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Evaluation of the Breeding Performance of a NaCl-UCl-Based Reactor System

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Abstract: The energy trilemma forms the key driver for the future of energy research. In nuclear technologies, molten salt reactors are an upcoming option which offers new approaches. However, the key would be closed fuel cycle operation which requires sufficient breeding for a self-sustained long term operation ideally based on spent fuel. To achieve these attractive goals two challenges have been identified: achieving of sufficient breeding and development of a demand driven salt clean up system. The aim is to follow up on previous work to create an initial approach to achieving sufficient breeding. Firstly, identification of a salt system with a high solubility for fertile material and sufficiently low melting point. Secondly, evaluation of the sensitivity of the breeding performance on the sort of fissile material, the fissile material loading, and the core dimension all based on a realistic salt system which provides the solubility for sufficient fertile material to achieve the required breeding in a homogeneous reactor without breeding blanket. Both points are essential to create an innovative solution to harvest the fruits of a closed fuel cycle without the penalty of the prohibitively huge investments. It is demonstrated that the identified and investigated NaCl-UCl based systems are feasible to deliver the requested in-core breeding within the given solubility limits of fertile material in the salt system using either uranium as start-up fissile component or plutonium. This result is enriched by the analysis of the achievable full power days per inserted mass of plutonium. These new insights support reactor optimization and lead to a first conclusion that systems with lower power density could be very attractive in the case of low fuel cost, like it would be given when operating on spent nuclear fuel.

Keywords: nuclear energy; nuclear reactors; molten salt reactors; closed fuel cycle; breeding; homogenous reactor; spent nuclear fuel; sustainability; low carbon technologies

1. Introduction

The current growing interest in molten salt reactor technologies has been recently highlighted by the IAEA: "Initially developed in the 1950s, molten salt reactors have benefits in higher efficiencies and lower waste generation. Some designs do not require solid fuel, which eliminates the need for manufacturing and disposing of it. In recent years, growing interest in this technology has led to renewed development activities." [1]. For the use of molten chlorides, much of this analysis is based on historic reports of the 1970 [2,3] which have been developed in the follow up of the first operation of a reasonable sized molten salt test reactor, the MSRE [4]. The newly growing interest is reflected by a variety of start-up companies formed around the development of molten salt reactors like Terrestrial Energy, Elysium Industries, TerraPower, and many more [5] as well as through the formation of international projects, like EVOL [6] and the follow up SAMOFAR [7], most of them dedicated on developing a new reactor system.

In the final result, we intend to go much further to achieve closed fuel cycle operation, which has already demonstrated for the whole process with sodium cooled fast reactors on a laboratory scale [8]. Closed fuel cycle operation would be one of the additional key objectives for improving the sustainability of innovative nuclear technologies in general, we could call it the 'holy grail' of nuclear energy production by fission with different levels of achievement – on a decreasing level of quality, a fully closed fuel cycle with or without the inclusion of the P&R request, breed and burn, significant increase of target burnup in existing reactors. However, up to now establishing a real closed fuel cycle on an industrial level has not been successful due to proliferation concerns, the challenges of the MOX recycling, and the prohibitively high cost. Only parts of the technology have reached the industrial level, e.g. sodium cooled fast reactor operation (BN-600 and BN-800 in the Russian Federation) or the reprocessing of uranium based light water reactor fuel (COGEMA reprocessing facility in La Hague, France, and THORP at Sellafield in the UK), while production of and reactor operation on MOX fuel and especially the reprocessing of irradiated MOX fuel are still lagging. There is the additional opportunity, which has been worked out in the last years for molten salt reactors with the aim of harvesting the leverage of closing the nuclear fuel cycle operation without the massive upfront investment into the reprocessing and the MOX production facilities required when solid fuels are used. This could be achieved by operating molten salt reactors fuelled with existing spent nuclear fuel from currently operating thermal nuclear power plants [9,10]. A preferable side effect would be reducing the challenges in the long-term nuclear waste management in a way comparable to the promises of the partitioning and transmutation approach [10,11] leading to a combined solution for nuclear system development, overcoming the traditional separation of waste management and power production [12,13]. To achieve these very attractive goals two basic challenges have to be solved:

- Achieving sufficient breeding in a molten salt system
- Developing of a demand driven salt clean up system [14]

The aim of this publication is to create an initial approach to the first point given above by:

- Identifying a molten salt system with high solubility for the fertile component, in our case uranium and sufficiently low melting point for reactor operation.
- Evaluating the sensitivity of breeding performance of systems on the identified salts based on the sort of fissile material, the fissile material loading, and the core dimension

This publication has to be seen in a much wider space of developing a promising nuclear energy system for the future to provide an urgently demanded low carbon opportunity to tackle the climate change challenge while reducing the long term waste challenge–iMAGINE, a disruptive nuclear energy system operating on spent nuclear fuel from LWR without prior reprocessing [9,13]. Thus, the general approach of achieving a closed fuel cycle system using molten salt reactors as already given in the existing work of Mourogov at al [15] on REBUS-3700 is consequently deepened here into another level, disrupting the whole nuclear energy system (reactor with the whole related fuel cycle). REBUS-3700 has been introduced as a future reactor concept based on a classical heterogeneous fast reactor core concept (core with breeding blanket) like in sodium cooled fast reactor technology [16] and with a salt clean-up based on classical reprocessing—not explicitly worked out, but most probably based on the pyro-reprocessing techniques that were available in 2006. Unfortunately, the system did not get the attention such an innovative work should have deserved. In contrast to REBUS-3700, iMAGINE will be based on a homogenous core design without breeding blanket, a demand driven salt clean-up and an optimization process between reactor physics and separation chemistry (more details are given in [14]). The homogeneous core without a blanket provides a much harder challenge to achieve sufficient breeding, but it provides a much more proliferation resistant system since no fresh, separable Plutonium will be formed in a separate system. To achieve this aim, a salt system with a higher fertile content has to be identified and consequently the analysis of the salt system as well as the detail of the breeding performance analysis will be executed in much more detail and over a wider space than in the existing work of Mourogov at al [15] to create a basis for an evidence based multi-disciplinary discussion and optimization of molten salt reactor development for closed fuel cycle operation.

2. The Salt System

The choice of the salt system is based on a literature search identifying [17] as one of the currently best available sources. The current major criteria for the choice of the salt system are:

- Request for a high load of fertile material to assure that sufficient breeding enables their operation in the future on spent nuclear fuel produced in existing thermal reactors
- Preference for low cost components of the salt system
- Preference for chlorine based salts due to the available experience in the UK on chlorine salts in advanced reprocessing projects and due to the available experimental facilities

Based on the above given criteria we have identified the NaCl-UCl_n system as a promising system to investigate with the focus on the NaCl-UCl_4 system as given in Figure 1. The following modelling and simulation study will be based on this system as reference point.

E1: 30%; $430 \pm 5\,°C$; $L \rightleftharpoons NaCl + 2\,NaCl{-}UCl_4$
C: 33.3%; $440 \pm 5\,°C$; $L \rightleftharpoons 2\,NaCl{-}UCl_4$
E2: 47%; $370 \pm 5\,°C$; $L \rightleftharpoons 2\,NaCl{-}UCl_4$ + unknown unknown
P: 57%; $415 \pm 5\,°C$; $L \rightleftharpoons 2\,NaCl{-}UCl_4$ + unknown compound

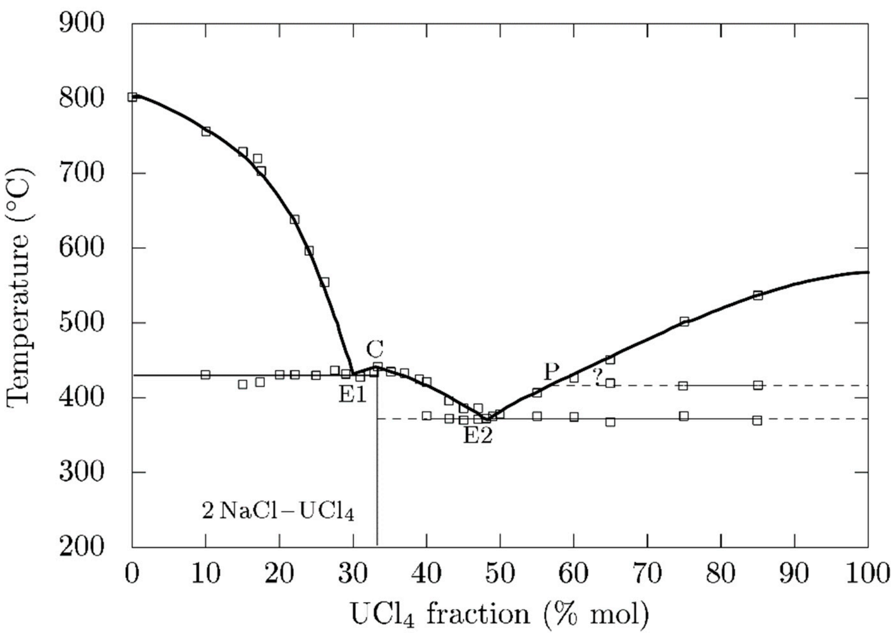

Figure 1. Invariant equilibria and phase diagram of the salt system NaCl-UCl_4 [17].

The figure delivers first data, but the publication date of 1943 of the original study [17] from which the data was obtained already indicates that one of the first requests would be to re-evaluate this system with modern analytical methods to provide a full data set on the thermo-physical properties which are required for modern modelling and simulation tools. A search for a reasonable density curve for the NaCl-UCl system lead to the publication [18] providing a density field for 1050K and the 3-component system NaCl-UCl_3-UCl_4, see Figure 2.

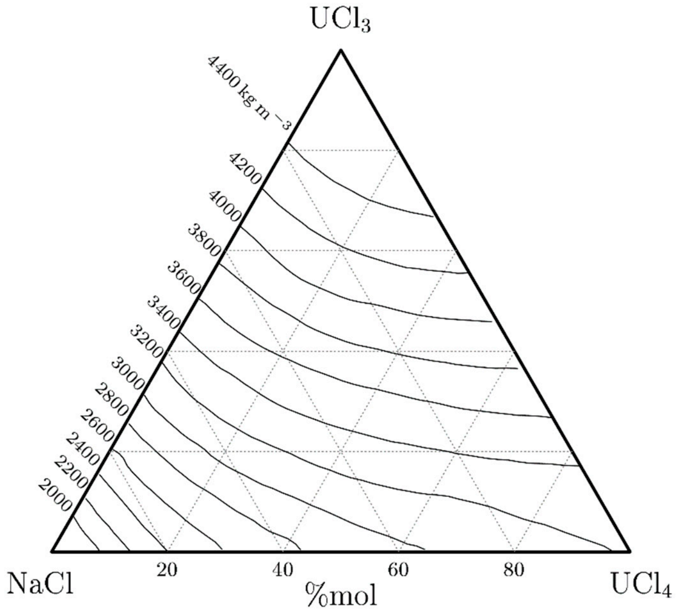

Figure 2. Iso-density curves (g/cm^3) of the tertiary melt of the mixture NaCl-UCl_3-UCl_4 at 1050 K [18].

This reference provides an additional salt system: NaCl-UCl_3-UCl_4 with the eutectic composition 42.5%–17.0%–40.5%, which seems to be worthy of further detailed investigation. The eutectic point of the NaCl-UCl_3-UCl_4 salt has a melting point of 338 °C [18], see Figure 3. The main advantage of this system would be the higher loading of fertile material at the eutectic point, which will be the optimal, but most probably not the exactly achievable operational point over the whole lifetime of the reactor. However, in general, this composition will request the same kind of 'upgrading' of the thermo-physical dataset and due to the close chemical compositions, this could be achieved in one experimental setup. The real reactor operation would most probably be somewhere in between, since the insertion of another trichloride component, $PuCl_3$ would drive the NaCl-UCl_4 system into the direction of the three-component system, by potentially replacing a part of the UCl_3 with $PuCl_3$.

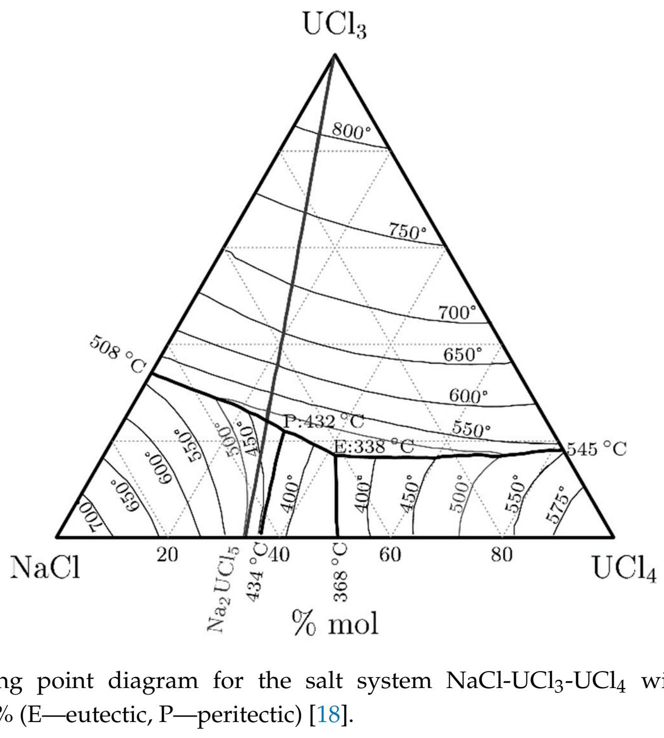

Figure 3. Melting point diagram for the salt system NaCl-UCl_3-UCl_4 with the eutectic at 42.5%–17.0%–40.5% (E—eutectic, P—peritectic) [18].

3. Materials and Methods

This section presents the salt data, the used code, and the developed model. Based on the given salt system investigation above, the applied standard salt composition for the study is based on [17] using the eutectic composition of NaCl-UCl_4 with 47% UCl_4 with a melting point of 370 ± 5°C as the reference case. However, the study is carried out for a slightly under stochastic composition to take care for the later operational requirement of keeping the chlorine level while uranium and plutonium are fissioned. For this instance and to avoid the appearance of pure and un-buffered UCl4 which could cause corrosion problems, a composition of $UCl_{3.8}$ is used for the determination of the nuclide composition. The requirement plutonium for the studies cases is added as independent part of $PuCl_3$ reducing the overall salt composition of NaCl-$UCl_{3.8}$ when inserted. The density is estimated to be 3.2 g/cm^3 at 1050 K, which is the only value which has been found based on the data in [18].

In addition, two major variations are investigated:

The use of enriched Cl-37 in the chlorine component of the NaCl versus natural NaCl with a significant natural Cl-35 share of 76%. It is expected that this change will have a negative influence on criticality and breeding, but the use of natural NaCl could offer clear cost advantages.

The use of the three component system as described in [18], consisting of NaCl - UCl_3 - UCl_4, 42.5% – 17.0% – 40.5%. In this case the inserted plutonium ($PuCl_3$) is estimated to replace a share of the UCl_3. In this case a density of 3.2 g/cm^3 is applied based on the diagram given in [18]. It is expected that this composition will show an improved breeding performance compared to the reference case.

The simulations for this feasibility and sensitivity study have been performed using the POLARIS module of the SCALE code system [19]. Polaris is a new module for SCALE 6.2 that provides a 2D lattice physics analysis capability that uses a multigroup self-shielding method called the Embedded Self Shielding Method (ESSM) and a transport solver based on the Method of Characteristics (MoC). In general, POLARIS and its cross section library has been developed and validated for light water reactors, thus we will provide at the end of this chapter a first verification/validation against the Monte-Carlo code SERPENT [20]. Polaris is integrated with ORIGEN for depletion calculations on a sub region basis [21]. For the calculations, the v7-252 cross section set and a set of ten burnup calculations at 0, 1, 5, 10, 20, 30, 40, 50, 60, 80 GWd/t_{HM} without consideration of online feeding and online salt clean-up, is used. It has to be mentioned here, that in contrast to a real molten salt reactor, the model will produce an artificial spatial burnup distribution in the material, which will not arise in the real liquid fuelled reactor. This is a limit of the application of a lattice code, which has been developed to model solid fuelled reactors. In addition, we have to clarify here, that this application is significantly outside of the scope of the validated standard applications of the code.

For the evaluation of the breeding, a very specific 2-D model (infinite cylinder) has been built for the simulation (see Figure 4 with included calculation regions) to take care of the very important effect of the neutron leakage from a molten salt reactor. The model consists of the reactor core with variable size (red), surrounded by a stainless steel vessel of 20 mm thickness (bright yellow) and a sodium reflector of 50 cm thickness (dark yellow). The arrangement is surrounded by a vacuum gap (blue rings) and the rest of the unit cell is filled with a strong absorber (outer blue) to avoid the reflection of leaking neutrons from the core/reflector system. This model is appropriate for evaluating the breeding performance which is the aim of this publication. For a real operational simulation, a more sophisticated multi-physics model would be required. A first approach has been delivered by the SCALE group of Oak Ridge National Laboratory [20].

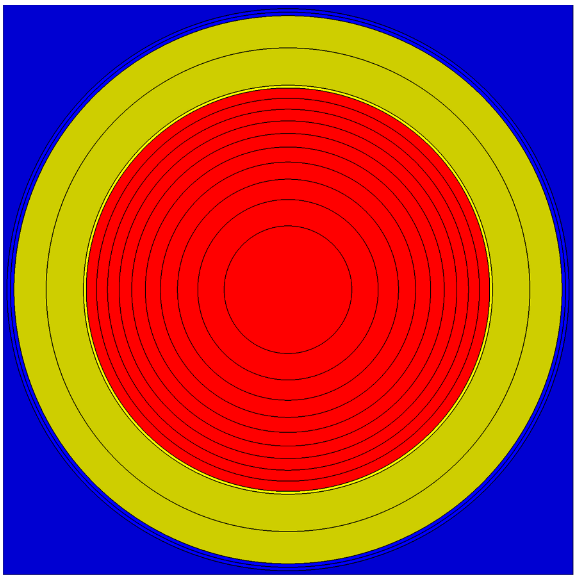

Figure 4. Model of the molten salt reactor core surrounded by absorber.

The fuel temperature in the core is set to an average of 750 °C, the vessel temperature is set to 600 °C, and the reflector temperature is 527 °C. The studies are based on the use of enriched uranium and on the use of plutonium with the vector given in Table 1. In the Pu based cases natural uranium is used as fertile material. The parameters studied are the radius and the fissile material content.

Table 1. Plutonium vector used in the study [14].

Pu-238	0.0025
Pu-239	0.6877
Pu-240	0.267
Pu-241	0.0176
Pu-242	0.0252

4. Results

4.1. Code Comparison for Model Validation

A first verification/validation of the quality of the SCALE/POLARIS application for a molten salt fast reactor is based on a comparison to the SERPENT Monte-Carlo code for one specific case (see Figure 5) based on using identical modelling approaches as given above.

A first comparison of the SCALE/POLARIS results and the Monte-Carlo results show a steady state offset of ~2400 pcm for the ENDF/B 7.1 library and an offset of ~1500 pcm for the ENDF/B 7.0 library. These numbers seems very high when compared to LWR modelling quality, but we have to consider here, that the system is a fast reactor system and fast reactor systems tend to be much more sensitive, thus there the calculation accuracy is lower. In addition, we have to keep in mind that the used libraries are not identical and that a part of the core composition is made up of chlorine, which isn't a standard nuclear reactor material (e.g., problems of ^{35}Cl(n,p) XS). In addition, the code in the current state uses a multi-group approach with a group structure developed for LWR analysis and the model is based on flat flux approximation within the, for a lattice code, very large calculation regions. The strong influence of the library in the two Monte Carlo simulations (more than 800 pcm) shows that the uncertainty is still high for the given combination of materials and neutron spectrum, which identified a demand for improving the cross section basis before going into deeper calculations. Taking all these factors into account and remembering that the calculations aim to demonstrate the feasibility of a breeding system, the application of SCALE/POLARIS seems appropriate. Over all, besides the initial offset, the burnup curve shows only a relatively robust bias over the whole burnup of ~2400 ± 200 pcm. Thus the results for the breeding evaluation will be rather robust, since both codes deliver comparable results.

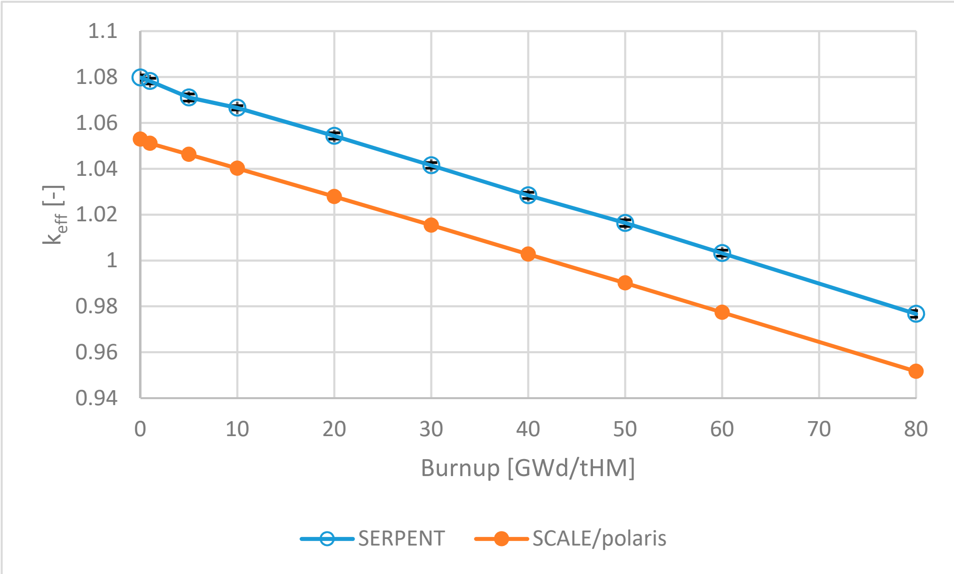

Figure 5. Code comparison regarding criticality over burnup.

A first justification of the bias and how large the influence of the bias is, is given in Figure 6. To compensate the bias an increase of ~1% in the ^{235}U enrichment would be required, which relates to ~5% error in the enrichment determination or the system size would have to be increased by ~8 cm in radius which relates to a change of ~7% in the estimated diameter. In a real breeder design, the preferred approach would be to increase the core size, since increasing the fissile loading tends to decrease the breeding efficiency [16]. Both results indicate that the accuracy of the simulation is acceptable for the envisaged feasibility study.

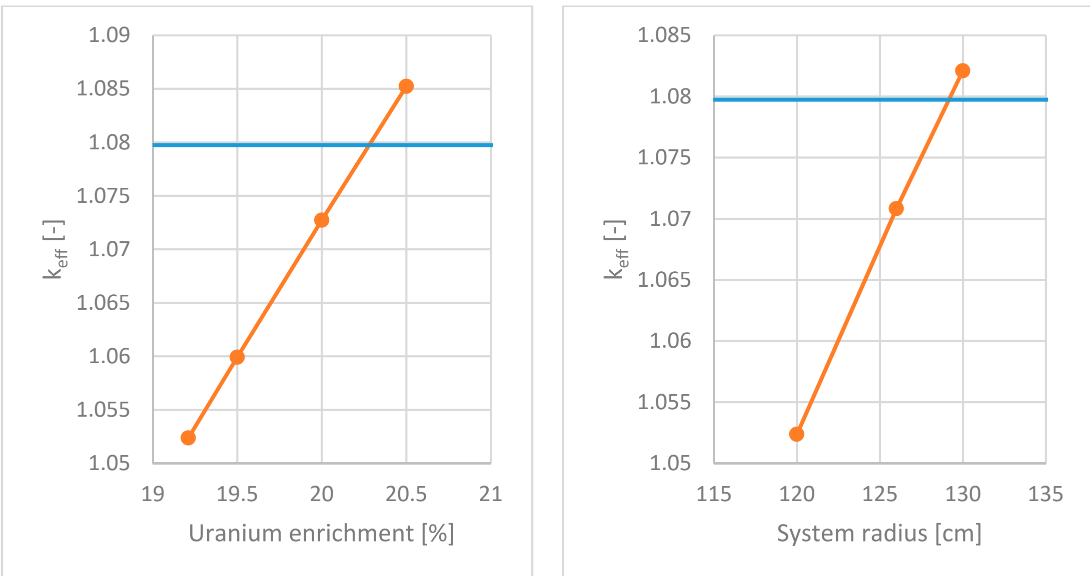

Figure 6. Required changes in uranium enrichment (**left**) and system radius (**right**) to adopt the POLARIS results to the SERPENT reference result.

However, when considering the change from a 2D system to a spherical 3D system, the leakage will significantly change and this would finally result in a very sharp drop in the criticality of the test system to k_{eff} = 0.911. Thus, in a future case when considering the 3D realistic core a significant correction has to be envisaged or the development of a more reliable and sufficiently efficient 3D tool.

4.2. Results and Discussion for the Two Component System

Natural chlorine consists of two isotopes, ^{35}Cl and ^{37}Cl, with a composition of 76% to 24%. Currently, most of the chlorine based molten salt reactor systems aim to use enriched ^{37}Cl. This has two reasons, the significantly higher neutron absorption cross section of ^{35}Cl and the possibility of the formation of ^{36}Cl by activation processes, a radioactive isotope with long half-life [22]. However, for a system with a very high content of uranium and a comparably low content of NaCl it could be interesting from economic point of view, to test the effect of natural NaCl (76% ^{35}Cl and 24% ^{37}Cl), while enriched ^{375}Cl is still used for the production of the $UCl_{3.8}$ where the Cl content is significantly higher. When natural NaCl is applied for the investigation $UCl_{3.8}$ this leads to a final composition to a ^{35}Cl content of ~18%. Thus, the decision should finally be made on the influence of the ^{35}Cl content on criticality, on the cost (and the safety case) of producing NaCl with enriched ^{37}Cl, and on the cost of the formed ^{36}Cl to be forwarded to the final disposal at the end of operation. Whereupon the formation of ^{36}Cl has to be seen and evaluated in recognition of all other long lived fission products, which will be formed in the reactor. The investigation of the effect on criticality is given in Figure 7. There appears a clear criticality reduction (~2200 pcm, compare the red line against the blue line) over the whole observed burnup period due to the ^{35}Cl in the NaCl component of the fuel. This loss in criticality has either to be compensated by a slight increase of the core size leading to a reduced leakage or by an increase of the content of fissile material. Or to look at it the other way round, the higher criticality achieved by using enriched ^{37}Cl in the sodium chloride can be used to reduce the initial enrichment of fissile material. To match both criticality curves the case with natural NaCl requires a ^{235}U enrichment of 19.21% to achieve a slightly positive Δk_{eff} integral over the considered burnup, which is required to compensate for the neutron leakage in the third dimension and to create a reasonable modelling of the breeding without a significant disturbance caused by the normalization of the fission source. The system with enriched ^{37}Cl in the NaCl achieves almost the same initial k_{eff} and the almost identical Δk_{eff} integral with only 18.32%. The detailed comparison of the k_{eff} over burnup in Figure 7 indicates a slightly improved breeding (less steep decrease of k_{eff} over burnup, see grey curve) caused by the reduced initial enrichment. The influence seems negligible in comparison with the expected accuracy of the calculations (comparison to the Monte-Carlo result). However, it is relevant, since it confirms the expectation that a core with lower fissile content tends to deliver improved breeding performance [16], and here we approach the level of a sensitivity evaluation since both results are produced with an identical code setting besides the enrichment of fissile material.

In general, an economic analysis will be required to identify the preferred solution, which will be influenced by the cost of the ^{37}Cl enrichment, the production of the specific NaCl, and the core size determining the amount of salt required as well as the follow up cost of the larger system and the cost of disposal of an additional radioactive material, the bred ^{36}Cl, the salt activation and the Sulphur production. However, there is an additional argument, as using natural NaCl is inherently safer as it requires a smaller quantity of chlorine at all and fewer unit operations to prepare the salt. We have to keep in mind, even without the nuclear part dealing with the chlorine will require special safety procedures. This gives evidence for the feasibility from a reactor physics point of view for future discussions on economy, chemistry, and nuclear waste management.

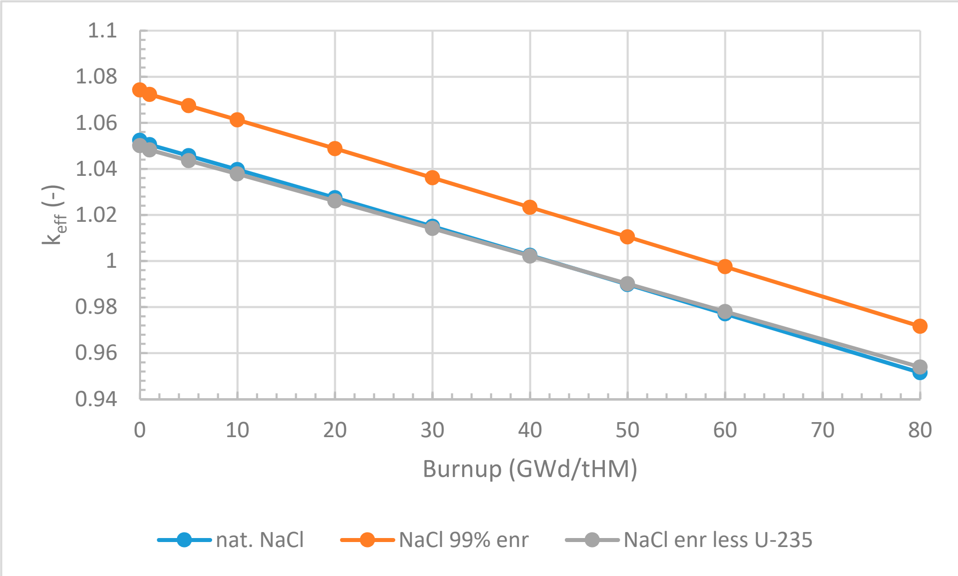

Figure 7. Comparison of the burnup behaviour of NaCl-UCl fully salt based on 99% enriched ^{37}Cl and a system using natural NaCl mixed with UCl based on 99% enriched ^{37}Cl.

The dimension search is started with a uranium based core composition using a two component system with 53% NaCl and 47% $UCl_{3.8}$. The first system size, which achieves with a low enriched uranium loading, a sufficiently positive Δk_{eff} integral that is compensated for the leakage in the third dimension over the considered burnup period of 80 GWd/t_{HM} has a radius of 120 cm in the model and required an initial ^{235}Uenrichment of 19.68%. The burnup specified here is a requirement for the simulation of the system to be kept in a reasonable balance over the whole period considered in order to get a reliable breeding estimation. The steep k_{eff} curve over burnup requires a significant excess reactivity at the beginning of the calculation, which would not be the case in an actual molten salt reactor. However, it indicates that the criticality of the system reduces significantly due to the consumption of the fissile material and a concomitant accumulation of fission products, this therefore indicates that there is insufficient breeding. The initial excess reactivity is needed to get the mentioned slightly positive integral of Δk integrated over burnup. This integration is essential to achieve a realistic model for the breeding under the approximations of a lattice code with normalization of the fission source in the k_{eff} calculation – in the case of a positive integral (0 to 40 GWd/t_{HM}) breeding is underestimated due to the normalization reducing the fission source, while in the negative integral (40 to 80 GWd/t_{HM}), breeding is overestimated due to the artificial increase of the neutron source by normalization. In a real molten salt reactor with online processing, the strong criticality decrease would be compensated by the feeding of additional fissile material, while the fission product accumulation would partly be reduced by the foreseen online salt clean-up – both online processes are not considered here, since the aim is just to investigate the breeding and the dependence on the system dimension based on a model with reduced complexity. The dimension search in Figure 8 shows the reduced loss in k_{eff} over burnup with an increased system size and a reduced initial enrichment requirement (16.53% for radius 145 cm and 14.73% for 170 cm).

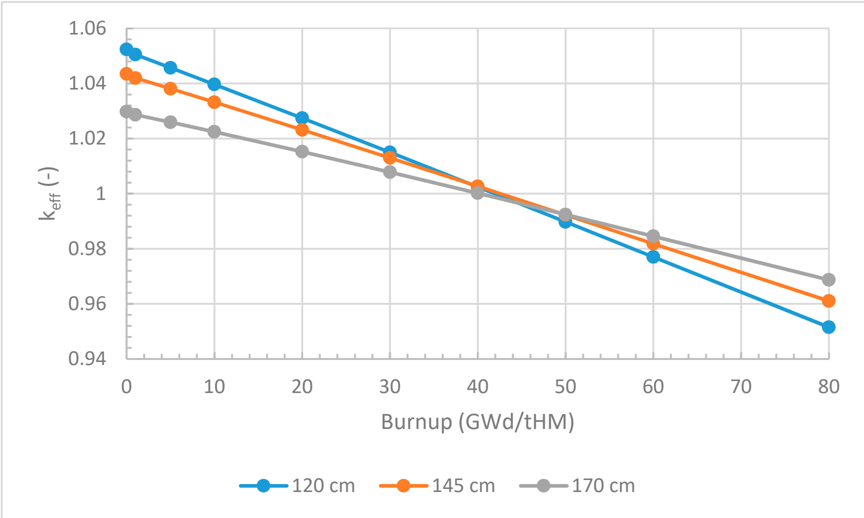

Figure 8. Burnup studies for a NaCl-UCl based molten salt reactor for different 2d reactor dimensions using fuel based on enriched uranium.

In general, the results for the increase of the system size indicate that a system with sufficient breeding can be found based on the studies sizes, but the size of the POLARIS model would have to be increased to move to larger core sizes. To avoid this change the next step will be performed by changing the nature of the fissile material from ^{235}U to plutonium. The use of plutonium as fissile material improves the breeding potential of the system significantly, see Figure 9. The k_{eff} over burnup curve is clearly flattened for each of the considered cases when plutonium is used as fissile material instead of ^{235}U, when comparing the dotted lines with circles and the solid lines with squares. The most interesting case is the 170 cm radius with plutonium as fissile material. This case delivers an almost flat k_{eff} over burnup curve over the whole burnup period of 80 GWd/t_{HM} considered, even for the applied reduced modelling conditions, which omit the effect of the salt clean-up.

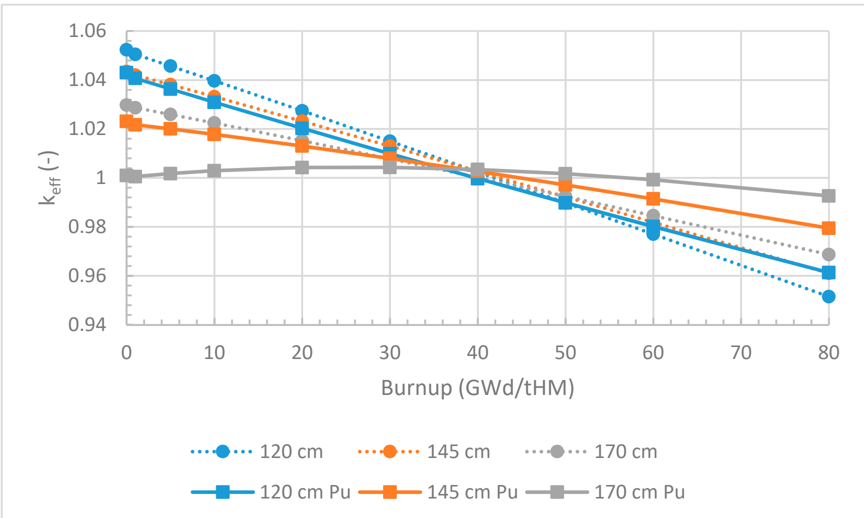

Figure 9. Comparison of burnup studies for a NaCl-UCl based molten salt reactor for different 2d reactor dimensions using fuel based on enriched uranium as well as natural uranium with plutonium as fissile component.

The results given in Figure 9 confirm that it will be possible to achieve sufficient breeding in a molten salt system based on the investigated NaCl-UCl_4 with 47% UCl_4 system under the given modelling conditions ($UCl_{3.8}$, 1050K average fuel temperature, 3.2 g/cm^3 salt density, 1239 MWth power), but for the current results only if Pu is used as fissile material. However, the results can only be seen as a preliminary outcome, since to achieve a really robust modelling and simulation result, the input database will have to be improved by applying a more appropriate reactor design and a robust modelling approach for a molten salt reactor with online processes for feeding and clean-up, as well as by improved experiments to determine the thermo- physical data of the salt composition in more detail. However, these first results indicate that despite the uncertainties described above, the proposed salt configuration can be used for the design of a breeding based molten salt reactor system with a reasonable reactor size of ~3.5 m diameter in a spherical configuration. A more precise modelling and simulation approach would be required to achieve a realistic design including the effects of the external loops required for cooling. However, it becomes obvious that the Pu driven core is more efficient since plutonium produces a higher number of neutrons per fission in the fast neutron spectrum than ^{235}U. Thus more neutrons are available for capture reactions leading to breeding.

A more detailed analysis of breeding in the case with Pu as a fissile material and with a wider varying model size is given in Figure 10. It is clearly visible that the breeding improves with increasing system size and the correlated lower initial content of Pu as fissile material, see Table 2. After the breakthrough is almost reached with a radius of 1.7 m, the two larger systems show a clear increase of k_{eff} during the operation, which clearly indicates that efficient breeding is taking place, there is even enough breeding to compensate the accumulation of fission products. However, it has to be kept in mind that these two cases start with a k_{eff} lower than one, thus on the one hand the start-up of the system wouldn't be possible, while on the other hand the result is slightly adulterated due to the normalization of the initial fission source creating a slightly higher fission source than in a realistic case. This will lead to a slight improvement of the initial breeding process, even if the integral of Δk over burnup is observed to be slightly positive. In addition, the results do not tell the complete truth since the Pu content is only given in absolute atom% of Pu, but the real absolute amount of Pu depends on the volume of salt in the core, while this volume depends on the radius and the burnup is normalized to the ton of heavy metal which also changes.

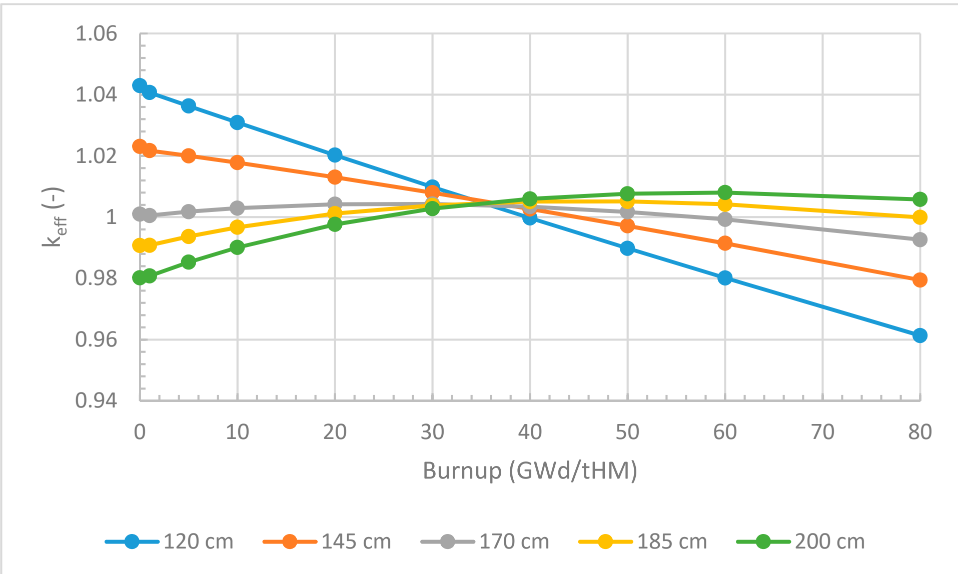

Figure 10. Study of the breeding performance of a NaCl-UCl system with fissile Pu component for different 2D system radii.

Table 2. Absolute Pu content in atom% over the radius of the system.

Radius [m]	Absolute Pu Content [%]
1.2	8.00%
1.45	6.89%
1.7	6.13%
1.85	5.81%
2	5.54%

To reflect the above described issue, the curves of Figure 11 have been renormalized taking into account the absolute amount of Pu in the core as well as the change in the absolute heavy metal content, while keeping the core power constant for all cases. In this figure it becomes clear that a larger core, with the identical power, thus consequently with a lower power density will operate for a much longer time period based on the initial loading and an improved breeding performance. Therefore, the inclusion of more Pu seems to be a very attractive alternative, at least as long as the fuel cost is low enough and as long as the plutonium doesn't degrade in the core, as it was observed in the CAPRA simulations. This brings us back to the initial study and why it would be important to be able to use natural NaCl. When this is combined with the operation of the reactor on spent nuclear fuel from light water reactors as proposed in [9,10], the larger core alternative could be of high interest which is in very strong contrast to typical fast reactor designs and they are typically designed around a very high power density mainly due to the high fuel cost related to the production of MOX fuel [23].

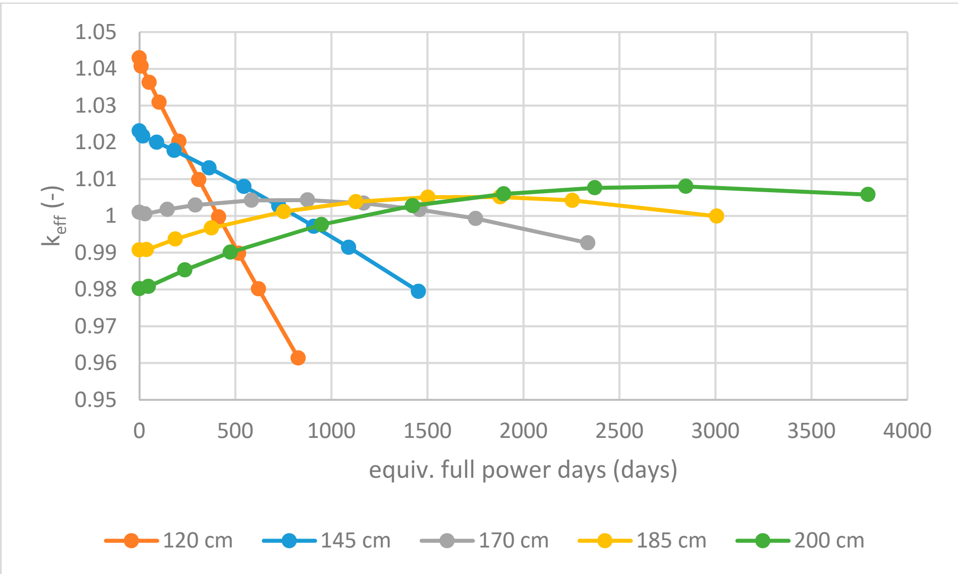

Figure 11. Achievable equivalent full power days in a one off-loading a NaCl-UCl system with fissile Pu component based on different 2D system radii and the loading for a spherical system.

However, since the core is based on Pu, which is in the case of UK is available from the reprocessing and the related stockpile, it will be worth to put an additional glance on the Pu utilisation in the different investigated core dimensions. The aim has to be to increase the Pu utilisation with increased core dimension and the required increased Pu loading in the core, only when this condition is fulfilled will the increase of the core dimension be efficient. This mentioned Pu utilisation in equivalent full power days per inserted ton of Pu is investigated in Figure 12.

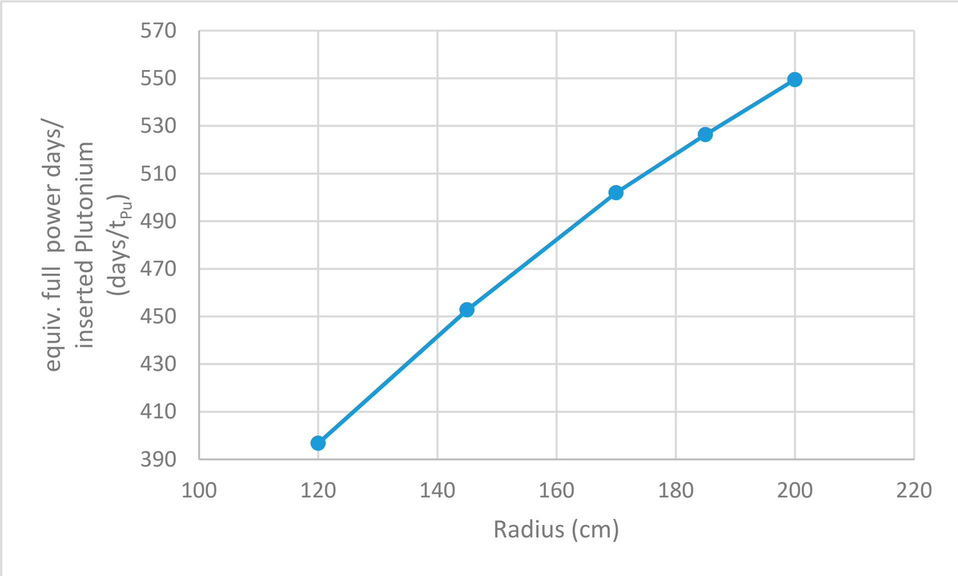

Figure 12. Achievable equivalent full power days per inserted amount of plutonium in a one off-loading a NaCl-UCl system with fissile Pu component based on different 2D system radii and the loading for a spherical system.

There is a clear increase in the number of full power days per inserted amount of Pu visible, which confirms that the number of full power days increases with increasing core radius faster than the amount of Pu that is required to operate the system. The result confirms the expectations that breeding is traditionally more efficient with reduced initial Pu loading per unit volume [16] as well as coinciding with the opposite observation that the breeding performance and the Pu fissile quality tend to degrade with very high Pu content in cores. This effect has been observed in the modelling and simulation investigations of the CAPRA project, which investigated plutonium burning [24]. Based on this final test, it seems from a reactor physics point of view a really attractive approach to accept a lower power density, based on the comparable low fuel cost for the proposed NaCl-UCl system (compared to MOX fuelled fast reactors) and the significantly better breeding performance. This could be of particular interest when considering the operation of a reactor on spent nuclear fuel from LWRs creating the link between power production [9,13] and waste management [10,12]. However, for a final decision it would be better to have a full economic analysis, which evaluates not only the reactor physics, but also the cost of construction and operation.

4.3. Results and Discussion for the Three Component System

Following the approach given in the introduction the detailed analysis of the two component system will be compared in the section with the three component system [18] with it's clearly higher UCl content (59.5% instead of 47 % in the two component system) in the salt composition. Figure 13 shows the dimension search for the 2-D system indicating an almost identical breeding behaviour than in the two-component system (cf. Figure 10). The results indicate that a system with a radius of 170 cm or more is required to achieve sufficient breeding in a self-sustainable mode over an acceptable operational burnup of at least 60 GWd/t_{HM}.

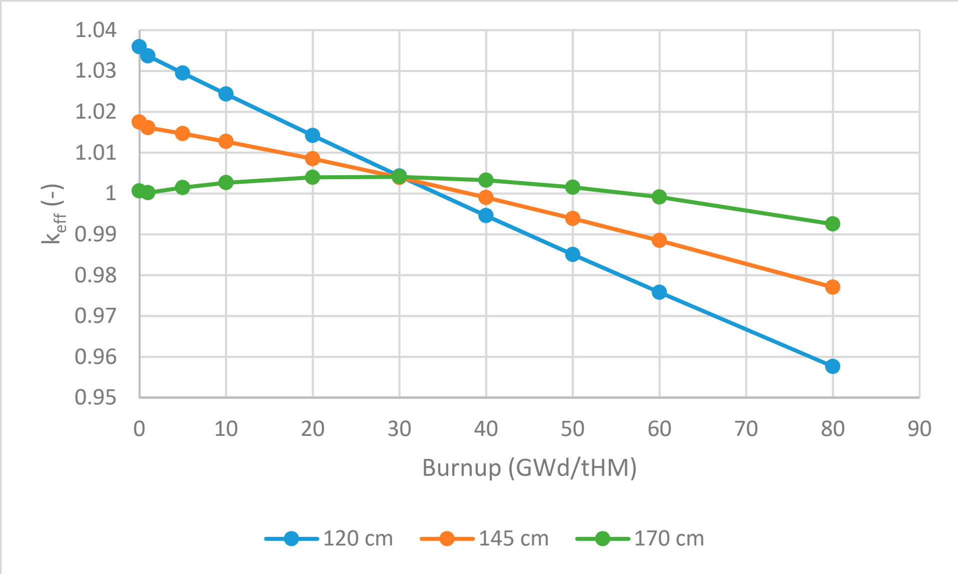

Figure 13. Study of the breeding performance of a three component NaCl - UCl3 - UCl4 system with fissile Pu component for different 2D system radii.

A more detailed comparison of the behaviour of the two and the three-component system is given in Figure 14 showing a very tiny improvement of the breeding in the three-component system. A more detailed look into the modelling of the different systems indicates why we see an almost identical behaviour. The two-component system was configured by adding plutonium to a given salt composition (47% UCl in 53% NaCl) since Pu typically forms $PuCl_3$. Thus, when adding about 6% PuCl to the leading salt composition, the overall heavy metal salt content in the mixture increases to 50%. This approach has been used due to the lack of information about the achievable composition and it is not investigated regarding the solubility issues of the composition. In contrast to this adding approach, a replacing approach has been used in the three-component system. The addition of the $PuCl_3$ has been compensated by reducing the UCl_3 component in the mixture to ensure that in this approach the solubility issues are considered. It seems most likely that the Pu component will replace a part of the U component with the identical oxidation state. Thus, this approach is from chemical and solubility point of view the more correct one. The detailed information on all the compositions calculated is given in Table A1 for comparison. A detailed comparison of the two 1.7 m cases (row 7 and row 12) shows that for the exactly identical Pu content, the fissile content, normalized on the overall heavy content, is lower in the three component case.

The three component system is only slightly better in breeding, but there is still the question of the quality of the density data and the mentioned solubility issue for the two-component system. In general, it should be expected that the three-component system delivers a higher density due to the higher uranium content, 47% in the 2-component system versus 59.5% in the three-component system (compare again row 7 and 12 of Table A1 for the HM content). However, the current database does not give sufficiently detailed data on the density to make a final conclusion to study all details. This leads to the conclusion, that improved basic thermo-physical knowledge is essential for more detailed and reliable studies on molten salt systems based NaCl-UCl systems and the composition related breeding performance.

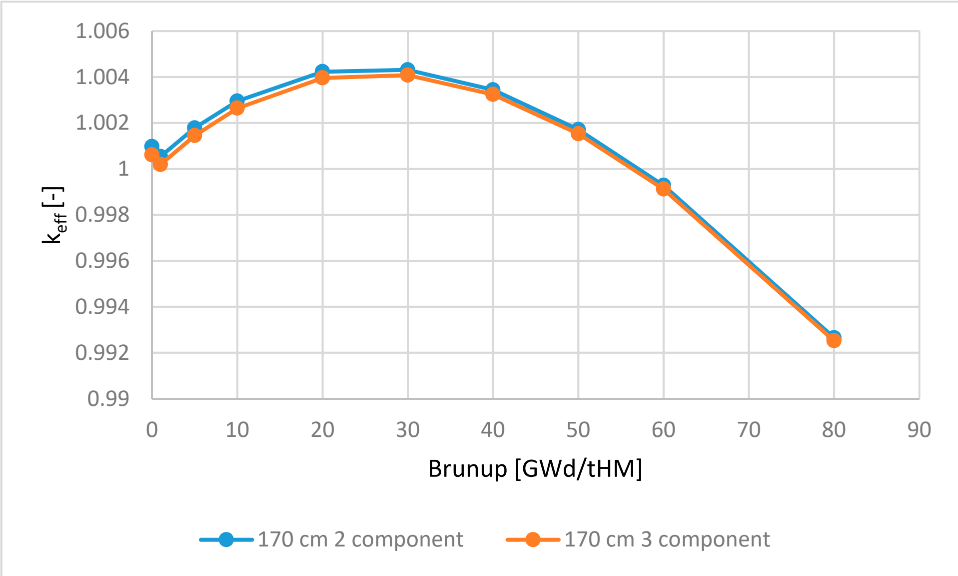

Figure 14. Comparison of the performance two and the three component system for the identical radius of 145 cm.

This leaves just one final question open, how could this system work for countries which do not possess a stockpile of separated Pu as is the case in the UK. Would it be possible to achieve a reasonable breeding in a system operated on enriched uranium? In general, it should be possible, since the Russian BN reactors are traditionally mainly operated on enriched uranium [25]. This final investigation is based on the three-component system, since improved breeding performance is expected due to the higher uranium loading for this salt composition. Figure 15 indicates that breeding in a uranium based system is possible, however only on the cost of a significantly larger system with a significantly higher amount of heavy metal in the core (see Table A1). The result confirms the expectation that it is possible to operate a fast reactor on ^{235}U as fissile material, but it is much less efficient than the operation on plutonium. The main reason is the significantly lower amount of neutrons created per fission event in uranium than with plutonium. At an incident neutron energy of 100 keV it is more than 0.5 neutrons more (+40% neutrons per fission) in ^{239}Pu and ^{241}Pu, than for ^{235}U and about the same at 1 MeV neutron energy (~+25% neutrons per fission). A detailed list of the results and input values like fissile enrichment is given in Appendix A, Table A1.

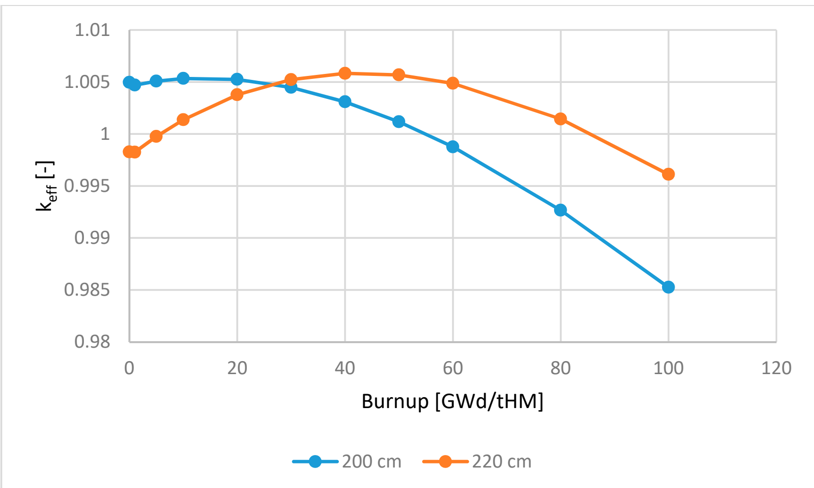

Figure 15. Study of the breeding performance of a three component NaCl-UCl3-UCl4 system with fissile U-235 component for different 2D system radii.

5. Conclusions

In nuclear technologies, molten salt reactors have gained growing interest since they can offer new approaches. However, the key for making nuclear an attractive, highly sustainable option for a future low carbon society would be to operate these reactors in closed fuel cycle mode providing sufficient breeding for a self-sustained long term operation based on a pure fertile feed or even based on spent nuclear fuel from existing reactors.

To achieve these very attractive goals two basic challenges have already been identified, the achievement of sufficient breeding in a molten salt system and the development of a demand driven salt clean up system. In this publication we followed up on the previous work to create an initial approach to the first point, achieving sufficient breeding by a two-step approach. Firstly, we identified two molten salt systems with the required high solubility for the fertile component essential to achieve the required breeding in a homogeneous reactor without breeding blanket. In our case we identified NaCl-UCl_n systems with specifically the two component system NaCl-UCl_4 and the 3-component system NaCl-UCl_3-UCl_4 both with sufficiently low eutectic melting points, 370 °C and 338 °C for reactor operation and high fertile solubility for breeding. Secondly, we evaluated the sensitivity of the breeding performance of a system based on the identified salts. We investigated breeding based on the sort of fissile material demonstrating the clearly better performance of the Pu based system. However, it has been shown that self-sustained breeding would also be possible in a slightly larger enriched uranium based system to confirm a possible startup of the system based on enriched uranium fuel. The fissile loading is adopted in all cases to achieve sufficient criticality to get a robust breeding estimation dependent on the core dimension. The demonstration of both points is essential to create an innovative solution to harvest the fruits of a closed fuel cycle without the penalty of the prohibitively huge investments required in the classical approach using aqueous reprocessing and sodium cooled fast reactors.

It is demonstrated that the identified and investigated sodium chloride – uranium chloride based systems are feasible to deliver the requested breeding based plutonium as the driver fed to a core with a radius between 170 and 185 cm in the used 2D model as well as based on enriched uranium requiring a system radius of 200cm in the 2D system. This result is enriched by the analysis of the achievable full power days per inserted mass of plutonium ~400 days in the 120 cm system, but more than 550 days in the 200 cm system. This results lead to the outcome that larger systems are much more efficient in the Pu use, even if a higher amount of plutonium will be required for the start-up of the system. This essential insight supports evidence based, multi-disciplinary innovative reactor optimization of future molten salt reactors and leads to a first, very important conclusion that systems with lower power density could be very attractive in the case of low fuel cost as it is in the case for spent nuclear fuel operated systems. This is supported by the analysis of the consequence of the use of natural NaCl as the carrier salt instead of the often discussed NaCl-37, which would lead to a significant reduction in cost and complexity of the fuel production.

The general outcome is in strong contrast to the design aims of traditional sodium cooled fast reactors and many current molten salt fast reactor proposals, where the designers head for high power densities due to the high cost of the use of mixed oxide fuel in SFRs. Based on these insights a more sophisticated study considering advanced salt clean-up and a more realistic reactor operation scheme should be envisaged. However, this kind of evaluation will require a new approach in modelling and simulation with more adequate tools to tackle the multi-disciplinary challenges of the interaction between reactor operation, breeding, fission product accumulation and salt clean-up.

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Appendix A

Table A1. Overview on the calculated cases.

Studied cases	Radius (m)	Pu Content Atomic %	Initial Keff	Fissile Content Atomic %	Power (W/gHM)	Sphere Volume (m^3)	Salt Mass (tons)	HM Mass (tons)	Fissile Mass (tons)	Power (MW)
Uranium cases	1.2	0	1.06456	19.68	100.00	7.24	23.16	12.39	2.44	1238.8
	1.45	0	1.04348	16.53	56.68	12.77	40.86	21.86	3.61	1238.8
	1.7	0	1.02976	14.73	35.17	20.58	65.85	35.22	5.19	1238.8
Two component Pu cases	1.2	8.00	1.04298	16.28	100.00	7.24	23.16	12.82	2.09	1238.8
	1.45	6.89	1.0231	14.26	56.68	12.77	40.86	22.51	3.21	1238.8
	1.7	6.13	1.00097	12.87	35.17	20.58	65.85	36.17	4.65	1238.8
	1.85	5.81	0.99072	12.27	27.29	26.52	84.87	46.56	5.71	1238.8
	2	5.54	0.98018	11.75	21.60	33.51	107.23	58.77	6.91	1238.8
Three component Pu cases	1.2	7.86	1.03307	13.20	94.01	7.24	23.16	13.18	1.74	1238.7
	1.45	6.80	1.01583	12.71	53.28	12.77	40.86	23.25	2.96	1238.6
	1.7	6.13	1.00061	11.45	33.06	20.58	65.85	37.48	4.29	1238.7
Three component U cases	2	0	1.00497	12.3	20.31	33.51	107.23	57.35	7.05	1238.8
	2.2	0	0.99827	11.7	15.26	44.60	142.73	76.34	8.93	1238.8

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