applied sciences

MDPI

Article

On the Dimensions Required for a Molten Salt Zero Power Reactor Operating on Chloride Salts

Bruno Merk [1,*], Anna Detkina [1], Seddon Atkinson [1], Dzianis Litskevich [1] and Gregory Cartland-Glover [2,3]

1 School of Engineering, University of Liverpool, Liverpool L69 3GH, UK; a.detkina@liverpool.ac.uk (A.D.); Seddon.Atkinson@liverpool.ac.uk (S.A.); d.litskevich@liverpool.ac.uk (D.L.)
2 Scientific Computing Department, Science and Technology Facilities Council, Cheshire WA4 4AD, UK; greg.glover@stfc.ac.uk
3 Daresbury Laboratory, SciTech Daresbury, Cheshire WA4 4AD, UK
* Correspondence: b.merk@liverpool.ac.uk

Abstract: Molten salt reactors have gained substantial interest in the last years due to their flexibility and their potential for simplified closed fuel cycle operation for massive expansion in low-carbon electricity production, which will be required for a future net-zero society. The importance of a zero-power reactor for the process of developing a new, innovative rector concept, such as that required for the molten salt fast reactor based on iMAGINE technology, which operates directly on spent nuclear fuel, is described here. It is based on historical developments as well as the current demand for experimental results and key factors that are relevant to the success of the next step in the development process of all innovative reactor types. In the systematic modelling and simulation of a zero-power molten salt reactor, the radius and the feedback effects are studied for a eutectic based system, while a heavy metal rich chloride-based system are studied depending on the uranium enrichment accompanied with the effects on neutron flux spectrum and spatial distribution. These results are used to support the relevant decision for the narrowing down of the configurations supported by considerations on cost and proliferation for the follow up 3-D analysis. The results provide for the first time a systematic modelling and simulation approach for a new reactor physics experiment for an advanced technology. The expected core volumes for these configurations have been studied using multi-group and continuous energy Monte-Carlo simulations identifying the 35% enriched systems as the most attractive. This finally leads to the choice of heavy metal rich compositions with 35% enrichment as the reference system for future studies of the next steps in the zero power reactor investigation. An alternative could be the eutectic system in the case the increased core diameter is manageable. The inter-comparison of the different applied codes and approaches available in the SCALE package has delivered a very good agreement between the results, creating trust into the developed and used models and methods.

Keywords: nuclear; nuclear reactors; reactor physics; nuclear experiments; zero-power reactors; modelling and simulation; molten salt reactors

Citation: Merk, B.; Detkina, A.; Atkinson, S.; Litskevich, D.; Cartland-Glover, G. On the Dimensions Required for a Molten Salt Zero Power Reactor Operating on Chloride Salts. *Appl. Sci.* **2021**, *11*, 6673. https://doi.org/10.3390/app11156673

Academic Editor: Guan Heng Yeoh

Received: 22 June 2021
Accepted: 16 July 2021
Published: 21 July 2021

1. Introduction

Historically, zero or very low power experiments have been seen as the first step into a new reactor programme or a new technology isotopic composition of the leftover, I fear that it is even not possible to operate a fast reactor based on this material. This is a problem which has already been observed for high Pu loads in the CAPRA core even if this was a fast reactor design. The degradation of the Pu vector in the thermal system is much more pronounced at it will not help to do P&T when 70% of the Pu, but 95% of the fissile is burnt. The leftover is still highly radiotoxic and the challenge to solve this problem is larger than before starting the process. [1]. The key requirements were to test new configurations in safe settings and to use the opportunity of these highly flexible experiments to assure effective learning at the beginning of a new technology.

Figure 1 shows the timeline of the MAGNOX development with several zero or very low power facilities: Gleep in 1947, BEPO in 1948, to Windscale-1 in 1952 at the beginning of the UK nuclear programme. At that early point of development, the major arguments for the zero power experiments were the comparably low cost and the opportunity of flexible, well-instrumented tests to demonstrate and improve the understanding of the system behaviour, to support the theoretical models, and to investigate a multitude of promising solutions in a short time period, promising rapid and effective learning. More recent ideas demonstrate that building a zero-power reactor offers a multi-fold opportunity to support the start-up of an innovative reactor program [2].

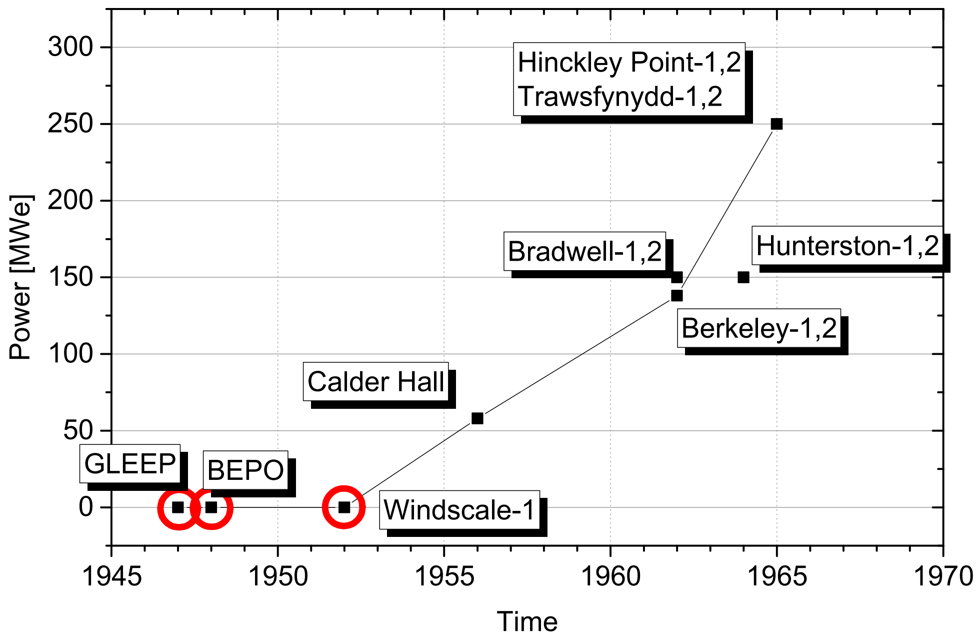

Figure 1. Development timeline of the MAGNOX technology following the data of [1].

At a first glance, zero power experiments have lost this high priority in more modern times, e.g., when observing the timeline of the German HTR programme [3] starting with the highly successful AVR experiment. However, looking deeper into the development of the programme, the importance of zero power experiments is even more highlighted due to the decision for the demand of a zero-power facility as basis to provide more hands on testing, leading to the KAHTER facility in 1973 [3]. The developers of the HTR programme confirmed that zero power tests have been essential for the optimization of the system and the confirmation of theoretical models [4]. Nowadays, scientists claim that it is possible to design a new reactor based on modelling and simulation only, which may be true for a known technology like light water reactors with a very broad validation base and a wide variety of specially developed codes. However, for any really new technology, these specialized modelling and simulation tools either do not exist or there is a strong demand for code validation on experimental results either demanded by the developers or in the next stage by the regulator in the design assessment process. Typically, validations are required for tools relevant for the determination of:

- core criticality and reactivity effects
- neutron flux distribution in space and energy as well as the resulting power distribution
- changes in reactivity and neutron flux resulting from density and temperature changes

where zero power experiments can deliver the first validation level. This first level of experiments is the basis for any next step in the process of developing a new, innovative reactor system, as given in Figure 2, where the validation request is only a part of the reasoning for the zero power experiment, but the reasoning also includes:

- the formation of a team of specialists who are able to develop the project

- the development and production of the first key components, e.g., the fuel
- the establishment of a supply chain
- the close interaction with the regulator to get the experiment licensed [3]

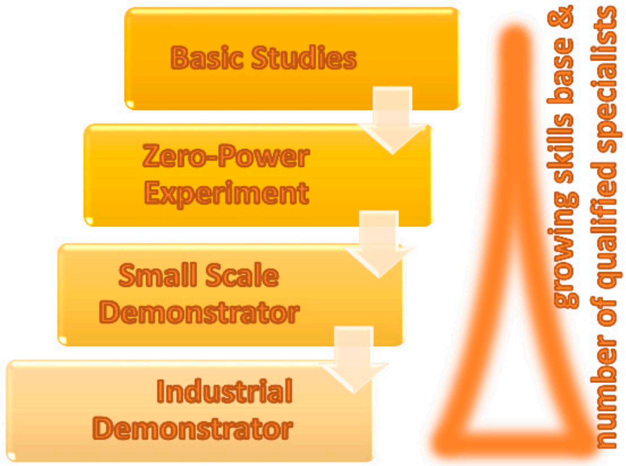

Figure 2. Process steps for the development of a new, innovative reactor system following the approach of [3].

The typical zero power experiments are almost kind of impossible to implement in power producing systems with high operational temperature and high neutron flux as well as the levels of radiation due to fission products. In contrast to this, a major point in the zero power experiments is the significantly larger operational envelope of a zero-power facility and the higher flexibility than in power producing systems, while not requiring an energy removal system due to low or at least moderate operational temperatures and very low power. The low flux and very low power levels allows comparably easy access for the experimenters due to the reduced radiation level caused by the absence of large amounts of fission products. These properties widen the space of available instrumentation significantly and allow rapid changes of the setup to investigate different configurations. The properties also allow for almost immediate accessibility to the experiments after shut down and assure extremely fast take-up of a relevant set of experimental results.

Following these arguments, the last, newly established technology, accelerator driven systems, and lead cooled reactors have been accompanied by two different zero power experiments, YALINA [5,6] and GUINEVERE [7,8], to create an understanding of the interaction between the accelerator and the sub-critical core as well as to deliver validation for lead-cooled reactor systems. Looking into molten salt reactor technologies, completely new approaches for operating reactors directly on spent fuel have come up through the proposed iMAGINE technology [3,9], and many companies like TERRESTRIAL Energy [10], TERRAPOWER [11], Elysium Industries [12], and others are planning reactors which will have demands for code and design validations. However, due to the homogeneous core composition (unity of coolant and fuel) of a molten salt reactor, a zero power reactor experiment is significantly different to existing, heterogeneous experiments. Thus, novel approaches for design, operation, control, and experimental setup must be scoped out, developed, and invented beginning with this series of publications. Meanwhile, Russia has already announced plans to develop their own research reference facility for their partitioning and transmutation molten salt reactor programme [13] and Terrapower has announced their approach using a Molten Chloride Reactor Experiment with low power (300 kW) to be located at the Idaho National Laboratory [14].

The general choice for the NaCl-UCl salt system has already been investigated in earlier works [15,16]. The major point for this decision is on the good potential for breeding (high heavy metal loading in the eutectic), which is required for the envisaged long-term self-sustained operation based on spent nuclear fuel, which is the core of the iMAGINE technology. The focus of this publication will be on answering the questions on the salt compositions (i.e., potential enrichment and heavy metal loading) and the dimensions required of a potential very low power experimental setup. This will narrow down the choices for a zero power experiment for a molten salt fast reactor based on a multitude of modelling and simulation results using different tools of the SCALE package. The study will focus on balancing the following, partly contradicting design objectives for a zero-power reactor experiment:

- Reasonably small core size—reduction of the fuel volume, thus production cost
- Reasonably low enrichment—reduction of fuel production cost, safeguarding concerns, criticality issues in fuel production and handling, but most of all being close to a future iMAGINE system with lower enrichment and/or fissile loading with a correspondingly larger core
- Reasonably low power—avoiding heat extraction while having sufficiently high flux to obtain acceptable measurement time and detector statistics
- Reasonably undisturbed flux distribution—keeping the flux distribution as close as possible to an undisturbed flux distribution of a potential future large scale core
- Reasonable core geometry—avoiding limitations to potential control system approaches due to the core geometry
- Potential operational safety—assure sufficiently strong feedback that the system is self-limiting without heat removal

The work started by providing a description of the codes, methods, and the salt configurations. This was followed by a 2-D analysis of the system radius and the reflector effect, depending on the salt composition and the uranium enrichment to get a first insight. This will be supported by studying the effect of the enrichment on the thermal feedback effect, which will allow the down selection of enrichment with which the zero power reactor can operate. This will be followed by an analysis of the integral of the chosen system and an investigation of the space-energy neutron flux distribution. The study will be finished by a 3-D analysis of the core volume required to achieve a critical operating condition for the down selected systems. The results of the study will be cross-verified using the different codes and calculation methods available in the SCALE package.

2. Codes and General Modelling

The salt system is based on detailed data on the density versus temperature curve from Russian literature [17]; see Table 1. The data is used for the analysis based on a reference temperature of 980 K with changes of ±50 K for the analysis of feedback effects with the aim to stay in the validity range of the available data given below for the reference case, as well as for the feedback analysis. Nevertheless, while being a good starting point for an initial analysis, the data is limited to a very narrow temperature window and for a temperature range which will not be the first choice for initial experiments in a zero-power environment. Especially, for a detailed study of a future zero-power experiment, the temperature range has to be extended down to room temperature to be able to judge the system behaviour under cold, solid conditions, which would be the perfect setting for first experiments. However, to be able to provide modelling and simulation results for room temperature, reliable thermo-physical data for the whole temperature range must be measured for the required salt system. This will require the creation of the salt mix in the molten state and then cooling it back down to room temperature.

Table 1. Concentration dependent densities for the system $NaCl-UCl_3-UCl_4$ with the uranium salt composition 30% UCl_3 + 70% UCl_4 and varying NaCl concentrations part reprinted from [1]. The experimental uncertainty in the density measurements is given with max ± 1.5%.

Concentration % NaCl	Validity Range [K]	A	b 10^{-3}	Standard Deviation 10^3
20.0	934–1018	5.3995	1.8646	4
30.0	934–1020	4.9360	1.5276	2
45.0	939–1029	4.2368	1.0256	3
60.0	872–1037	3.8237	0.8774	2
80.0	985–1119	3.2382	0.8012	2

Using the coefficients given in Table 1, the temperature dependent density is calculated according to the formula:

$$\rho\left[\frac{\mathrm{g}}{\mathrm{cm}^3}\right] = a - bT[\mathrm{K}] \tag{1}$$

Based on the given salt system investigation [15], the applied standard salt composition for the study for the nominal case is the eutectic 42.5% NaCl–40.5% UCl_4–17% UCl_3. This composition is close enough to the third row of Table 1 to provide a reasonable density estimation. In addition, a heavy metal rich case is investigated with 20% NaCl–56.35% UCl_4–23.65% UCl_3. The fuel temperature for both compositions is set with a uniform spatial distribution to 980 K in the core since no variation is expected in a zero-power system that typically has a thermal power of less than 1 kW.

The simulations for this initial study of zero-power reactor configurations have been performed using the POLARIS module of the SCALE code system [18]. Polaris is a new module for SCALE 6.2 that provides a 2-D 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, validation we have recently published [16] was an initial verification/validation against the Monte-Carlo code SERPENT [19]. This will be extended here by comparisons to the multi-group deterministic S_n transport calculation sequence TRITON-NEWT and multi-group as well as continuous energy Monte-Carlo code Keno VI, all parts of the SCALE package [20].

Polaris uses the v7-252 cross-section set of the SCALE package, based on ENDF/B 7.1, which is also the basis for the TRITON-NEWT as well as the multi-group (MG) Keno VI calculations, while the continuous energy version uses the ce_v7.1_endf library of the SCALE package.

For the study, very specific 2-D and 3-D models of a fast molten salt reactor were built for the simulation. First, the POLARIS model, see Figure 3, consists of 2-D discretized ring core surrounded by a steel vessel and a reflector region with a large amount of absorbing material around the core. The outer region is required due to the reflecting boundary conditions at the outside of the square (a given setting in POLARIS). The arrangement models a leakage boundary condition due to the absorber. Second, the NEWT model, reflects a 2-D quarter of the core with x-y discretization consisting of the ring core, the steel vessel surrounded by the reflector, while the rest of the cell is filled with vacuum using reflective boundaries at the bottom and the right side and vacuum boundaries at the other two sides; see Figure 4. Third, the 2-D and 3-D Keno VI models are given in Figure 5. The 2-D model is based on a cylindrical core surrounded by a vessel, a reflector, and vacuum boundary conditions (pictured left as a horizontal cut). In the vertical cut (central picture), the system has reflecting boundary conditions at the top and bottom to model the system as a 2-D problem. The 3-D model is a finite cylinder (vertical cut, pictured right) with a reflector on the top and bottom and vacuum boundary conditions. The aim for the boundary condition setting for all three models is to come as close as possible to reality with full leakage of all neutrons from the outside of the reflector.

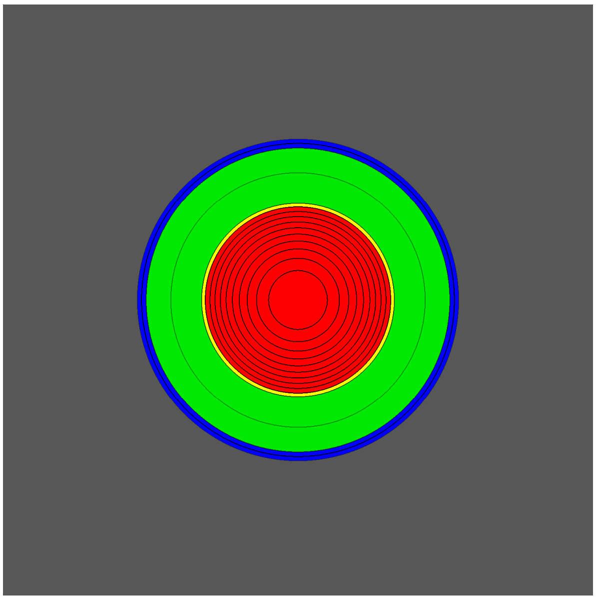

Figure 3. The 2-D SCALE/POLARIS model (molten salt fuel core—red, stainless steel—yellow, NaCl reflector—light green, vacuum—blue, absorber material—dark grey) used for the general analysis.

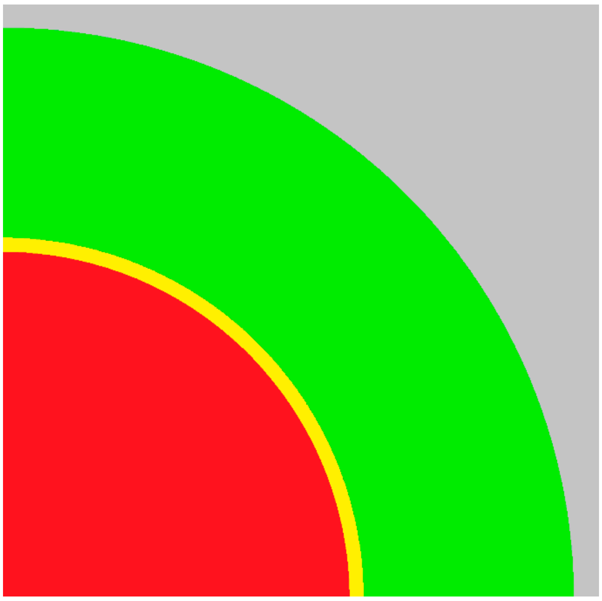

Figure 4. The 2-D SCALE/TRITON-NEWT model (molten salt fuel core—red, stainless steel—yellow, NaCl reflector—light green, vacuum—grey) used for the analysis of the neutron flux distribution.

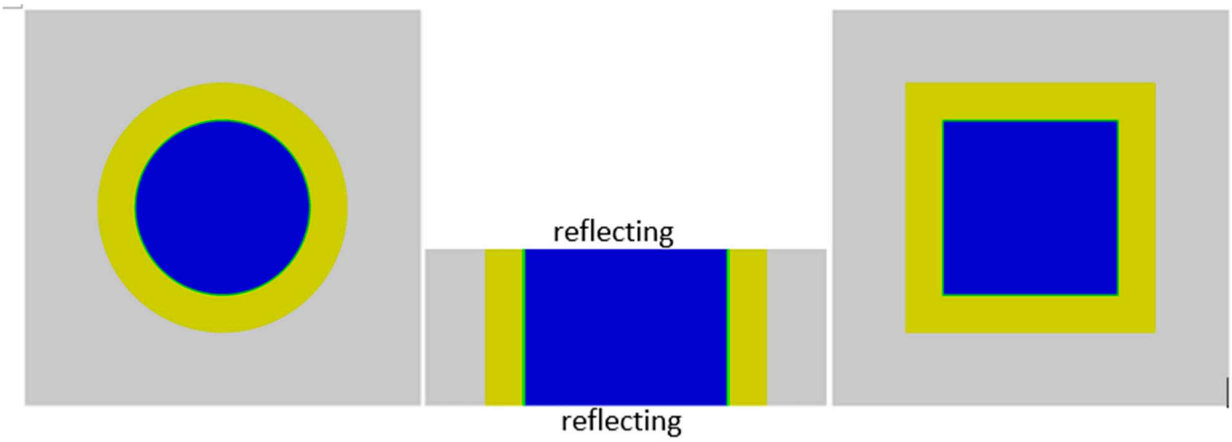

Figure 5. The general 2-D (left and centre) Monte-Carlo model of the configuration (left, top view x-y, centre front view x-z) and 3-D (left and right) Monte-Carlo model of the configuration (left, top view x-y, right front view x-z) with molten salt fuel core—blue, stainless steel vessel—light green, reflector—yellow, and vacuum—grey used for the validation of the results and the 3-D studies.

The limit for the dimensional iterations in POLARIS, NEWT, and KENO VI was set to ±100 pcm for the determination of the critical dimension. For the Monte-Carlo simulations, a setting of 1000 generations with 10,000 particles each was used for the iteration and final evaluation of 5000 generations with 50,000 particles have been used. This setting leads to an accuracy of ±12 pcm and below at a 95% confidence interval, which is more than sufficient in comparison to the spatial dimension iteration limit.

3. Results and Discussion

3.1. 2-D Criticality Versus Dimension Studies

The first important information for designing a small scale zero-power core is the demanded size, the required salt amount, and the related amount of nuclear material. The required core size is studied through the variation of the enrichment, followed by the determination of the radius required to achieve a critical configuration in the 2-D system with an iteration accuracy of ±100 pcm. The thickness of the reflector has been kept identical at 30 cm for each step of the study. The mentioned size of the core is strongly dependent on the salt composition—eutectic composition versus heavy metal rich composition, and on the fissile enrichment in the uranium chloride salt. The system radius and thus the required amount of salt is, as expected, to be continuously decreasing with increasing U-235 enrichment showing a kind of asymptotic behaviour for very high enrichment; see Figure 6.

The observation holds for the reflected and the unreflected system (modelled by replacing the reflector material with a vacuum) as well as for the eutectic and the heavy metal rich system. The volume gain

$$volume\ gain = \frac{vol_{HMR} - vol_{ET}}{vol_{HMR}}\ [\%]$$

for the heavy metal rich (HMR) system stays almost constant with a volumetric reduction of approximately 30% compared to the eutectic system (ET), while for the 20% enriched system, the volume gain is slightly higher. The slight variation in the curves seem to be a result of the iteration limit for the dimensional iteration (±100 pcm). From the point of the required salt volume, a good gain can be achieved by increasing the U-235 enrichment to 35%—thus, a value higher than the safeguarding limit of 20%. The effect of going to higher values does not seem to give the required significant improvement to justify moving to a higher enriched fissile material.

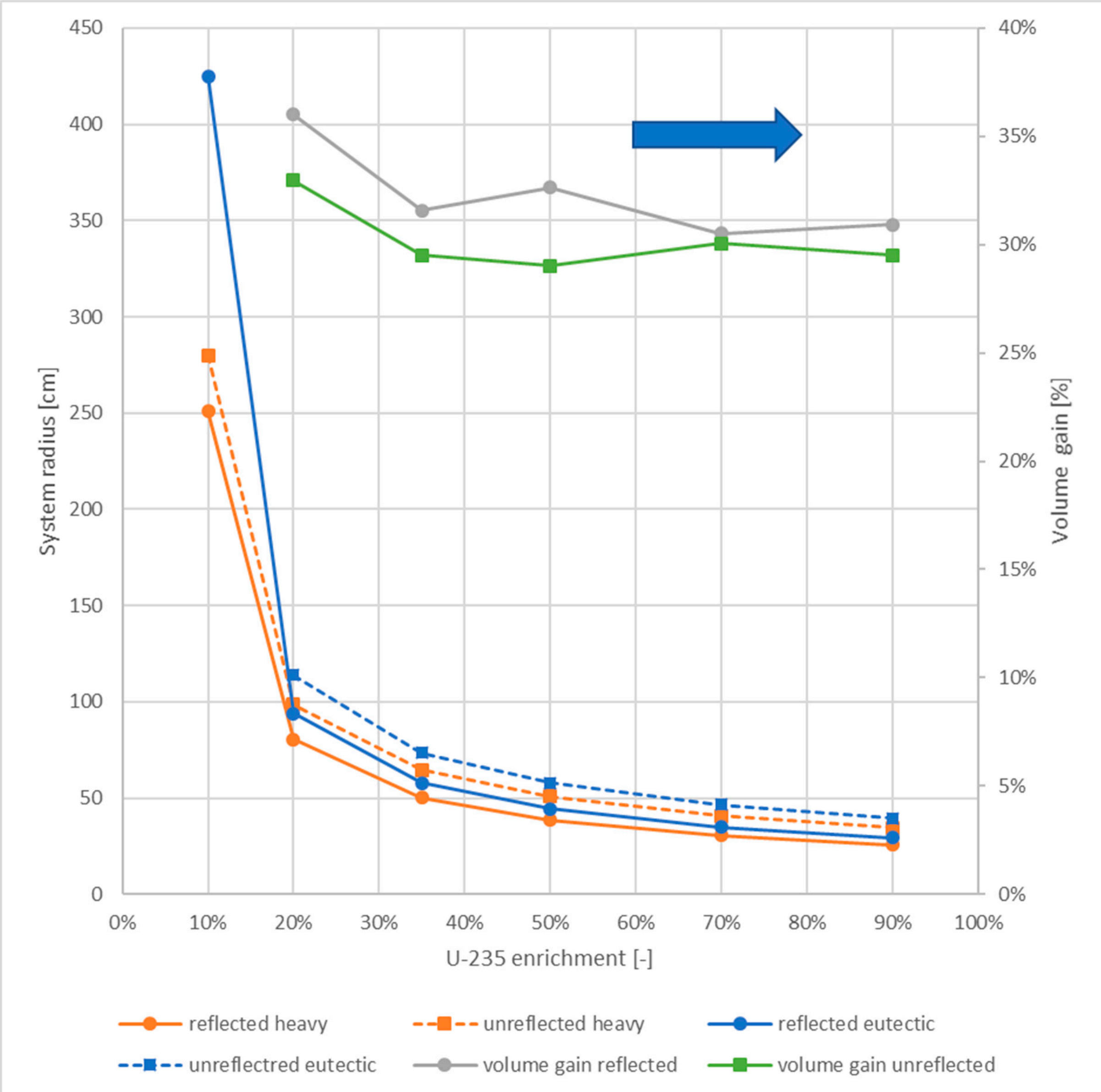

Figure 6. 2-D system radius for the eutectic and the heavy metal rich system depending on uranium enrichment for the reflected and the unreflected systems as well as the volume gain per step archived by changing from the eutectic to the heavy metal rich system (calculated through POLARIS).

The second step of the study analyses the efficiency of the reflector dependent on the U-235 enrichment and the two different salt configurations (Figure 7). For this step, all reflected critical configurations of the last step have been recalculated with the reflector region filled with vacuum, while the thickness of the reflector itself has been kept identical at 30 cm for each step. The figure indicates a clear change in the effect of the reflector with the expected growing gain caused by the reflector effect with increasing U-235 enrichment and the correlated decrease of the size of the critical system. The k_{eff} of the drop caused by eliminating the reflector reaches up to more than 20,000 pcm for the highest enrichment, while the effect starts to saturate at high enrichments. The effect of removing the reflector is slightly stronger for the heavy metal rich system for lower enrichments, while this difference disappears for higher enrichment. The results of this step indicate a good opportunity to use the reflector as a future control system for a zero-power reactor in the case the enrichment is sufficiently high, at least 35% or higher. The control- and shut-down system will be discussed and analysed deeper in a second part of this study [21] and the effects of the use of different reflector materials in a third part [22].

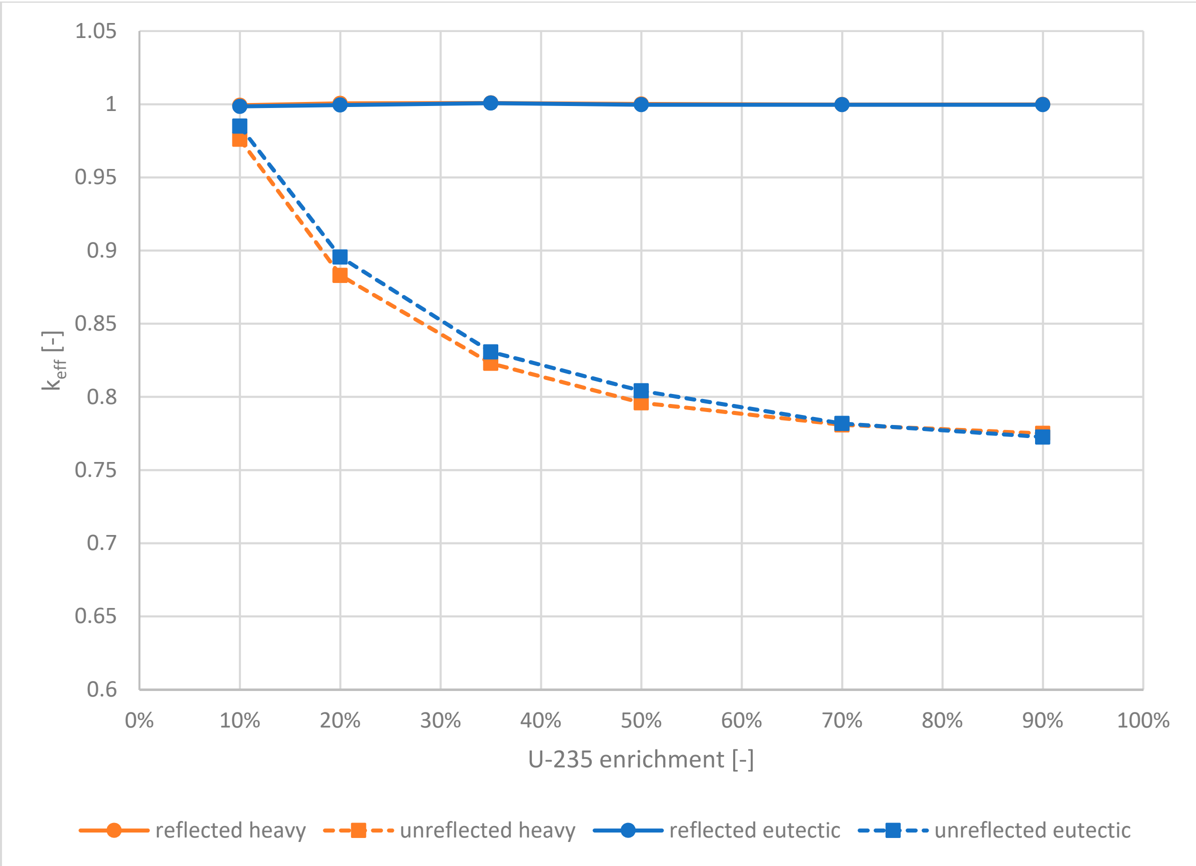

Figure 7. 2-D system criticality reduction by eliminating the reflector for the eutectic and the heavy metal rich system depending on uranium enrichment (calculated through POLARIS).

3.2. Thermal Feedback Study

Another important feature for the control of the reactor is its stability due to the inherent temperature feedback effect. This feedback effect is combined from two major components in a molten salt reactor, one is the density change of the molten salt which acts in unity as coolant as well as fuel. The other effect is the Doppler or fuel temperature effect, which leads to a higher level of neutron absorption in the reactor fuel due to resonance broadening. The feedback effect is determined through the change in density using Equation (1) to provide new number densities combined with the change of the temperature of the molten salt applied for the cross-section correction due to the Doppler effect. In general, molten salt reactors possess a very strong negative feedback behaviour. The effect of the density changes in the salt leads to a reduction of the density of fissile atoms in the salt volume of the core. Thus, the criticality will be strongly reduced when the salt expands, while the classical Doppler effect is in a comparable range to other reactors operating over a comparable temperature level. However, there are two things which are of interest: How much dependence does the temperature feedback effect show with regards to the variation of the uranium enrichment and thus the core dimension? The second point would be the feedback effects at low operational temperature around room temperature with solid salt, but the density over temperature data is currently not available for this high importance temperature window.

The temperature feedback increases by increasing U-235 enrichment, but it shows the same style of saturation as already seen in the dimensional study and the reflector study.

Thus, the effect of increasing the enrichment is stronger at lower enrichment than for very high enrichment; see Figure 8. In general, the effect grows for ~−3 pcm/°C to −20 pcm/°C for the eutectic system and ~−7.5 pcm/°C to −34 pcm/°C for the heavy metal rich system. The growing effect due to the increased enrichment as well as for the heavy metal rich case can be explained through the higher amount of fissile material, which is pushed out of the core in the case of a reduction in density due to temperature increase.

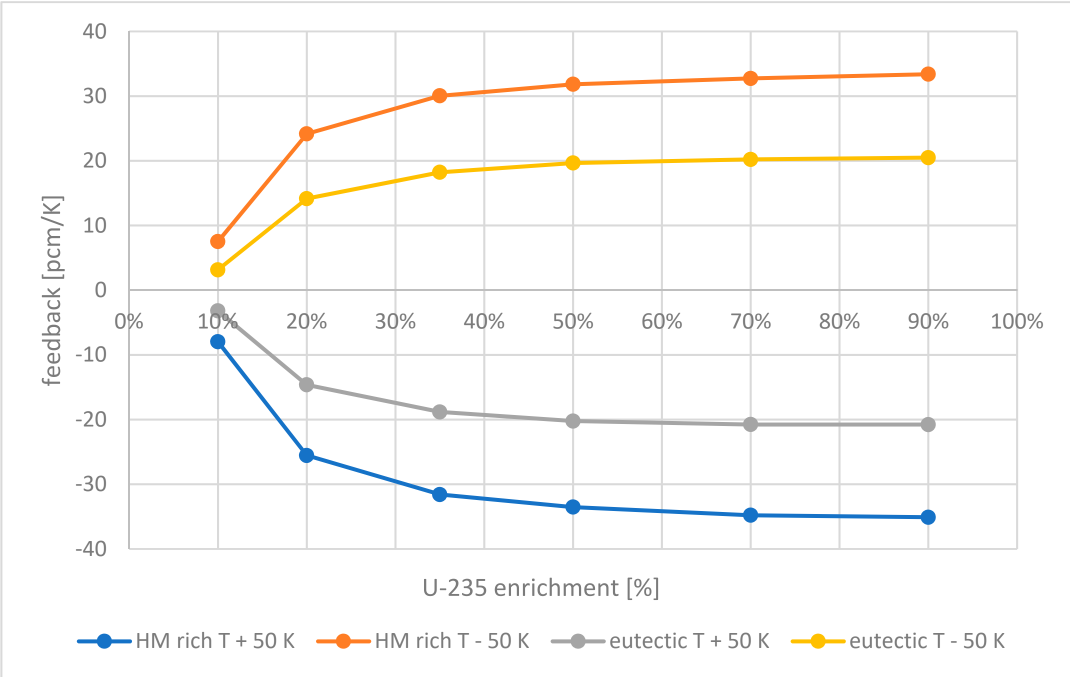

Figure 8. 2-D system thermal feedback effects (salt density and fuel temperature) for the eutectic and the heavy metal rich system depending on uranium enrichment (calculated through POLARIS).

3.3. 2-D Integral Neutron Spectrum Studies

The next part shows an investigation of the influence of different system specifications: heavy metal content of the salt, use of the reflector, and enrichment on the neutron spectrum in the system. The general integral neutron spectrum (integrated over the whole system) in the NaCl-UCl system is highly comparable with a fast reactor, where almost no neutron flux below 100 eV is observed and there is a clear peak between 200 keV and 1 MeV. The spectrum is slightly harder than in a classical sodium cooled fast reactor fuel assembly of EFR type [23]; see the grey line in Figure 9. The molten salt reactor spectrum is characterized by a much smaller, but still visible, low energy tail formed by the sodium with a clearly lower number of neutrons below 100 keV and a higher number of neutrons above this value.

The change from the eutectic to the heavy metal rich system leads to a slightly harder neutron spectrum with a decrease in the number of neutrons below ~100 keV and a slight increase in the number of neutrons around 1 MeV; see Figure 9. However, the change in the neutron spectrum is only very limited; the general spectral distribution stays almost constant.

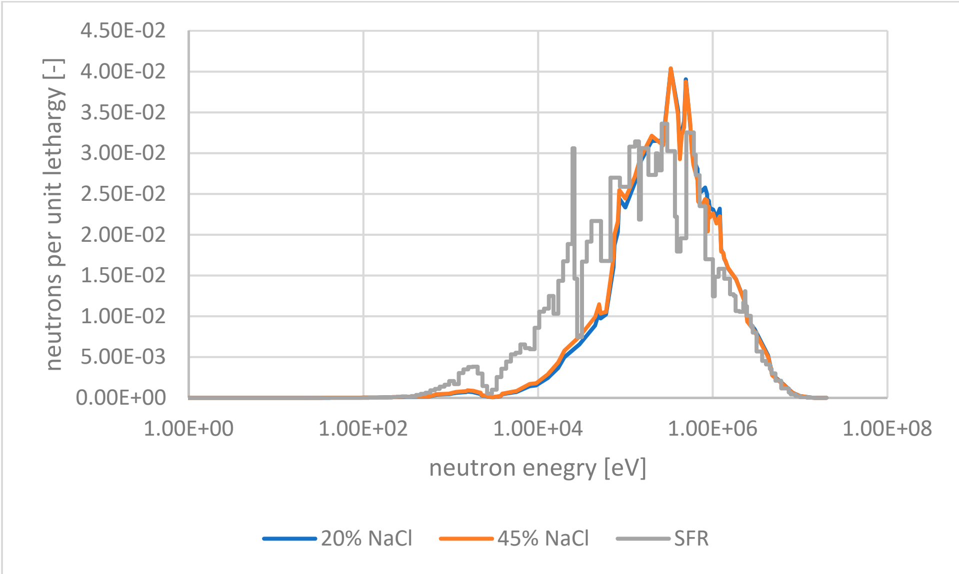

Figure 9. Integral neutron spectrum 35% enriched for the eutectic and the heavy metal rich system in comparison with the neutron spectrum in a classical sodium cooled fast reactor (SFR) fuel assembly of EFR type (calculated through POLARIS, the SFR case is based on HELIOS [24]).

The influence of the NaCl reflector on the neutron flux spectrum is much stronger than the influence of the salt composition; see Figure 10. The reflector creates a new peak around 1 keV, which disappears when the reflector is removed. This low energy peak reflects neutrons that are undergoing several collisions without leading to neutron reactions, which happens mainly in the reflector. Besides this, the neutron spectrum of the system is clearly softened by the reflector creating a higher neutron flux below ~200 keV and a lower neutron flux around 1 MeV.

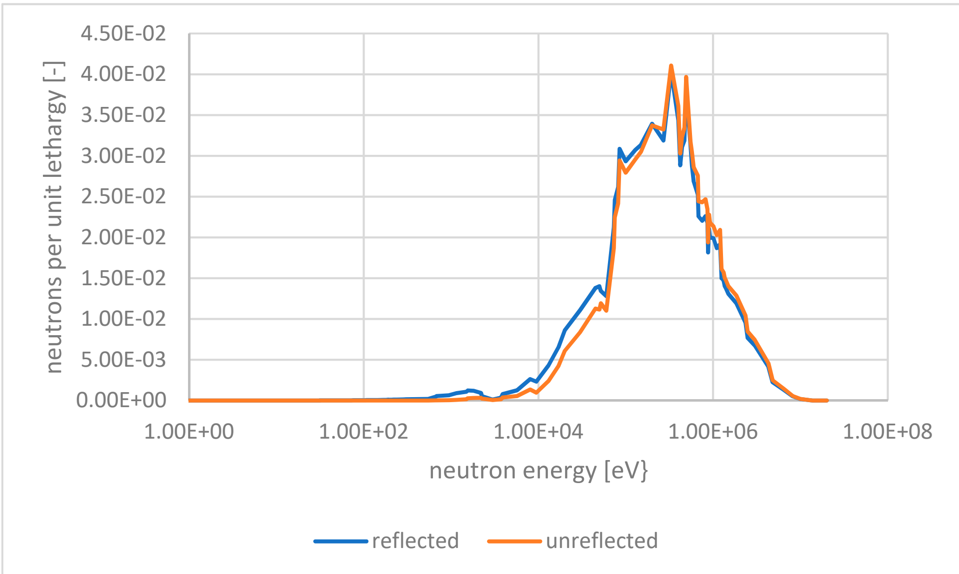

Figure 10. Neutron spectrum 20% enriched for the reflected and reflected eutectic system (calculated through POLARIS).

The strongest change in the neutron spectrum is caused by the change in the U-235 enrichment; see Figure 11. The use of highly enriched uranium leads to a clear reduction in system size; see Figure 6. The small system size leads, on the one hand, to a significantly higher neutron leakage and less collisions, while on the other hand, the strong reduction of the U-238 amount diminishes the parasitic absorption of neutrons in the system since most of the heavy metal is in the 90% enriched system fissile material. Overall, these changes lead to a decrease in the number of neutrons in the energy range below 200 keV and an increase of the number of neutrons above 500 keV. The neutron spectrum of the 90% enriched system is much closer to the fission neutron spectrum.

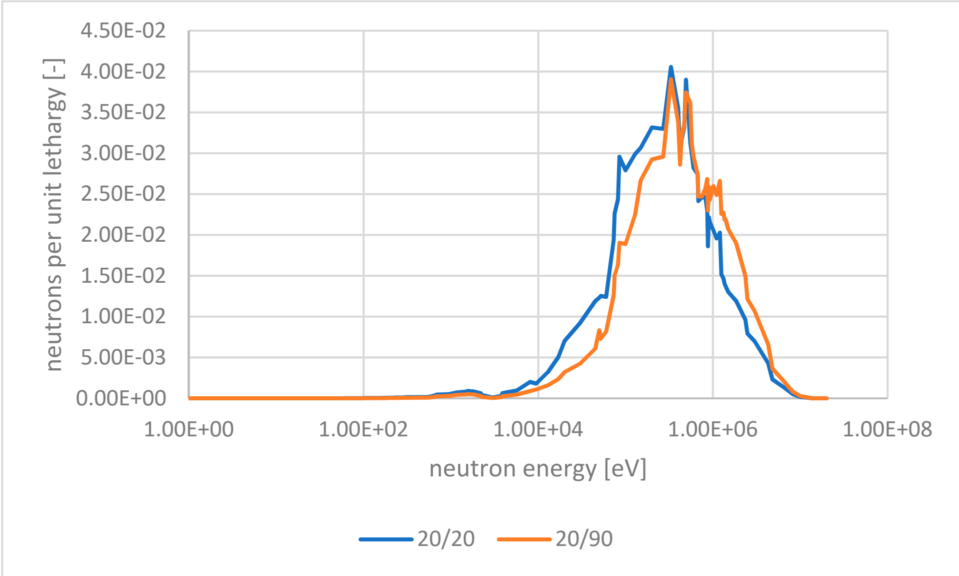

Figure 11. Neutron spectrum for the heavy metal rich system (20% NaCl) with 20% enrichment (20/20) compared to the 90% enriched system (20/90) (calculated through POLARIS).

3.4. Verification of the 2-D Results through Different Codes

All results presented and discussed up to now have been based on the described POLARIS model and the application of POLARIS, a code that was developed originally for lattice calculations for light water reactors modelling infinite grids of unit cells and fuel assemblies by applying reflective boundary conditions. This effect has been eliminated by a very elaborate model using a large amount of absorber. The reliability of the results using this model and the POLARIS code is investigated in Table 2. The aim of this comparison is to assure that the results of POLARIS are robust regarding the used model, the method, and the cross-section basis. The method of the characteristic model is evaluated against another transport code by applying the S_n method on an unstructured mesh using the TRITON-NEWT sequence and the identical cross section set, but with another procedure for the self-shielding. The results show a good agreement too within one centimetre in core radius for all the three tested cases with POLARIS being for all cases on the conservative side. The quality of the two deterministic results is evaluated by a comparison to the results gained with the Monte-Carlo code Keno VI of the SCALE package using the multi-group self-shielded cross-section set produced with the identical procedure, as for the deterministic NEWT transport solver. The Keno VI multi-group results show only a very small deviation (less than 1.5 cm in radius) to the NEWT results and to the POLARIS results—thus, the results of both significantly different transport methods agree very well. Finally, the last approximation, the use of the multi group library relying on the 252-group master library traditionally weighted for light water reactor application, is investigated. This task is accomplished by a comparison to the application of the Keno VI code using a continuous energy approach based on the ENDF-B VII.1 library to evaluate the potential bias of the multi-group master library. The Keno VI continuous energy results once more coincide very well with all the other results; thus, all results stay in less than ±1 cm from the highest level as well as highest complexity results provided by the continuous energy Monte-Carlo calculation, which confirms the quality of the significantly faster operating models using a higher level of approximation. The quality of all results is definitively sufficient for this kind of first step study for the analysis of the potential dimensions of a zero-power chloride-based molten salt reactor experiment.

Table 2. Comparison and evaluation of different relevant results for the expected core radius of the 2-D system using deterministic and stochastic simulation methods based on multi-group and continuous cross-section sets.

	42.5% NaCl	**20% NaCl**	**20% NaCl**
	35% enr	**20% enr**	**35% enr**
Scale/Polaris core radius [cm]	**57.7**	**80.6**	**50.3**
Scale/Trition/NEWT core radius [cm]	56.9	80.5	49.3
Scale/Keno VI multi-group core radius [cm]	56.3	79.63	49.1
Scale/Keno VI continuous core radius [cm]	56.84	80.26	49.54

3.5. 2-D Neutron Flux Distribution Analysis

After the test on the reliability of the different codes, a first glance will be taken into the spatial effects on the spectrum caused via the third dimension leakage. This can be evaluated through the TRITON-NEWT sequence and the spatial distribution of the neutrons of a set of arbitrarily defined neutron groups, while trying to reflect on the characteristic for the neutron spectrum of the system. The comparison of the different settings for the height of the core for the spectral analysis is given in Figure 12. The spectrum of the 2-D system without leakage in axial direction (labelled inf med) is the softest with the highest amount of lower energy neutrons. As soon as an axial leakage is taken into account (labelled crit flux 120 and 180), the spectrum starts to harden with decreasing axial dimensions. However, the strongest hardening of the spectrum appears to happen in the case when the radial reflector is eliminated for the infinite medium core (labelled unreflect); see grey line in Figure 12. In addition to the spectral information, the energy groups for the 2-D spatial analysis of few group neutron flux distribution are given in this figure with the energy groups I to V separated by the blue vertical lines to get a higher fidelity of the analysis of the space-energy neutron flux distribution, which will allow understanding to be gained of where each of the groups have their highs and lows in the spatial distribution.

The group wise 2-D spatial neutron flux distribution for the extra5 group scheme (trying to capture the major characteristic of the neutron flux profile), defined in Figure 12, is shown in Figure 13. In the three higher energy groups, the highest neutron flux (red) is concentrated in an inner core ring (see mark at 40 cm radius), while the rest of the core up to 57 cm radius is covered by lower flux with intermediate intensity. All three groups have almost the same intensity of neutron flux; see scale at bottom, right. While the neutron flux in the reflector is already very low in group one (left, top), there is an increased flux visible in the reflector in group 2 (top, right), which is becoming stronger in group 3 (centre left). Thus, it becomes apparent that the flux distribution flattens out with lower energies. Group 4 has an overall much lower intensity and an almost flat profile from the centre until about half of the reflector, followed by a rather sharp decrease in the outer half of the reflector. Group 5 has an overall very low intensity with a peak in the reflector and a small neutron flux increase at the outer boundary of the core, which indicated that the lower energy neutrons slowed down in the reflector penetrate only to a small depth into the core due to the self-shielding caused by the high importance of the low energy neutrons. This undesired effect creates a slight increase in the power production close to the reflector and thus an undesired power and flux distribution cannot be completely avoided, but in the ideal case limited through the choice of the reflector. However, in this case, the effect is expected to be of limited influence due to the low over all intensity. Nevertheless, this behaviour should be kept in mind when investigating different reflector materials since the use of more efficient reflector materials like polyethylene or graphite could strongly enhance this undesired effect due to the high neutron thermalization power of these materials. A comparable effect has been discovered in the YALINA-Booster experiments where a fast system was surrounded by a system thermalized by polyethylene. This has led to problematic behaviour in the time evolution of the neutron flux and has complicated the analysis of the experiments significantly [25].

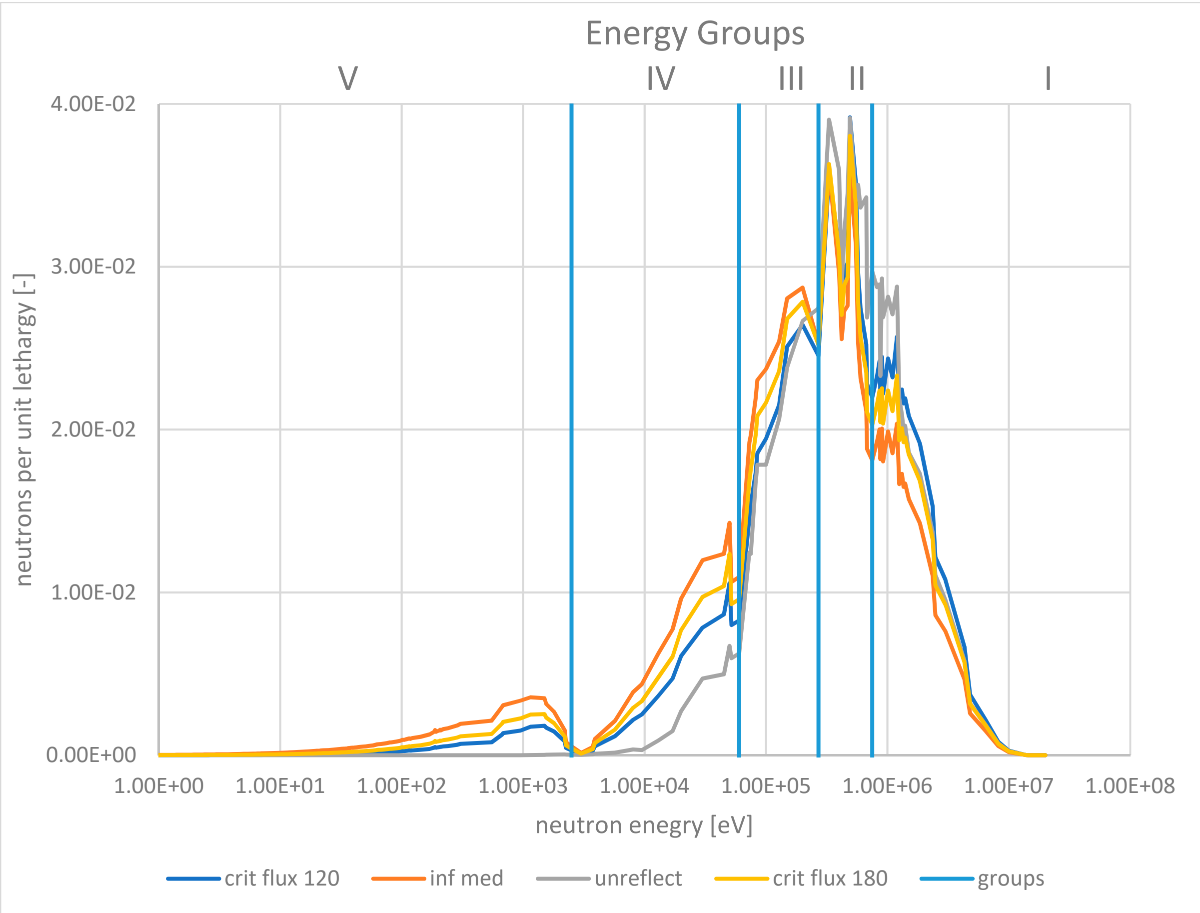

Figure 12. Neutron flux spectrum for the reflected and the unreflected core as well as for different axial leakage approximation (infinite medium, 120 cm core height, and 180 cm core height) using different core heights with added neutron group structure for the spatial flux distribution analysis.

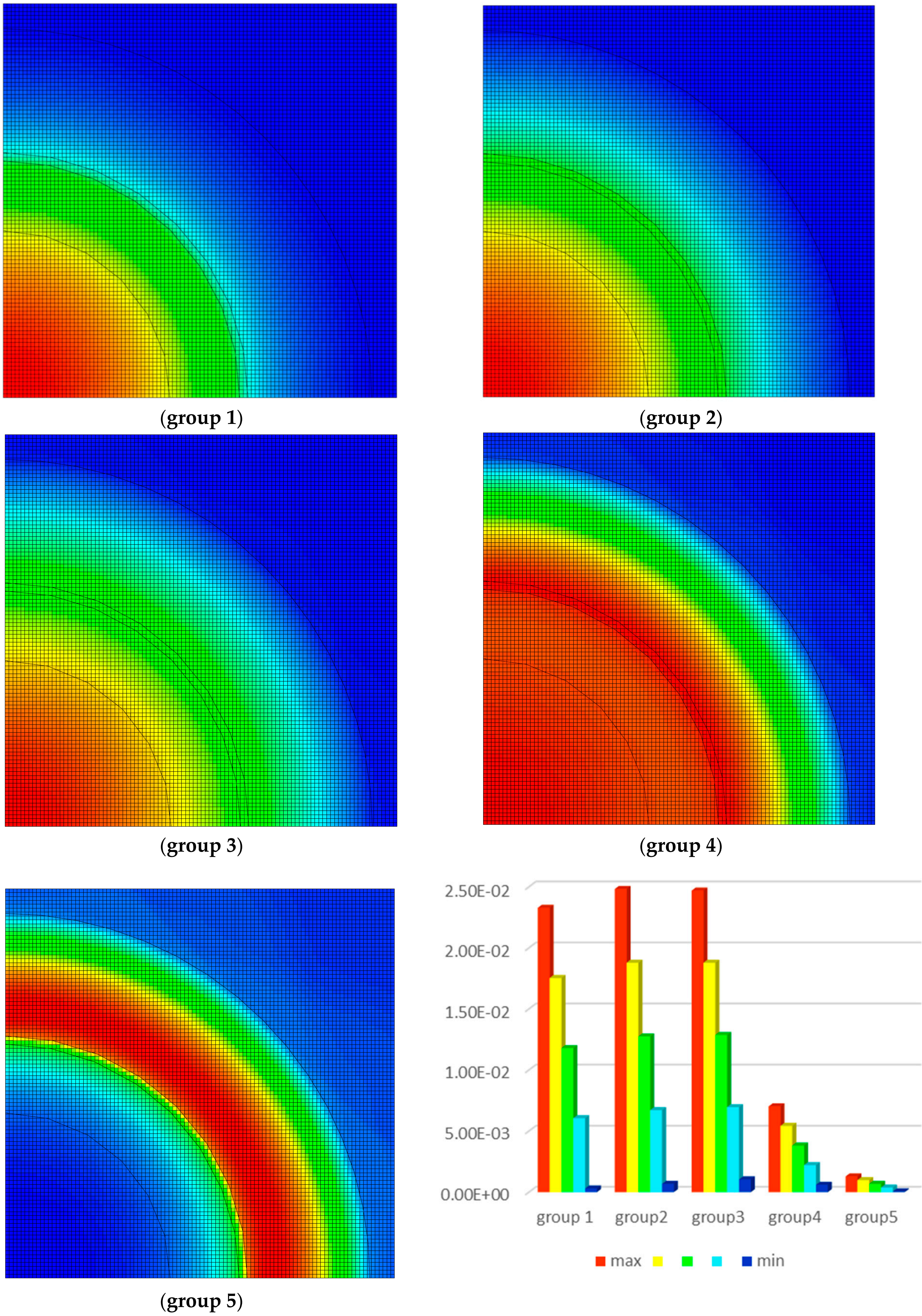

Figure 13. Spatial neutron flux distribution in the 5 energy groups defined in Figure 12 and related neutron flux values in each group with the colour map given in the bottom right figure (calculated using a TRITON-NEWT sequence).

3.6. 3-D Investigation of Core Volumes

Following the extensive 2-D analysis, the potential configurations are narrowed down to three different compositions for the 3-D analysis to create a first guess on the amount or volume of fuel, which will be required to achieve a critical configuration. The three compositions are:

- A eutectic composition with 35% enriched uranium
- A heavy metal rich composition with 35% enriched uranium
- A heavy metal rich composition with 20% enriched uranium

The choice for these compositions has been taken on the basis of the partly contradicting objectives given in the introduction. The representative choice is used to get a deeper understanding of the advantage in reducing the core size and consequently to the cost of the system achievable through the use of the heavy metal rich composition instead of the eutectic composition on the one hand; on the other hand, the extra volume is analyzed, which would be required if the aim is to create a core based on low enriched uranium. However, a final decision will have to be based on a larger set of criteria, potential configurations, and technological issues (e.g., max. permitted enrichment in the fuel production to assure a criticality safe production). The calculations performed are based on the 3-D model given in Figure 5 using for the primary investigation, the multi-group Monte Carlo code Keno VI and for a later cross-check the continuous energy version of the same code.

Table 3 indicates the expected results with the lowest core volume of less than 1.8 m^3 for the heavy metal rich system and an U-235 enrichment of 35%, which seems in the current state of the study the most promising combination. The change to the eutectic salt compositions would increase the required volume to achieve a critical system by almost 50%, while the switch to the more proliferation resistant fuel system with only 20% enrichment would increase the volume by almost a factor of 4. This would result in a core with a hardly acceptable size and volume for a zero power experiment. However, comparing the amount of UCl for the two 35% enriched cases with only less than 5% difference in the required UCl amount, it becomes obvious that the reduction of the core size when using the HM rich composition does not lead to a massive reduction of the fuel cost itself as long as we consider the UCl component as the cost intensive part. A real advantage of the HM rich system is only visible if the core size and the resulting size of the set-up is of importance, e.g., when the experiment should be set up in an existing facility. However, looking back to Figure 6, it would be worth an analysis to reduce the core size by a slight increase in the enrichment for the eutectic composition. In any of the 35% enriched cases, specific safeguarding requirements will have to be fulfilled which will not significantly change by a moderate increase of the enrichment as long as the criticality safety in the fuel production and handling can be assured. The comparably weak influence of the choice of a taller, but thinner core indicate that the results for the eutectic composition open the space for potential optimization of the core for a control system, which might work better in such a core geometry.

Table 3. Studied cases for the 3-D Monte-Carlo analysis to determine the minimum volume required to achieve a critical configuration.

	42.5% NaCl, 35% enr			**20% NaCl, 20% enr**			**20% NaCl, 35% enr**		
	Radius [cm]	**Height [cm]**	**Volume [m^3]**	**Radius [cm]**	**Height [cm]**	**Volume [m^3]**	**Radius [cm]**	**Height [cm]**	**Volume [m^3]**
Multigroup	67.2	100	2.84	104.25	100	**6.83**	65.6	65	**1.76**
	64.2	120	3.11						
	74.25	75	**2.60**						
Continuous Energy	75.25	75	2.67	105.55	100	7.00	66.55	65	1.81

The cross-comparision with the continuous energy solution indicates only a small difference below 3% regarding the system voume. These results confirm the quality and usability of the 252 group set used, not only for the 3-D, but also for the 2-D Monte-Carlo and the 2-D deterministic Sn transport solutions.

The detailed results given in Table 3 are depicted in Figure 14. From the visualization, it is clear that there is very good agreement of the results of both Monte-Carlo calculations (multi-group versus continuous energy). In addition, the clear advantage of the heavy metal rich system regarding volume optimization and the massive advantage of the use of the 35% enriched uranium is highlighted. Based on this outcome, a first recommendation for further investigation is to concentrate on the heavy metal rich system (in the case volume is relevant), but in any case with an enrichment of 35%, which seems to be a reasonable value when compared to other recent reactor physics experiments like GUINEVERE [26] with 30% enrichment and YALINA-Booster with an enrichment of 36% in the foremost lead fast zone [6].

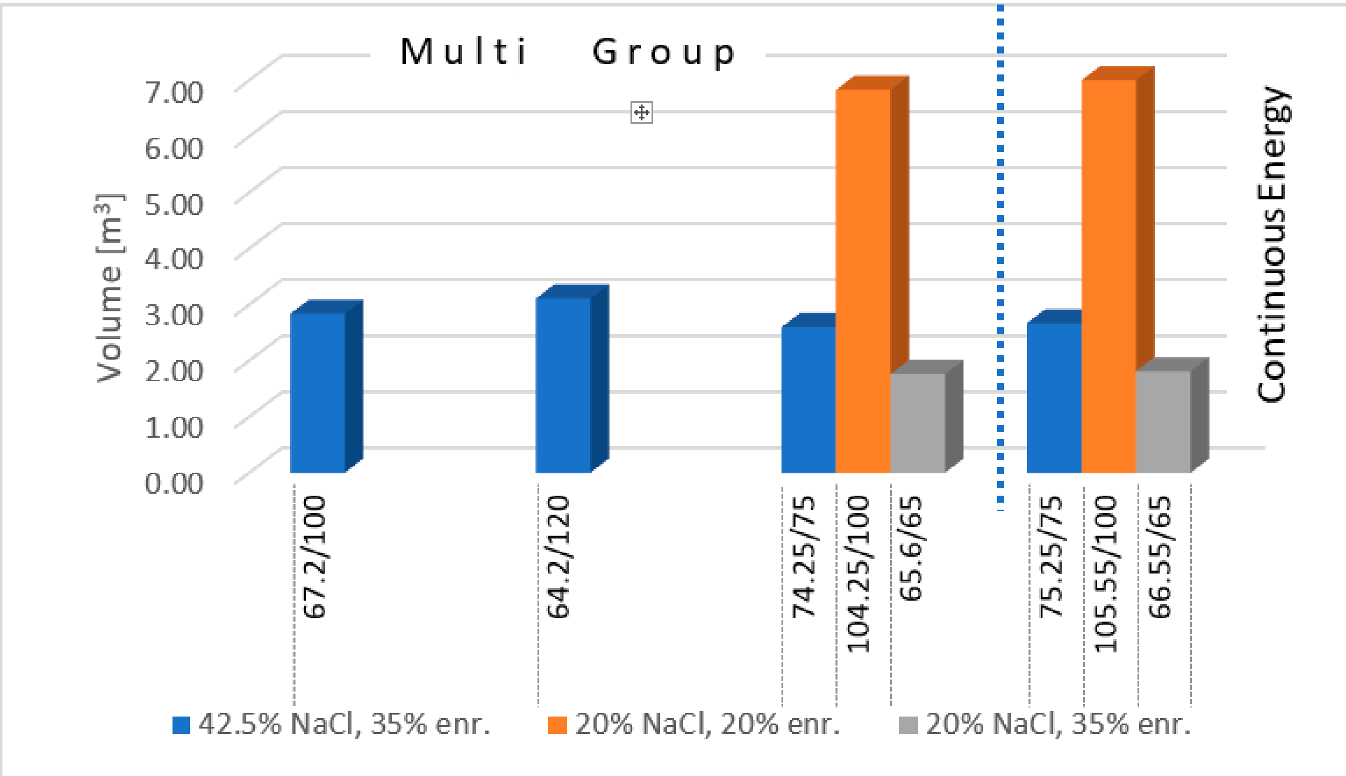

Figure 14. Expected core volumes for the different studied cases using multi-group and continuous energy Monte-Carlo simulations.

4. Summary and Conclusions

The investigated topic is introduced by making the case for the importance of a zero power reactor for the process of developing a new, innovative rector concept, such as a molten salt fast reactor. This is based on historical developments as well as the current demand for experimental results and relevant key factors for the success of the next step in the development process.

The focus of the modelling and simulation work in this publication is on answering the questions on salt compositions and the required dimensions of a potential experimental setup to narrow down the choices for a zero power experiment for a molten salt fast reactor. A multitude of modelling and simulation results using different tools of the SCALE package is delivered studying two different salt systems, the eutectic and the heavy metal rich systems. The studies on the 2-D system radius show the volume reduction to be strongest in the step from 20% enrichment to 35% enrichment and the reflector effect to be continuously increasing with enrichment. This results in reductions of the system radius, while the heavy metal rich system requires a smaller volume in all cases. The study of the thermal feedback effects (salt density and fuel temperature) shows a strong increase in the feedback effect with increasing enrichment and decreasing system radius with some indication of saturation at very high uranium enrichment.

The study of the neutron spectrum indicates that the systems have a slightly harder neutron spectrum than in a typical sodium cooled fast reactor with some limited spectral hardening in the heavy metal rich system, increased spectral hardening in the case of the absence of the reflector, and significant spectral hardening in the cases with very high uranium enrichment. The analysis of the spatial neutron flux distribution of the reflected system using five specifically defined energy groups demonstrates that a major part of the lower energy flux is created in the reflector through collisions, which will lead to a slight increase of the power production in the outermost core region while the majority of the fast neutron flux follows a distribution with a strong central neutron flux peak.

Based on the results and considerations on cost (required amount of fuel and potential cost increase as a function of core size) and non-proliferation/safeguarding requirements (potential use of high enriched Uranium >20%), three configurations have been chosen for the follow up 3-D analysis—the eutectic composition with 35% uranium-235 enrichment and the heavy metal rich compositions with 20% and 35% enrichment. The expected core volumes for these configurations have been studied using multi-group and continuous energy Monte-Carlo simulations, identifying the 35% enriched systems as the most attractive ones, but always with the caveat of the increased safeguarding requirements. Finally, if adding the potential facility size resulting from core size, the choice is disposed to the heavy metal rich composition of 35% enrichment as the reference system for future studies on zero power reactor investigation. However, if core size is not a limiting criterion, the eutectic system could be attractive too, since the required amount of UCl does not differ significantly between both compositions. The extra volume of the eutectic system is almost completely made up by the comparably cheap NaCl. The inter-comparison of the different applied codes and approaches available in the SCALE package has delivered a very good agreement between the results, creating trust in the developed and applied models and methods. However, this will not be sufficient to avoid the need for validation experiments to assure the best possible results for future modelling and simulation of molten salt reactor systems.

The current analysis is only applied for a very narrow window of operational temperature around 980 K in molten salt, which is related to the currently limited data availability. Thus, a much more comprehensive study is required as soon as the relevant thermophysical properties for the considered salt mixtures are available in room temperature to create a better understanding of a potential first core based on a solid salt mixture.

Author Contributions: Conceptualization, B.M.; Data curation, A.D., S.A. and G.C.-G.; Investigation, B.M. and A.D.; Methodology, B.M., D.L. and G.C.-G.; Software, A.D. and S.A.; Supervision, B.M.; Validation, D.L. and G.C.-G.; Visualization, A.D. and D.L.; Writing—original draft, B.M.; Writing—review & editing, D.L., A.D. and G.C.-G. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Engineering and Physical Sciences Research Council (EPSRC), grant number EP/V027239/1 162528/9.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Conflicts of Interest: The main author is part of the academic editorial board of the special edition, but was not involved in the review and decision-making process on this manuscript. The authors declare no conflict of interest.

References

1. Government and Industry Roles in the Research. *Development, Demonstration, and Deployment of Commercial Nuclear Reactors: Historical Review and Analysis*; EPRI: Washington, DC, USA, 2017; p. 3002010478.
2. Merk, B.; Litskevich, D.; Detkina, A.; Cartland-Glover, G.; Atkinson, S.; Bankhead, M. A zero-power facility as multi-fold opportunity to support quick progress in innovative reactor development. *ATW Int. J. Nucl. Power* **2021**, *66*, 59–64.
3. 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.
4. Drüke, V.; Filges, D. The critical HTGR test facility KAHTER—An experimental program for verification of theoretical models, codes, and nuclear data bases. *Nucl. Sci. Eng.* **1987**, *97*, 30–36. [CrossRef]
5. Chigrinov, S.E.; Kievitskaya, A.I.; Rakhno, I.L.; Serafimovich, I.G.; Rutkovskaya, K.K.; Khilmanovich, A.M.; Marstinkevich, B.A. Experimental and theoretical research on transmutation of long-lived fission products and minor actinides in a subcritical assembly driven by a neutron generator. In Proceedings of the 3rd International Conference on ADTTA, Prague, Czech Republic, 7–11 June 1999.
6. Kiyavitskaya, H.; Bournos, V.; Fokov, Y.; Martsynkevich, B.; Routkovskaia, C.; Gohar, Y.; Persson, C.-M.; Gudowski, W. *YALINA-Booster Benchmark Specifications for the IAEA Coordinated Research Projects on Analytical and Experimental Benchmark Analysis on Accelerator Driven Systems and Low Enriched Uranium Fuel Utilization in Accelerator Driven Sub-Critical Assembly Systems*; IAEA: Vienna, Austria, 2007.
7. GUINEVERE: Generator of Uninterrupted Intense Neutron at the lead VEnus Reactor. Available online: https://science.sckcen.be/en/Projects/Project/GUINEVERE (accessed on 25 November 2020).
8. Baeten, P.; Abderrahim, H.A.; Bergmans, G.; Kochetkov, A.; Uyttenhove, W.; Vandeplassche, D.; Vermeersch, F.; Vittiglio, G.; Ban, G.; Baylac, M.; et al. The Guinevere project at the VENUS-F Facility. In Proceedings of the European Nuclear Conference (ENC 2010), Barcelona, Spain, 30 May–2 June 2010; pp. 388–392.
9. Merk, B.; Litskevich, D.; Whittle, K.R.; Bankhead, M.; Taylor, R.; Mathers, D. On a long term strategy for the success of nuclear power. *Energies* **2015**, *8*, 12557–12572. [CrossRef]
10. Resilient and Sustainable—Terrestrial Energy. Available online: https://www.terrestrialenergy.com/technology/sustainable-clean-energy/ (accessed on 25 November 2020).
11. Southern Company and TerraPower Prep for Testing on Molten Salt Reactor. Available online: https://www.energy.gov/ne/articles/southern-company-and-terrapower-prep-testing-molten-salt-reactor (accessed on 25 November 2020).
12. ELYSIUM INDUSTRIES Molten Chloride Salt Fast Reactor. Available online: https://thoriumenergyalliance.com/wp-content/uploads/2020/02/Elysium-MCSFR-TEAC10-Update.pdf (accessed on 19 July 2021).
13. На ГХК прошло рабочее совещание по вопросу создания жидкосолевого реактора. Available online: https://www.sibghk.ru/news/9068-na-gkhk-proshlo-rabochee-soveshchanie-po-voprosu-sozdaniya-zhidkosolevogo-reaktora.html (accessed on 25 November 2020).
14. Latkowski, J. Terra Power's Molten Chloride Fast Reactor. 22 February 2021. Available online: https://www.nationalacademies.org/event/02-22-2021/merits-and-viability-of-different-nuclear-fuel-cycles-and-technology-options-and-the-waste-aspects-of-advanced-nuclear-reactors-february-22-and-23-2021-meeting (accessed on 6 July 2021).
15. Merk, B.; Detkina, A.; Litskevich, D.; Atkinson, S.; Cartland-Glover, G. The interplay between breeding and thermal feedback in a molten chlorine fast reactor. *Energies* **2020**, *13*, 1609. [CrossRef]
16. Merk, B.; Detkina, A.; Atkinson, S.; Litskevich, D.; Cartland-Glover, G. Evaluation of the breeding performance of a NaCl-UCl-based reactor system. *Energies* **2019**, *12*, 3853. [CrossRef]
17. Desyatnik, V.N.; Katyshev, S.F. Volumetric and surface properties of the NaCl-UCl3-UCl4 melts. *Zhurnal Fiz. Khimii* **1980**, *54*, 1606–1610.
18. Jessee, M.A.; Jarrell, J.J.; Wieselquist, W.A.; Williams, M.L.; Kim, K.S.; Evans, T.M.; Hamilton, S.P.; Gentry, C.A. POLARIS—2D light water reactor lattice physics module. In *Scale Code System*; Rearden, B.T., Jessee, M.A., Eds.; Oak Ridge National Laboratory: Oak Ridge, TN, USA, 2017.
19. Leppänen, J.; Pusa, M.; Viitanen, T.; Valtavirta, V.; Kaltiaisenaho, T. The serpent monte Carlo code: Status, development and applications in 2013. *Ann. Nucl. Energy* **2015**, *82*, 142–150. [CrossRef]
20. Rearden, B.T.; Jessee, M.A. (Eds.) *Scale Code System, ORNL/TM-2005/39 Version 6.2.2*; Oak Ridge National Laboratory: Oak Ridge, TN, USA, 2016.
21. Merk, B.; Detkina, A.; Atkinson, S.; Litskevich, D.; Cartland-Glover, G. Evaluating Reactivity Control Options for a Chloride-Salt Based Molten Salt Zero Power Reactor. 2021. Available online: https://www.preprints.org/manuscript/202106.0552/v1 (accessed on 19 July 2021).
22. Merk, B.; Detkina, A.; Atkinson, S.; Litskevich, D.; Cartland-Glover, G. Innovative Investigation of Reflector Options for the Control of a Chloride-Salt Based Molten Salt Zero Power Reactor. 2021. Available online: https://www.preprints.org/manuscript/202106.0559/v1 (accessed on 19 July 2021).
23. Merk, B.; Rohde, U.; Glivici-Cotruţă, 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] [PubMed]
24. Merk, B.; Fridman, E.; Weiß, F.P. On the use of a moderation layer to improve the safety behavior in sodium cooled fast reactors. *Ann. Nucl. Energy* **2011**, *38*, 921–929. [CrossRef]

25. Merk, B.; Glivici-Cotruţă, V.; Weiß, F.P. On the use of different analytical solutions for recalculation of the YALINA-Booster experiment SC3A. *Progress Nucl. Energy* **2012**, *58*, 11–20. [CrossRef]
26. Uyttenhove, W.; Baeten, P.; van den Eynde, G.; Kochetkov, A.; Lathouwers, D.; Carta, M. The neutronic design of a critical lead reflected zero-power reference core for on-line subcriticality measurements in Accelerator Driven Systems. *Ann. Nucl. Energy* **2011**, *38*, 1519–1526. [CrossRef]