On the Feasibility of Breed-and-Burn Fuel Cycles in Molten Salt Reactors

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Abstract. Reactors operated in an open cycle generally suffer from poor fuel resources utilization, typically below 1%. An exception to this are Breed-and-Burn reactors, which after long irradiation of the feed fuel are able to operate with minimum fissile input and fuel processing without actinides separation, which makes them particularly attractive. Historically, the concept has essentially been considered for solid-fuel fast reactors, more specifically, for sodium-cooled fast reactors. Their performance in Breed-and-Burn is however limited by the neutron fluence necessary to reach break-even neutron generation and thus high fluence on the fuel cladding, as well as their spatially inhomogeneous fuel burn-up. However, molten salt reactors do not suffer from such limitations, but have other constraints due to their use of a homogeneous fuel. In this paper, the neutronics constraints in term of fuel selection and utilization and reactor dimensions are studied and reported**.**

Key Words: Molten Salt Reactor, Breed-and-Burn, Fuel cycle, Chlorides

1. Introduction

One major objective of the so-called Generation IV reactors is a substantial increase in sustainability compared to reactors of the previous generations [1]. This goal is twofold: on the one hand, better utilization of natural resources thanks to the efficient burning of natural uranium and thorium, and on the other hand, a decrease of the amount of long-lived radioactive waste produced by the reactor and its fuel cycle.

A key in achieving both the former and the latter is breeding, that is, the capability of a reactor to produce fissile material from a poorly fissile feed. Additionally, high breeding capabilities increase the availability of neutrons to be spent for waste burning. The most resource-efficient use of breeding is in an ideal closed fuel cycle where all actinides are recycled up to approximately 95% utilization (because of losses to reprocessing). However, it may be both technically and economically challenging. On the other hand, an open cycle with low conversion ratio may not be sustainable enough, because reaching a high burn-up does not entirely compensate the necessary initial enrichment. For example, a typical Pressurized Water Reactor, despite reaching a burn-up of 5% FIMA (Fissions per Initial Metal Atoms), only has a natural uranium utilization of approximately 0.5% because of the required fuel enrichment. For both reasons, Breed-and-Burn (BNB) fuel cycles that are both open and sustainable due to their very high breeding could be attractive.

In this paper, the feasibility of implementing a Breed-and-Burn fuel cycle in Molten Salt Reactors (MSRs) using liquid fuels is investigated from a neutronics and fuel cycle point of view. First, a more detailed definition of BNB is presented, followed by the description of the method used for the computations in this paper. Finally, a characterization of the performance of potential fuel salt compositions and reflector materials for use in a reactor.

2. Model of a Breed-and-Burn Fuel Cycle

Breed-and-Burn is a mode of operation in which the reactor breeds and subsequently burns the fuel necessary for its operation while using a poorly fissile feed of fertile material [2, 3]. Such a fuel cycle requires the reactor to have an excellent neutron economy (high breeding, low parasitic neutron captures), which is typically only possible in a fast neutron spectrum. Sodium-cooled Fast Reactors have mainly been proposed for implementation of this fuel

cycle. However, the very high cladding fluence needed to reach self-sustaining BNB operation are technologically challenging.

Molten Salt-fueled Reactors have the particularity of using a liquid fuel, which does not suffer from radiation damage, contrarily to the more common solid fuels. Moreover, in the most common implementations, the fuel is not contained within cladding tubes because it serves both as fuel and as coolant. Liquid fuels can therefore theoretically remain in the core indefinitely, if sufficient solubility and stability of fission products can be guaranteed at high fuel burn-ups. Furthermore, refueling operations could potentially be simpler: a small fraction of the fuel volume would be emptied and replaced by feed.

In this section, the method used in this paper to evaluate the Breed-and-Burn capabilities of candidate fuel salts is detailed. It consists of three main steps performed for each fuel salt:

1. A standard depletion calculation with constant flux level of a unit cell containing the fertile fuel salt is performed from 0% to approximately 100% burn-up and taken as a reference.
2. The adequate weighting function describing the distribution of the amount of time spent by a given volume of fuel in the core for a given in-core residence time is computed.
3. Averages of quantities of interest such as the infinite multiplication factor are computed using the two previous steps.

In the following subsections, each of these steps is described in further details.

2.1. Depletion of the representative fresh fertile fuel

In this step, a simple unit cell (cube with sides of 1 cm) containing the fresh fuel is depleted at constant flux (in this work, a flux of 1×10^{15} n/cm^2.s was used, corresponding to a power density range of 15-30 W/cm^3 at equilibrium for cases of interest) from 0% to approximately 100% FIMA[1]. The purpose of the depletion calculation is to obtain representative values of parameters of interest during the life of the fuel.

Two parameters, k_{inf} and the net number of neutrons generated $P_{n,}$ are evaluated as functions of irradiation time t:

$$k_\infty(t) \quad \text{and} \quad P_n(t) = \bar{\nu}(t) R_F^{total}(t) - R_A^{total}(t)$$

Knowing the representative evolution of the fuel during burnup, it is assumed that the performance of a reactor with a mixture of fuels of different burnups can be approximated with an average value of k_{inf} and of the neutron excess P_n.

2.2. Burnup distribution in the BNB core

To obtain the averages mentioned before, adequate distribution functions should be used for the case at hand. Their purpose is to describe the simultaneous presence of different burnups, which can also be seen as fuels of different "ages" in the core.

In solid fuel reactors, the distribution is flat, because the entirety of the fuel goes through the irradiation time equally and the entirety of the contents of a given assembly remains in the

[1] Due to technical limitations of the codes used, the depletion could not effectively be conducted up to 100% FIMA, however, the obtained results were linearly extrapolated until that point.

core for a fixed amount of time. Hence, from fresh fuel to the fuel discharged at irradiation time T all burnups are equally present (see FIG. 1) and the distribution has the following form:

$$p(t) = 1/T \quad \text{for} \quad t \in \langle 0, T \rangle \quad \text{and} \quad p(t) = 0 \quad \text{for} \quad t > T$$

Therefore, T is both the average and maximum irradiation time in the case of solid fuel.

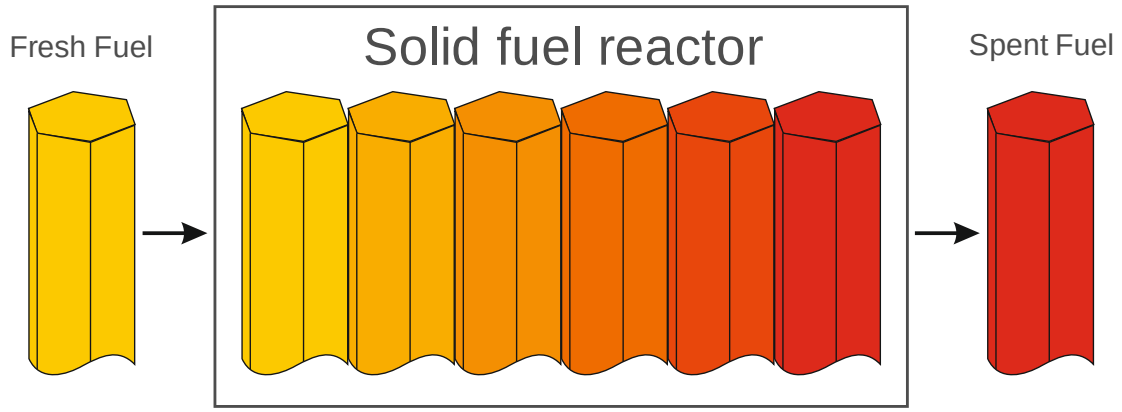

FIG. 1 Illustration of fuel burn-up distribution in the case of a solid fuel reactor.

In a liquid fuel reactor, however, the discharged fuel is the average fuel because everything is mixed in the liquid state. Consequently, a given amount of fuel of all burnups is necessarily discharged. Moreover, part of the fuel will remain in the core longer than the average irradiation time, contrarily to the solid fuel case (see **Error! Reference source not found.**).

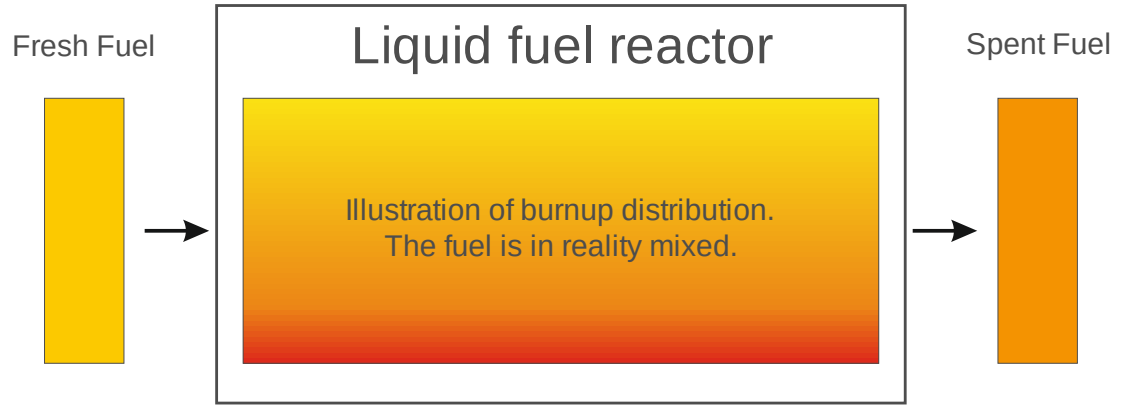

FIG. 2 Illustration of fuel burn-up distribution in the case of a liquid-fuel reactor. The discharged fuel is an average of all the burn-ups.

The removal of fuel is described by a similar equation as exponential decay; considering a total volume of fuel V_{fuel}^{total} and a volumetric discharge rate of $\dot{v}_{feed}$, one gets:

$$\frac{dN}{dt} = -\frac{\dot{v}_{feed}}{V_{fuel}^{total}} N = -\lambda N .$$

Accordingly, the salt removal speed is equal to the decay constant and the fuel burnup distribution function in liquid fuel reactor has following form:

$$p(t) = \lambda e^{-\lambda t} .$$

The average irradiation time T of the discharged fuel is thus:

$$T = \int_0^\infty tp(t)dt = \int_0^\infty t\lambda e^{-\lambda t}dt = \frac{1}{\lambda} = \frac{V_{fuel}^{total}}{\dot{v}_{feed}} \quad (1)$$

An illustration of the difference in the distribution functions for solid and liquid fuel is given in FIG. 3.

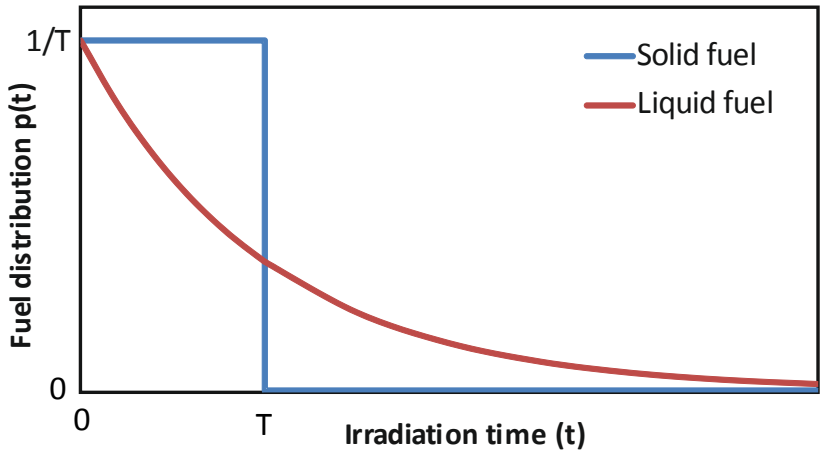

FIG. 3: Distribution functions as function of irradiation time for solid and liquid fuel. The solid-fuel case follows a flat distribution, while the liquid-fuel case follows and exponentially decreasing distribution.

Using the above-mentioned distributions, one can then derive the average quantities needed to evaluate the performance of the candidate fuel salts for a given discharge time or burn-up.

2.3. Average values for a given discharge time

For a solid fuel reactor with discharge time T, the average neutron excess is thus:

$$\bar{P}_n(T) = \int_0^\infty p(t)\left(\bar{\nu}(t)R_F^{total}(t) - R_A^{total}(t)\right)dt = \frac{1}{T}\int_0^T \left(\bar{\nu}(t)R_F^{total}(t) - R_A^{total}(t)\right)dt$$

Knowing that the irradiation flux is constant the equation can be rewritten as follows:

$$\bar{P}_n(T) = \frac{\Phi}{T}\int_0^T \bar{\nu}(t)\Sigma_F^{total}(t)\left(1 - \frac{R_A^{total}(t)}{\bar{\nu}(t)R_F^{total}(t)}\right)dt = \frac{\Phi}{T}\int_0^T \bar{\nu}(t)\Sigma_F^{total}(t)\left(1 - \frac{1}{k_\infty(t)}\right)dt$$

Similarly, the average k_∞ for the selected discharge time T can be expressed as:

$$\bar{k}_\infty(T) = \int_0^\infty p(t)k_\infty(t)dt = \frac{1}{T}\int_0^T k_\infty(t)dt$$

In a similar fashion as the solid-fuel case, the average neutron excess can be derived for liquid fuels using the burnup distribution function as follows:

$$\bar{P}_n(T) = \int_0^\infty \lambda e^{-\lambda t}\bar{\nu}(t)\Sigma_F^{total}(t)\left(1 - \frac{1}{k_\infty(t)}\right)dt = \frac{1}{T}\int_0^\infty e^{-\frac{t}{T}}\bar{\nu}(t)\Sigma_F^{total}(t)\left(1 - \frac{1}{k_\infty(t)}\right)dt,$$

where the Eq. 1 was applied for the substitution. The average k_∞ in a liquid fuel reactor for the selected discharge time T can be expressed as:

$$\bar{k}_\infty(T) = \int_0^\infty p(t)k_\infty(t)dt = \int_0^\infty \lambda e^{-\lambda t} k_\infty(t)dt = \frac{1}{T}\int_0^\infty e^{-\frac{t}{T}} k_\infty(t)dt$$

2.4. Transformation of irradiation time T in burn-up B

The average fission rate can be used to express the burnup in FIMA %. Its value can be expressed equally as the average k-infinity. For solid fuel it is thus:

$$\bar{R}_F^{total}(T) = \int_0^\infty p(t)R_F^{total}(t)dt = \frac{1}{T}\int_0^T R_F^{total}(t)dt$$

Or in the following manner for liquid fuel:

$$\bar{R}_F^{total}(T) = \int_0^\infty p(t)R_F^{total}(t)dt = \int_0^\infty \lambda e^{-\lambda t} R_F^{total}(t)dt = \frac{1}{T}\int_0^\infty e^{-\frac{t}{T}} R_F^{total}(t)dt$$

Since the irradiation flux is constant the reaction rate integrals can be replaced by macroscopic cross-sections integrals and multiplying them by the said flux.

Using the average fission rate, one can derive the average discharge burn-up (expressed in % FIMA) by normalizing with the initial number of actinide atoms N_0:

$$B(T) = 100\frac{T\bar{R}_F^{total}(T)}{N_0}$$

Burn-up being a more practical quantity to use, from this point on all results are presented in this unit.

2.5. Verification of the model

To verify the accuracy of the method, a test case comprising a unit cell of NaCl-UCl_3 (60-40 mol%) was depleted and its equilibrium quantities derived using this method. Additionally, the same cell was explicitly depleted to equilibrium with the EQL0D procedure [7] and the Serpent 2 code [8] with the ENDF/B-VII.0 nuclear data library using different reprocessing rates. The model predictions of k-infinity are subsequently compared to the direct calculations on FIG. 4.

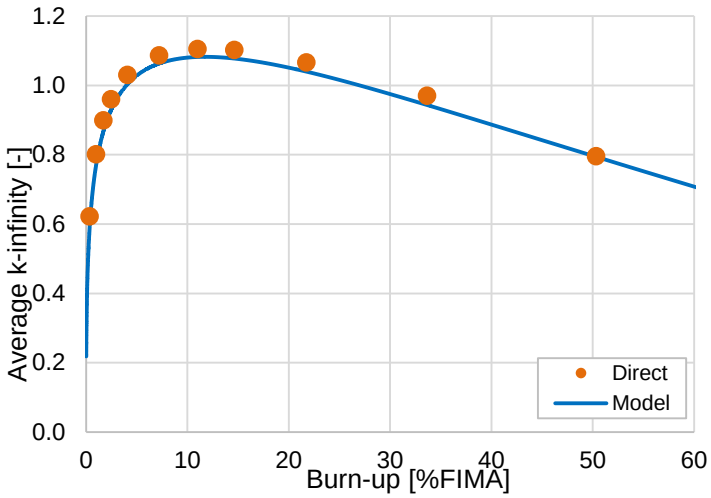

FIG. 4: Comparison of model predictions and direct calculations at selected burn-ups for the selected test case (NaCl-UCl3, 60-40 mol.%).

As depicted by FIG. 4, the model predictions are close to the results obtained by direct calculation. Depending on the case, the model may slightly over- or underestimate the infinite multiplication factor k-infinity, but within a margin that is acceptable for scoping studies. In the following section, the results of the investigations of several salt candidates obtained using this method are presented.

3. Evaluation of Candidate Fuel Salts

Using the method detailed previously, scoping studies have been performed on several fuel salts to compare their performance depending on various parameters. In the following, the maximum achievable k-infinity in BNB was investigated by using the method with several chloride salt compositions using both the Uranium-Plutonium and Thorium-Uranium cycles. Previous work has shown that, essentially, only chloride salts where the chlorine is enriched in its isotope 37 is of interest for Breed-and-Burn [6], therefore, all the salts investigated in this paper are of this type (Chlorine is considered to be pure Chlorine 37). TABLE I summarizes the salt compositions used for the scoping studies presented here.

To obtain a good performance in a BNB cycle, one must find a salt composition with an actinide density that is as high as possible. This, however, usually comes at the cost of a higher melting point, which is impractical. Nonetheless, salts containing only actinides chlorides were investigated for comparison purposes. While salts containing UCl_4 generally have a lower melting point than those containing UCl_3, the former is expected to be thermally unstable at high temperatures, making it a questionable choice. Additionally, the capability of pure actinide chloride salts to retain a substantial amount of fission product chlorides in solution in the vicinity of the original melting temperature is uncertain. However, salts compositions containing both compounds were nonetheless investigated from a neutronics point of view.

Finally, previous results [6] have shown that uranium-bearing salts are expected to perform better than thorium-bearing salts. A salt composition containing some Light Water Reactor (LWR) Plutonium was also added to quantify the improvement in reachable burn-up with some added fissile material.

TABLE I: CANDIDATE FUEL SALT COMPOSITIONS AND PROPERTIES

Salt	Composition	Tot. density	Act. density	Melting T.

Unit	mol.%	g/cm^3	g/cm^3	°C
NaCl-UCl_3	68-32	3.32	1.66	520
NaCl-UCl_3	60-40	3.64	1.97	590
UCl_4-UCl_3	80-20	3.79	2.38	545
UCl_4	100	3.56	2.20	590
NaCl-$ThCl_4$	55-45	3.15	1.61	375
$ThCl_4$	100	3.82	2.33	770
NaCl-(Th,Pu)$Cl_{4,3}$	55-40-5	3.15	1.76	375

The first quantity of interest is the infinite multiplication factor that can be reached in BNB mode as function of the discharge burn-up. Additionally, the maximum and minimum burn-up achievable for a given value of k-inf are also of importance. The results were derived using the method described above using the EQL0D procedure and the Serpent 2 code [8] with the ENDF/B-VII.0 nuclear data library. The results are depicted on FIG. 5 below. Black markers have been added at the point of maximum k-inf for each salt. For each cases, a minimum and maximum burn-up can be observed where the infinite multiplication factor first and last reaches unity. The infinite multiplication factor first increases with increasing burn-up (or irradiation time) because at low cycle times, the fuel is removed too fast, therefore what is bred is removed and replaced by poorly fissile feed.

On the other hand, at high burn-up (or irradiation time), not enough fission products are removed and replaced by fertile feed, which is detrimental to the breeding performance. The maximum infinite multiplication factor is reached when the addition of fertile feed is sufficient to breed but not enough for the fertile salt to dominate, and when the removal of fission products is sufficient not too impede breeding to much while maintaining the losses of bred products to removal low enough.

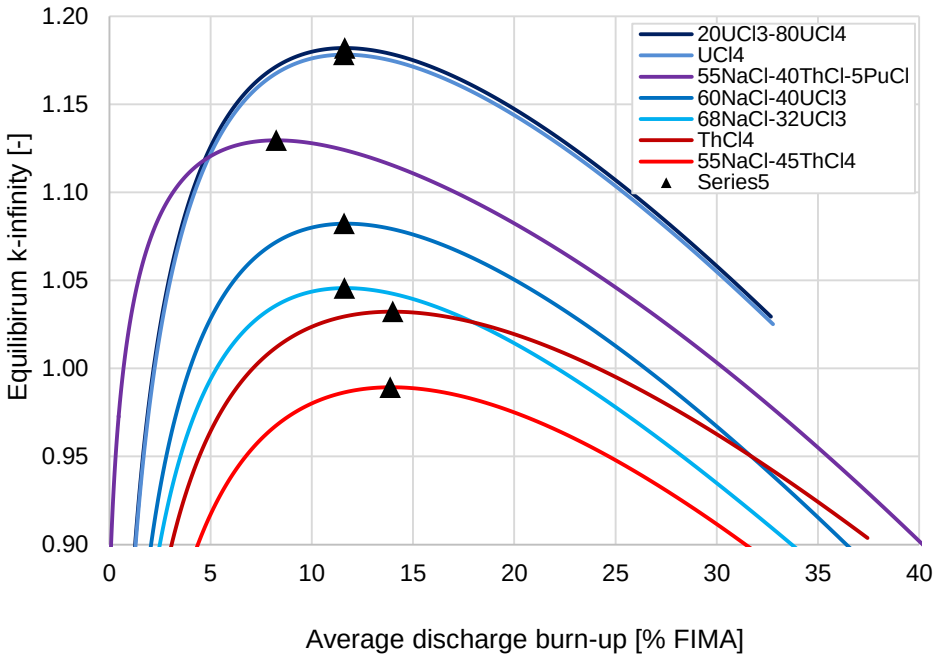

FIG. 5: Infinite multiplication factor for selected salts as function of discharge burn-up.

As depicted on FIG. 5, the best performance is obtained for the actinide-only Uranium chloride salts, which show both the highest possible k-inf and discharge burn-up of ca. 35%. The worst performance is obtained by Thorium-bearing salts without Plutonium support, e.g. the mixed Thorium chloride salt barely reached criticality on an infinite lattice and the pure Thorium tetrachloride salt reaching 24% maximum burn-up. The composition with 5 mol.% $PuCl_3$ performs better however, reaching a maximum burn-up of ca. 31%. Mixed Uranium chloride salts have an intermediate performance, with the 40 mol% UCl_3 reaching 27% burn-up and the 32 mol.% one reaching 22% burn-up. Obviously, a reactor cannot be operated below the minimum burn-up or above the maximum burn-up since the equilibrium k-infinity is lower than unity in those cases.

An open question is the possibility of maintaining the accumulated fission products (particularly, lanthanides which form chloride compounds not removed by bubbling) in solution at such fuel burn-ups. For example, in the $NaCl$-UCl_3 case, the composition at approximately 11% FIMA is expected to contain up to 2.69 mol.% Nd, 1.53 mol.% Ce, 0.82 mol.% Sm, and so on. Individual fission products with concentrations in the molar range are not expected to change the melting point of the mixture substantially, but the question of their joint solubility is open, as the experience with the Molten Salt Reactor Experiment is not directly applicable to this case due to the low burn-ups achieved and the use of fluoride salts.

Moreover, these results do not account for neutron leakage, since they are computed at the infinite lattice level. For a reactor of finite dimensions, a reactivity margin must be considered to compensate for neutron leakage. To minimize the leakage and thus the critical dimensions of the core, a neutron reflecting material must be added around it. In the following section, a finite-size reactor comprising several candidate reflector materials is investigated in conjunction with several fuel salt candidates to quantify the achievable size, volume, and inventory of the reactor.

4. Critical Core Dimensions and Reflector Options

Having determined that BNB is feasible at the infinite lattice level for several salt compositions, the subsequent step is to investigate the resulting dimensions of a finite-size core. To minimize the physical size of the core, an adequate reflector material must be found; a challenging task due to the neutron “transparency” of chlorides salts which combine a low actinide density with a hard neutron spectrum, making finite-sized cores more susceptible to neutron leakage.

TABLE II: PROPERTIES OF CANDIDATE REFLECTOR MATERIALS

Material	Density	Isotopic composition
Fe	7.5 g/cm^3	Natural
Zr	6.4 g/cm^3	Natural
Pb	10.3 g/cm^3	Natural
Pb-208	10.3 g/cm^3	100% Pb-208

The critical dimensions of cores surrounded by iron-, zirconium-, lead, and lead-208-based reflectors were determined using the EQL0D procedure for the above-mentioned salt compositions, apart from the sodium-thorium chloride salt. The core geometry is assumed to be cylindrical with a height-to-radius ratio of 1.84. The reflector thickness is assumed to be 100 cm in all directions. Their equilibrium compositions were derived for discharge burn-ups

corresponding to maxima of k-infinity, as previously predicted. The properties of the reflector materials investigated in this work are given on TABLE II above. Using the above-mentioned reflector materials, the equilibrium critical dimensions of a BNB core using a given salt were computed. FIG. 6 reports the core volumes in these conditions.

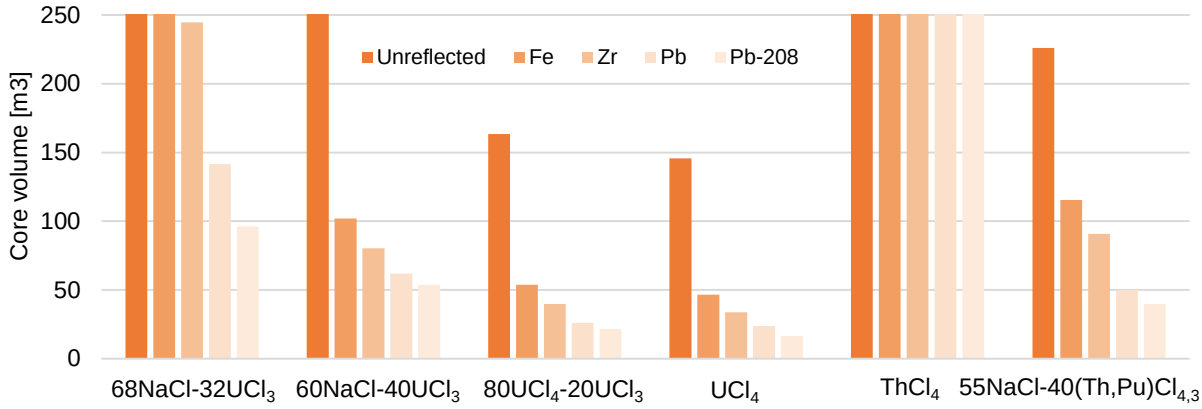

FIG. 6: Equilibrium critical core volume for several salts and reflector materials

The results show that a high actinide density truly is critical to obtaining a feasibly small physical size, indeed the lower density sodium-uranium chloride salt 68NaCl-32UCl_3 necessitates a core volume of 100 m^3 and above depending on the reflector material. The high-density Uranium-bearing salt show much smaller core volumes, while the Thorium-bearing high-density salt has unfeasibly high core volumes regardless of the reflector material used. The mixed Thorium-Plutonium salt however requires core volumes below 100 m^3 for Zr or Pb reflectors. Regardless of the salt composition, lead reflectors (natural or enriched) have the best performance, with Zr being second and Fe last.

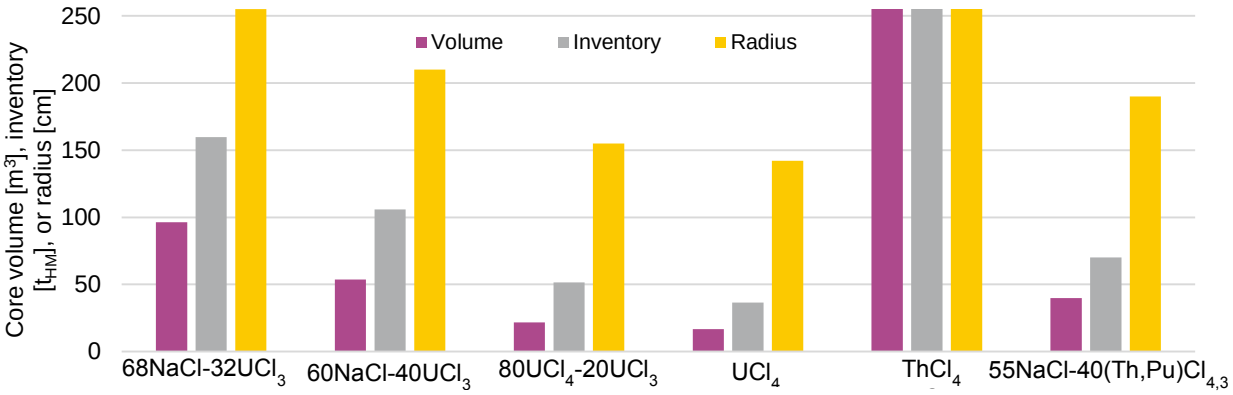

FIG. 7: Minimal core radius, volume and inventory of the investigated salts.

While a higher actinide density can lead to lower core volumes, the combined effect of higher density and lower volume on the total inventory must also be determined. On FIG. 7, the minimum core radii, volumes and Heavy Metal (HM) inventories for each salt, and in combination with a 208-Pb reflector, are given.

Assuming a potent reflector material such as Pb-208, the volumes and inventories needed for BNB can reach values as low as 17 m^3 for pure UCl_4 and 22 m^3 for the eutectic mixture of UCl_3 and UCl_4. For less speculative fuel salts, the necessary volumes reach the level of 54 m^3 for the NaCl-UCl_3 (60-40 mol.%), which is still acceptable, given the resulting HM inventory of 106 tons.

5. Conclusion

The feasibility of using a Breed-and-Burn fuel cycle in Molten Salt Reactors was investigated from a neutronics and fuel cycle point of view using a simple model based on the depletion of a representative unit cell of fertile material and the use of a residence time distribution of the fuel in the case of a liquid-fuel reactor.

The investigations presented in this paper show that Breed-and-Burn is indeed feasible in Molten Salt Reactors using a suitable salt composition and reflector material, if the salt used is chloride-based where the chlorine is enriched in chlorine 37. The smallest reactor inventories are reached when using salts with high actinide densities. Burn-ups of 10-12 % FIMA can be reached at the smallest core size.

The feasible core dimensions and volumes with most salts and reflector materials remain large, however, compared to other Molten Salt Reactor concepts. Moreover, the technical feasibility of the concept remains to be demonstrated from other points of view (engineering, chemistry, materials).

Acknowledgements

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