuel for conventional solid-fueled systems [9].

3.3. Parametric Study of Different Fission Product Contents

The basis for the simulation was given by the case with SNF feed as calculated, based on light water reactor fuel with an average burnup of 50 GWd/tHM using HELIOS. The use of SNF (black line with squares) instead of tailings (red line with circles) as feed led to a slightly longer potential operation (see Figure 8). The system achieved higher criticality in the first operational period without salt cleanup. This effect is due to the higher share of fissile material in the SNF feed (~1% Pu and ~1% U-235) as compared to ~0.3% U-235 in tailings. Obviously, this observed difference was mainly important in the first operational stage without salt cleanup, since the following steps did not show major differences anymore. However, it is an interesting outcome that the effect of the fissile material added seems to be stronger than the effect of adding the fission products contained in the SNF. The main reason is that most FPs have significantly higher absorption XS in the thermal neutron spectrum as compared to the fast spectrum. Thus, the fission products have a much stronger poisoning effect in light water reactors than in the iMAGINE system.

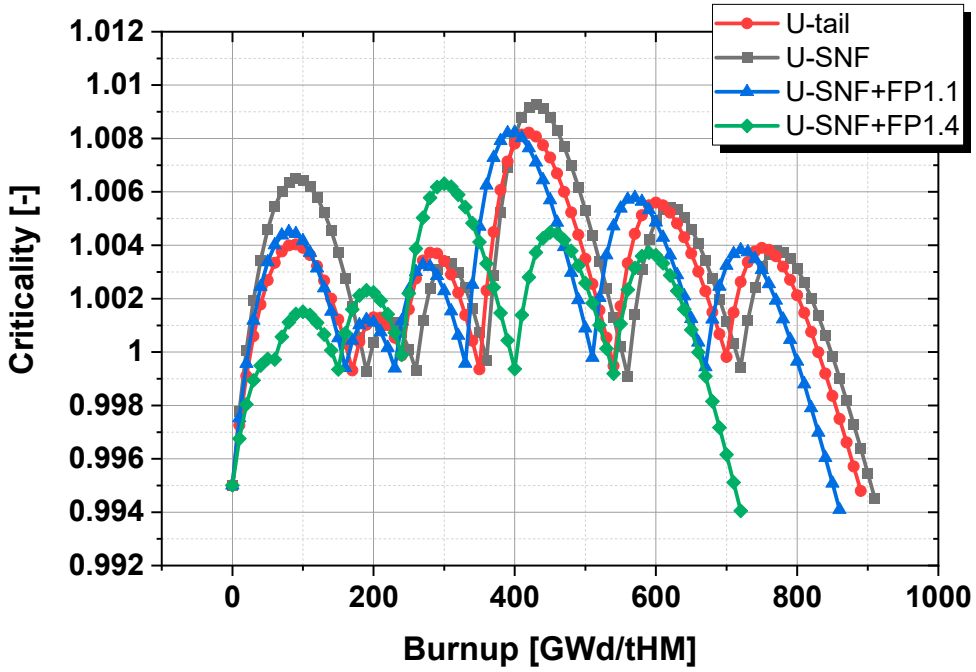


Figure 8. Uranium started system with successive increase in the number of elements separated and with different feeding streams used to replace the U-238, which is consumed through breeding for the self-sustained long-term operation.

Next, a higher amount of FPs was inserted into the system while keeping the amount of fertile and fissile materials (uranium and plutonium) in the feed stream unchanged. The idea was to model two feed streams with different amounts of vitrified waste by only increasing the amount of fission products in the SNF by two and five times. This modeling approach ensured conservatism by bringing all fission products and minor actinides into the core. In this way, it is not considered that gaseous and volatile fission products are only contained to a small share in the vitrified waste, while the major share has to be handled in a different way in the reprocessing plant. However, the effect is limited to the first calculation cycle since all gaseous and volatile fission products will anyway be released at the end of each 10 GWd/tHM cycle.

The insertion of double the amount of fission products (blue line with triangles) had a minor overall effect compared to the case using tailings. However, compared to the case of the SNF feed, it became clear that the increased amount of fission products had mainly a strong effect in the first stage before the cleanup was activated. The long-term effect was very limited. This can be explained by the overall increase of the amount of

fission products in the core as well as with the effect of the cleanup, which reduces not only the fission products created through the burnup, but also the fission products which are inserted in addition.

Inserting five portions of fission products (green lines with diamonds) had a very strong influence in the first operational period and did not even allow the reactor to become critical in the averaged criticality analysis per cycle. In general, the insertion of such a large amount of fission product significantly penalizes the operation, reducing the total burnup by ~20% from 910 GWd/tHM to 720 GWd/tHM compared to the SNF case. However, it clearly shows that the reactor could still be operated with such a high fission product load, which confirms that the insertion of larger amounts of fission products, e.g., through the feeding of dissolved vitrified waste, would be possible as a waste management method. On the one hand, this offers the opportunity to burn minor actinides which have not been separated previously in reprocessing; on the other hand, the approach will allow the use of the reverse reprocessing method with the opportunity of the element-wise separation of fission products, leading to new possibilities for conditioning of the waste streams.

The comparative analysis of the plutonium accumulation (Figure 9) shows that the Pu concentration in the core depended only weakly on the different feed scenarios. The Pu content in the scenario with the tailings feed was slightly lower, while feeding the largest amount of fission products delivered a marginally higher Pu content. However, there is no real effect on the overall tendency of the increase in the Pu amount and the formation of an almost asymptotic concentration at the end of the observation period. The amount of Pu fed (dashed lines) was identical in all SNF cases, which verifies the correct implementation of the model for the fission product feed.

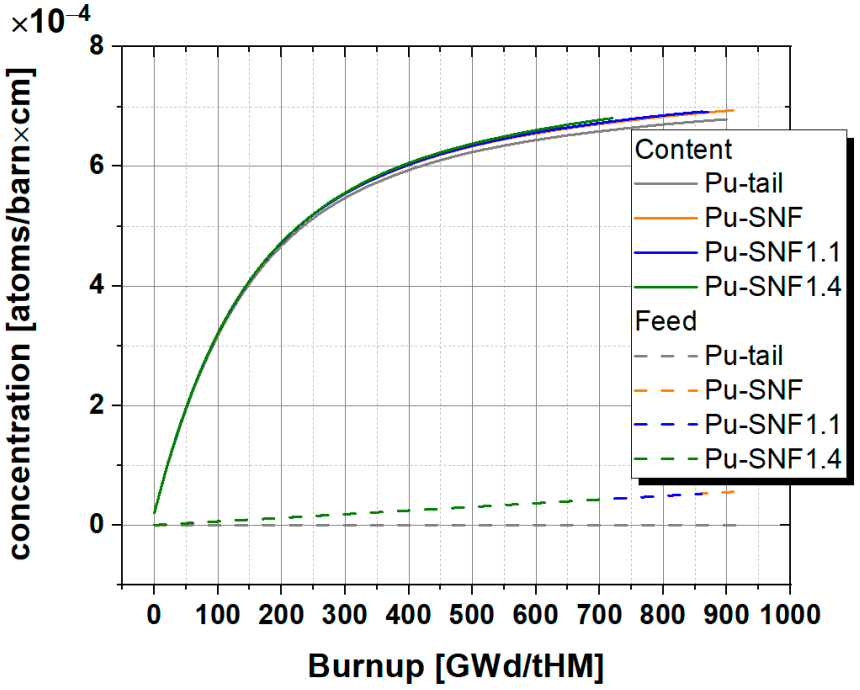


Figure 9. Comparison of the Pu production and the plutonium feed over burnup, dependent on the feed for the uranium started system.

The analysis of the Pu management showed that the amount resident in the system was in all cases slightly higher than in the reference case. However, in all SNF feeding cases the overall Pu amount was slightly reduced compared to the reference case with the tailings feed. The inserted Pu amount consisted of the Pu fed into the system and the Pu bred in the system. The Pu reduction was calculated using Equation (1). Obviously, the difference between the slightly higher Pu concentration at the end of the observation period is lower than the amount of Pu fed into the system through the SNF feed. The results indicate that

some small amount of Pu burning was achieved in the SNF fed scenarios (see Table 1). The different amounts of fission products had only a very marginal effect.

$$Element\ reduction\ (case) = 1 - \frac{amount\ resident\ (case) - amount\ fed\ (case)}{amount\ resident\ (Pu - tail)} \quad (1)$$

Table 1. Detailed analysis of the Pu management based on feeding and the amount resident at the end of the observation period.

	Pu-Tail [atoms/barn × cm]	Pu-SNF [atoms/barn × cm]	Pu-SNF1.1 [atoms/barn × cm]	Pu-SNF1.4 [atoms/barn × cm]
Amount fed	0	5.56×10^{-5}	5.26×10^{-5}	4.40×10^{-5}
Amount resident	6.79×10^{-4}	6.94×10^{-4}	6.90×10^{-4}	6.81×10^{-4}
Pu reduction		−6%	−6%	−6%

The comparative analysis of the americium content in the core (Figure 10) shows that the general tendency of the accumulation of Am was only slightly influenced by the feed. This indicates that the Am concentration in the core was mainly driven by the operation of the reactor and the breeding processes. Increasing the Am amount in the core through the feed led to a slightly higher concentration, but the effect was smaller than the increase due to operation. The Am feed is indicated through the dashed lines. It is already apparent from these lines that the inserted amount of Am was larger than the difference in the concentration appearing in the core over the observation period.

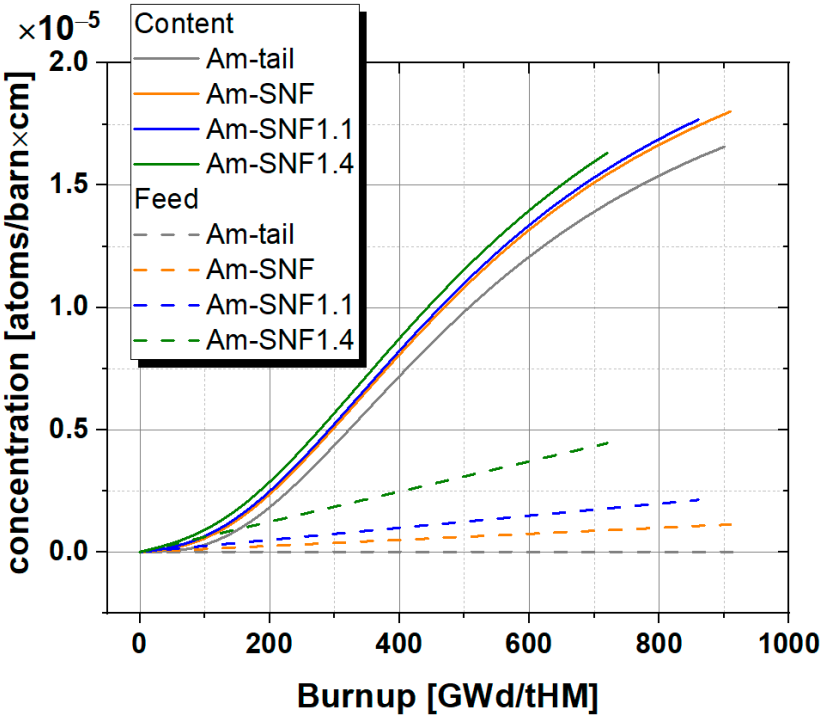


Figure 10. Comparison of the Am production and the Am feed over burnup, dependent on the feed for the uranium started system.

A more detailed analysis of the Am management (see Table 2) confirms this observation for the cases with higher fission product feed, but this was not the case for the unmodified SNF feed. The amount resident at the end of the observation period depended on the operational time and the amount of Am fed into the system, leading to the lowest resident amount for the SNF 1.4 case. Figure 10 supports here the understanding that the lowest amount was obviously a result of the shorter observation period, while the concentration was highest through the whole period. The overall Am change was calculated using

Equation (1). The higher Am content was because the increased Pu content in the SNF feed case led to a higher breeding of Am, since the SNF contained greater amounts of higher Pu isotopes as precursor, especially in the beginning, since a fully developed LWR Pu vector at 50 GWd/tHM was inserted, while the clean configuration based on enriched uranium was initially Pu free. In the tailings-based system, the breeding will start at a lower average mass of the Pu isotopes (see the delay in the Am concentration for the tailings fed case). The breeding process creates only Pu-239 in the first time period of operation; the formation of higher Pu isotopes requires a longer irradiation time (see Figure 5). In the cases of higher loads of Am through the feeding stream, Am reduction takes place and the reduction efficiency increases with increased Am concentration, which coincides with the observations that have been made in a large number of P&T studies delivered in the past [32–34].

Table 2. Detailed analysis of the Am management based on feeding and the amount resident at the end of the observation period.

	Am-Tail [atoms/barn × cm]	**Am-SNF [atoms/barn × cm]**	**Am-SNF1.1 [atoms/barn × cm]**	**Am-SNF1.4 [atoms/barn × cm]**
Amount fed	0	1.12×10^{-6}	2.13×10^{-6}	4.45×10^{-6}
Amount resident	1.66×10^{-5}	1.80×10^{-5}	1.77×10^{-5}	1.63×10^{-5}
Am change		2%	−6%	−28%

The Cm content over the observation period rose for all cases (see Figure 11). No saturation level was achieved and the concentration of Cm in the salt was still growing almost linearly at the end of the observation period. However, there was a clear difference between the base case where no Cm was fed into the core and all other cases. In all cases, Cm was bred inside the core from neutron capture reactions in Am: this means the production was strongly dependent on the Am content in the core and thus delayed in a clean core where the Am had to be bred first. However, the delay in the increase of the Cm concentration was much more pronounced than for Am (compare Figures 10 and 11). It is obvious in the other cases that the rapid increase in the Cm concentration at the beginning was caused by the feed through the SNF and additionally through the 'vitrified' waste. This is shown by the identical gradient of the concentration (solid lines) and the content inserted through the feed (dashed lines) up to 100 GWd/tHM. The massive insertion of Cm in the SNF 1.4 scenario and the almost identical slope of the feed and the content indicates that efficient burning takes place when the concentration is high enough.

A more detailed analysis of the Cm management and the concentrations as well as the feed is given in Table 3. The overall change in Cm was calculated using Equation (1). The concentration of Cm in the system (amount resident) at the end of the observation period increased by 27%, 28%, and 31%, respectively, for the scenarios with increased Cm feed. Obviously, the feeding amount was more influential in the case of Cm on the amount resident than the breeding process. However, the higher the feed, the greater was the amount of Cm which was transmuted during the operation of the reactor. In the scenario with the highest Cm feed, almost all material added through the feeding process was already burnt at the end of the observation period, which indicates a successful transmutation process, even if the concentration was still increasing. The cases with the lower feed support some transmutation, but the process was not yet efficient enough due to the low amount of Cm added.

In the final step, the effect of the different scenarios on fission product accumulation and salt cleanup was evaluated using technetium as the representative fission product (see Figure 12). The initial analysis indicates that the amount of all fission products inserted through the feeding stream was small compared to the accumulation of Tc, which is created due to reactor operation. One of the main reasons was that the LWR fuel, which was inserted through the feed stream, had a limited burnup of ~50GWd/tHM, much lower than the overall burnup in the fuel over the observation period. A more important observation

for the long-term operation is that the asymptotic value of Tc after activation of the salt cleanup seemed to be only marginally influenced by the feed. This confirms the expectation that the effect of the higher amount of fission products in the feed stream will be almost completely balanced through the cleanup system. This effect is interesting since it could lead to the conclusion that it would be promising to start with a comparably high fission product load while initiating the cleanup system earlier and using the fission products as a removable poison. These could be siphoned off when the reactor physics demands the removal of a specific fission product to reduce the number of different fission products to be tackled in the cleanup system in early operation stage.

Table 3. Detailed analysis of the Cm management based on feeding and the amount resident at the end of the observation period.

	Cm-Tail [atoms/barn × cm]	Cm-SNF [atoms/barn × cm]	Cm-SNF1.1 [atoms/barn × cm]	Cm-SNF1.4 [atoms/barn × cm]
Amount fed	0	5.42×10^{-7}	1.02×10^{-6}	2.14×10^{-6}
Amount resident	1.72×10^{-6}	2.18×10^{-6}	2.20×10^{-6}	2.26×10^{-6}
Cm reduction		−5%	−32%	−93%

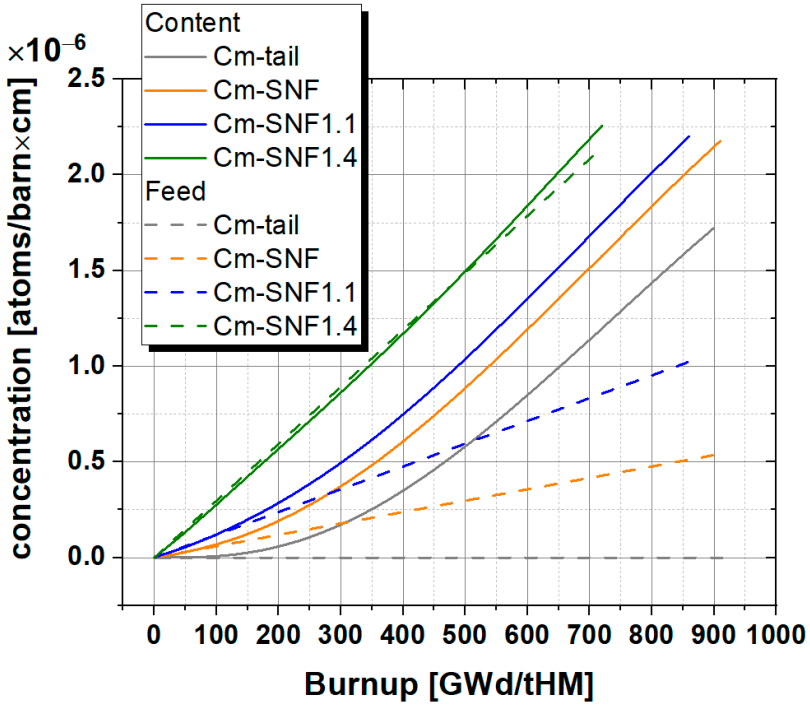


Figure 11. Comparison of the Cm production and the Cm feed over burnup, dependent on the feed for the uranium started system.

Finally, an investigation of the feedback effects was performed to ensure that all studied systems can be operated in a safe manner (see Table 4). The feedback effects were calculated in HELIOS for a fuel temperature and density change of +100 K, based on the model used for the operational study. It was approximated that the density change of the fuel salt was independent of the feeding stream. This seemed to be adequate since the largest part of each feeding stream is anyway based on U-238. The thermal feedback effect of the fuel was significantly stronger than in sodium-cooled fast reactors, where the doppler effect has been determined as −0.14 pcm/K in [35]. The main reason is that in solid-fueled fast reactors, all fuels stay within the core and thus the density change does not play a major role. In contrast to this, in a molten salt reactor with an adequate design, the change of the fuel temperature leads to a much stronger density change and this change leads to a loss of fissile material from the core, a combination leading to a much stronger feedback effect.

Additionally, unity of the coolant and fuel in a molten salt reactor leads to hardening of the neutron spectrum and thus, higher absorption probability. The feedback at the beginning of operation/life (BOL) was ~ −6 pcm/K for all cases (since only the feed stream was changed). In the tailings case, the feedback effect stayed almost constant and at the end of operation/life (EOL) was −5.5 pcm/K. In all cases with the SNF-based feed stream, the feedback decreased, which was primarily due to the change in the Pu vector, as higher Pu isotopes are known to reduce the feedback effect. In general, the feedback effect study demonstrated that even with high loading of fission products, the major prerequisite for safe reactor operation, inherent negative feedback effects are ensured within the expected quality of the simulations.

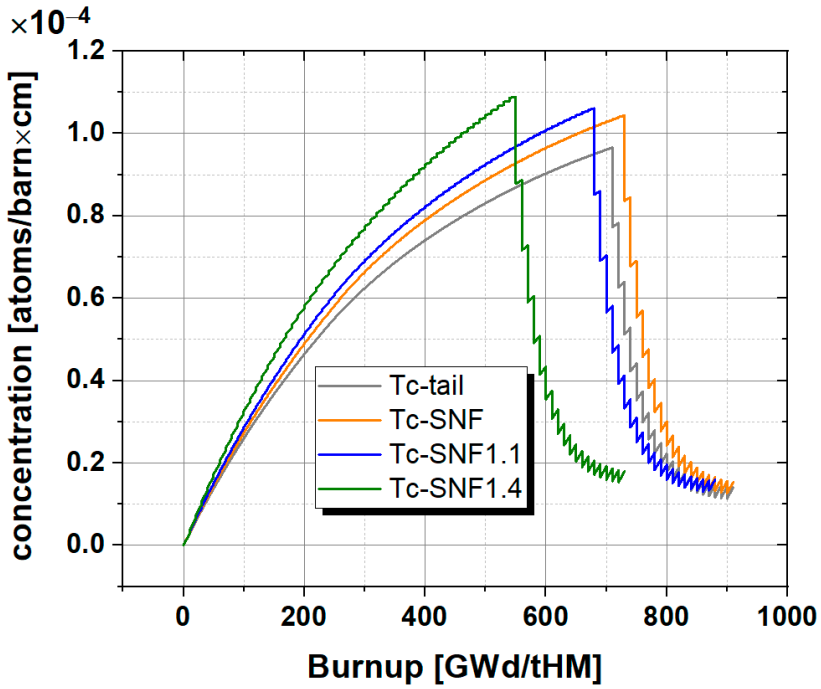


Figure 12. Evolution of the fission product content in the different scenarios using Tc as the representative element.

Table 4. Detailed analysis of the temperature feedback effect at the beginning and the end of the observation period.

	Feedback [pcm/K]
BOL	−6.0
EOL-tail	−5.8
EOL-SNF	−3.7
EOL-SNF 1.1	−3.8
EOL-SNF 1.4	−3.8

The flow of the molten salt fuel outside of the core through the heat exchangers will reduce the amount of delayed neutrons in the core, which is an often discussed phenomena that people tend to relate to reduced safety of the system. However, the main effect of the delayed neutron fraction is limiting the speed of the excursion due to the reactivity insertion. In modern molten salt reactor design with internal breeding, there is no requirement for excess reactivity, which is in contrast to classical solid fueled reactors. Thus, the risk of excursions is significantly limited. Anyway, it would be worth a study of this topic using realistic reactivity insertion speed related to re-feeding of cleaned material to create a deeper understanding of the limits.

4. Conclusions

Nuclear waste management, through partitioning and transmutation, has been researched since the 1990s. Currently, the general agreement is that partitioning of minor actinides is a prerequisite for the process. The high level of innovation delivered through iMAGINE is demonstrated in this paper through long-term operation modeling and simulation, which confirmed that it is possible to achieve the aims of P&T without prior partitioning. Although it seems less efficient, this is only due to the small fraction of MA in the LWR fuel which makes the process inefficient. The high concentration of MAs required for efficient burning are impossible to obtain in a short time without separation technologies. The amounts of MA inserted mainly stay in the system until their concentrations are high enough to reach a steady state. These high concentration levels were not achieved in this study, which shows that this proposed nuclear waste management approach will be a long-term effort when it is accomplished without partitioning/separation technologies. However, none of the analyses contradicts this effort. The key points are: (a) when the technology for treating the waste is possible and reliable, the time horizon will not be a major concern; (b) the waste management is now linked to energy production instead of requiring dedicated costly facilities, thus creating opportunities to deliver waste management with a promising economic basis; (c) the waste management is now associated with massive and long-term energy production by extracting all energy content of the mined uranium, thus unlocking a precious, practically inexhaustible energy resource through smart technology development. In comparison to classical fast reactors based closed fuel cycle, the iMAGINE system has the following advantages: (a) waiting times of ~10 years until the fuel can be reprocessed are no longer required; (b) the amount of fuel in the fuel cycle is not dominated by the long waiting time; (c) no fissile material has to be separated, avoiding proliferation concerns; (d) solid fuel with MA content does not have to be produced, which means there will be no heavy radiation exposure to humans in the fuel production; (e) MSFRs do not need excess reactivity, which is one of the main accident initiators in solid-fueled fast reactors.

From a reactor physics point, it has been demonstrated that the reactor can be kept critical even with the insertion of massive amounts of already separated FPs—either vitrified or in the so-called "soup" in countries with active reprocessing programs. Long-term self-sustained reactor operation has been demonstrated through modeling and simulation for a system started with enriched uranium for all investigated cases. After the initial start-up based on enriched uranium, the system is sufficiently robust to operate on an SNF feed or even a feed stream with additional FPs from already reprocessed waste. Analysis of the feedback effects has shown that the modeled system has a strong negative feedback effect of ~ -6 pcm/K, significantly stronger than solid-fueled fast reactors, and even the reduction due to feeding of spent nuclear fuel to ~ -3.8 pcm/K ensures the basis for a safe operation. The effect of the insertion of the dissolved vitrified waste is negligible.

The required chemical processes for the dissolution of vitrified material as well as the effects on the reactor chemistry and solubilities would have to be investigated in detail. However, this has to be weighed against the opportunities: (a) re-inserting the minor actinides into the reactor and burning them in the long-term without partitioning; and (b) the opportunity to open the way for improved conditioning or even reuse of some materials due to the reverse reprocessing leading to an element-wise sorted waste stream as well as the materials being captured in the off-gas treatment.

It has been demonstrated that burning of the minor actinide isotopes is accelerated with increasing amounts of previously separated waste inserted. This could be used as an optimization scheme; the insertion could be very high in the beginning, as a kind of 'burnable absorber' when the Pu production is strong. The subsequent initiation of the cleanup system at a later stage will then reduce the effect of the inserted fission products, which will free additional criticality reserve to keep the core stable. In addition, this strategy of an increased initial load will help to improve the burning rate of the trans-plutonium isotopes due to the correlation between the reduction rates and the amounts of Cm and Am

resident in the core. Thus, an early insertion would be beneficial to achieve high burning rates without the demand for partitioning.

The general conclusion is that the objectives of P&T are achievable without prior partitioning. This is a ground-breaking result; however, it will take significantly long operational times, which does not seem to be problematic as long as energy production based on nuclear power production is continued. In the case of a phase-out decision, this strategy would not really be promising. In this case, a much more efficient transmutation scenario would have to be envisaged, as already discussed several years ago for Germany [14].

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References

1. Arkhipov, V. Future Nuclear Energy Systems: Generating Electricity, Burning Wastes. Iaea Bulletin, 39/2/1997. 1997. Available online: https://www.iaea.org/sites/default/files/publications/magazines/bulletin/bull39-2/39204783033.pdf (accessed on 27 February 2015).
2. Merk, B.; Litskevich, D.; Detkina, A.; Noori-kalkhoran, O.; Jain, L.; Derrer-Merk, E.; Aflyatunova, D.; Cartland-Glover, G. iMAGINE—Visions, Missions, and Steps for Successfully Delivering the Nuclear System of the 21st Century. *Energies* **2023**, *16*, 3120. [CrossRef]
3. Merk, B.; Litskevich, D.; Whittle, K.R.; Bankhead, M.; Taylor, R.J.; Mathers, D. On a Long Term Strategy for the Success of Nuclear Power. *Energies* **2017**, *10*, 867. [CrossRef]
4. Merk, B.; Litskevich, D.; Peakman, A.; Bankhead, M. IMAGINE—A Disruptive Change to Nuclear or How Can We Make More Out of the Existing Spent Nuclear Fuel and What Has to be Done to Make it Possible in the UK? *Atw Int. J. Nucl. Power* **2019**, *64*, 353–359.
5. Merk, B.; Detkina, A.; Litskevich, D.; Patel, M.; Noori-kalkhoran, O.; Cartland-Glover, G.; Efremova, O.; Bankhead, M.; Degueldre, C. A First Step towards Zero Nuclear Waste—Advanced Strategic Thinking in Light of iMAGINE. *Energies* **2022**, *15*, 7209. [CrossRef]
6. Knebel, J.U.; Fazio, C.; Ait Abderrahim, H.; D'hondt, P.; Benamati, G.; Monti, S.; Gonzalez, E.; Pillon, S.; Warin, D. IP EUROTRANS: A European Research Programme for the Transmutation of High Level Nuclear Waste in an Accelerator Driven System. In Proceedings of the Actinide and Fission Product Partitioning & Transmutation Eighth Information Exchange Meeting, Las Vegas, NV, USA, 9–11 November 2004. Available online: http://www.oecd-nea.org/pt/docs/iem/lasvegas04/11_Session_V/S5_01.pdf (accessed on 10 June 2013).
7. Madic, C.; Boullis, B.; Baron, P.; Testard, F.; Hudson, M.; Liljenzin, J.-O.; Christiansen, B.; Ferrando, M.; Facchini, A.; Geist, A.; et al. Futuristic back-end of the nuclear fuel cycle with the partitioning of minor actinides. *J. Alloys Compd.* **2007**, *444–445*, 23–27. [CrossRef]
8. Merk, B.; Litskevich, D.; Peakman, A.; Bankhead, M. The Current Status of Partitioning & Transmutation and How to Develop a Vision for Nuclear Waste Management. *Atw Int. J. Nucl. Power* **2019**, *64*, 261–266.
9. Khaperskaya, A.; Khomyakov, Y.; Shadrin, A.; Feinberg, O. Conceptual approaches and the main directions of R & D on partitioning and transmutation of minor actinides and long-lived fission products in the Russian Federation State atomic energy corporation "Rosatom", Manchester. In Proceedings of the 15th International Exchange Meeting on Partitioning and Transmutation, Manchester, UK, 2 October 2018.
10. Merk, B.; Litskevich, D.; Bankhead, M.; Taylor, R.J. An innovative way of thinking nuclear waste management—Neutron physics of a reactor directly operating on SNF. *PLoS ONE* **2017**, *12*, e0180703. [CrossRef]
11. Acatech (Ed.) *Partitioning and Transmutation of Nuclear Waste*; Opportunities and Risks in Research and Application (acatech POSITION PAPER); Acatech: Munich, Germany, 2014.

12. Renn, O. *Partitionierung und Transmutation Forschung—Entwicklung—Gesellschaftliche Implikationen (Acatech STUDIE Dezember 2013)*; acatech–Deutsche Akademie der Technikwissenschaften: Munich, Germany, 2014.
13. Merk, B.; Detkina, A.; Litskevich, D.; Noori-Kalkhoran, O.; Cartland-Glover, G. A HELIOS-Based Dynamic Salt Clean-Up Study for iMAGINE. *Appl. Sci.* **2022**, *12*, 8748. [CrossRef]
14. Merk, B.; Litskevich, D. Transmutation of All German Transuranium under Nuclear Phase Out Conditions—Is This Feasible from Neutronic Point of View? *PLoS ONE* **2015**, *10*, e0145652. [CrossRef]
15. Merk, B.; Rohde, U.; Glivici-Cotruta, V.; Litskevich, D.; Scholl, S. On the Use of a Molten Salt Fast Reactor to Apply an Idealized Transmutation Scenario for the Nuclear Phase Out. *PLoS ONE* **2014**, *9*, e92776. [CrossRef]
16. *HELIOS2 Methods Manual*; Version 2.03.01; SSP-11/452 Rev 6; Studsvik Scandpower, Inc.: Idaho Falls, ID, USA, 12 January 2021.
17. Villarino, E.A.; Stammler, R.J.J.; Ferri, A.; Casal, J.J. HELIOS: Angularly dependent collision probabilities. *Nucl. Sci. Eng.* **1992**, *112*, 16–31. [CrossRef]
18. Wemple, C.A.; Gheorghiu, H.-N.M.; Stamm'ler, R.J.J.; Villarino, E.A. Recent Advances in the HELIOS-2 Lattice Physics Code. In Proceedings of the International Conference on the Physics of Reactors "Nuclear Power: A Sustainable Resource", Interlaken, Switzerland, 14–19 September 2008.
19. Evaluation and Viability of Liquid Fuel Fast Reactor System EVOL, DELIVERABLE D2.1, Design Parameters Definition for Most Stable Salt Flux, Rev 3 30/04/2012. Available online: https://cordis.europa.eu/project/id/249696 (accessed on 8 March 2022).
20. Brovchenko, M.; Kloosterman, J.-L.; Luzzi, L.; Merle, E.; Heuer, D.; Laureau, A.; Feynberg, O.; Ignatiev, V.; Aufiero, M.; Cammi, A.; et al. Neutronic benchmark of the molten salt fast reactor in the frame of the EVOL and MARS collaborative projects. *EPJ Nuclear Sci. Technol.* **2019**, *5*, 2. [CrossRef]
21. Merk, B.; Detkina, A.; Atkinson, S.; Litskevich, D.; Cartland-Glover, G. Catland-Glover: Evaluation of the Breeding Performance of a NaCl-UCl-Based Reactor System. *Energies* **2019**, *12*, 3853. [CrossRef]
22. Kozlowski, T.; Downar, T.J. *OECD/NEA and U.S. NRC PWR MOX/UO_2 Core Transient Benchmark*; Final Specifications, Revision 2; Purdue University: West Lafayette, IN, USA, 2003. Available online: https://www.oecd-nea.org/upload/docs/application/pdf/2020-10/mox_benchmark_specifications_2003.pdf (accessed on 5 June 2023).
23. *Aurora User Manual*; SSP-11/451 Rev 8; Studsvik Scandpower, Inc.: Idaho Falls, ID, USA, 12 January 2021.
24. *Zenith User Manual*; SSP-11/460 Rev. 5; Studsvik Scandpower, Inc.: Idaho Falls, ID, USA, 8 December 2020.
25. Merk, B.; Litskevich, D. On the Burning of Plutonium Originating from Light Water Reactor Use in a Fast Molten Salt Reactor-A Neutron Physical Study. *Energies* **2015**, *8*, 12557–12572. [CrossRef]
26. Merk, B.; Litskevich, D. A disruptive approach to eliminating weapon-grade plutonium—Pu burning in a molten salt fast reactor. *PLoS ONE* **2018**, *13*, e0201757. [CrossRef] [PubMed]
27. Rachamin, R.; Wemple, C.; Fridman, E. Neutronic analysis of SFR core with HELIOS-2, Serpent, and DYN3D codes. *Ann. Nucl. Energy* **2013**, *55*, 194–204. [CrossRef]
28. Detkina, A.; Litskevich, D.; Drury, M.; Merk, B. Zero Power Experiment for Molten Salt Reactor—Code Verification and Validation Request. In Proceedings of the International Conference on Physics of Reactors, Pittsburgh, PA, USA, 15–20 May 2022.
29. Merk, B.; Litskevich, D.; Gregg, R.; Mount, A.R. Demand driven salt clean-up in a molten salt fast reactor—Defining a priority list. *PLoS ONE* **2018**, *13*, e0192020. [CrossRef] [PubMed]
30. Merk, B.; Detkina, A.; Litskevich, D.; Drury, M.; Noori-kalkhoran, O.; Cartland-Glover, G.; Petit, L.; Rolfo, S.; Elliott, J.P.; Mount, A.R. Defining the Challenges—Identifying the Key Poisoning Elements to Be Separated in a Future Integrated Molten Salt Fast Reactor Clean-Up System for iMAGINE. *Appl. Sci.* **2022**, *12*, 4124. [CrossRef]
31. Degueldre, C.; Merk, B. Use of chloride-37 in Molten Salt Fast Reactor for ecological and economical reasons. *Prog. Nuclear Energy* 2023, *Submitted*.
32. Jaag, S.; Kiefhaber, E. *Americium Incineration—Challenge in Plutonium Burning and Minor Actinide Transmutation*; American Nuclear Society: La Grange Park, IL, USA, 1999.
33. Merk, B. Moderating Material to Compensate the Drawback of High Minor Actinide Containing Transmutation Fuel on the Feedback Effects in SFR Cores. *Sci. Technol. Nucl. Install.* **2013**, *2013*, 172518. [CrossRef]
34. Gulevich, A.V.; Eliseev, V.A.; Klinov, D.A.; Korobeinikova, L.V.; Kryachko, M.V.; Pershukov, V.A.; Troyanov, V.M. Possibility of Burning Americium in Fast Reactors. *At. Energy* **2020**, *128*, 88–94. [CrossRef]
35. Merk, B.; Devan, K.; Bachchan, A.; Paul, D.; Puthiyavinayagam, P.; Srinivasan, G. Can enhanced feedback effects and improved breeding coincide in a metal fueled, sodium cooled fast reactor? *Ann. Nucl. Energy* **2017**, *105*, 205–218. [CrossRef]

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