

Parametric studies on the fuel salt composition in thermal molten salt breeder reactors

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Abstract

In this paper the salt composition and the fuel cycle of a graphite moderated molten salt self-breeder reactor operating on the thorium cycle is investigated. A breeder molten salt reactor is always coupled to a fuel processing plant which removes the fission products and actinides from the core. The efficiency of the removal process(es) has a large influence on the breeding capacity of the reactor. The aim is to investigate the effect on the breeding ratio of several parameters such as the composition of the molten salt, moderation ratio, power density and chemical processing. Several fuel processing strategies are studied.

1. Introduction

There is a renewed interest in molten salt reactors (MSR) since the design was chosen for Generation IV. The MSR uses a molten salt fluid fuel containing actinide salts and other salts providing low melting point and good heat transfer properties. The concept of fluid fuel has numerous advantages on fuel fabrication and fuel processing and last but not least has the possibility of making changes to the composition during operation. Besides the advantages of the high temperature operation (hydrogen production, efficient electricity production) the MSR is a promising actinide burner or breeder reactor.

MSRs were extensively studied both experimentally and theoretically by ORNL in the 60's and 70's. The studies of Oak Ridge showed that the MSR is an attractive breeder reactor on the thorium fuel cycle. Recently many studies were

carried out on the physics and chemistry of the thorium fuelled MSR. The Molten Salt Breeder Reactor (MSBR) (Robertson et al., 1970), the original design from ORNL, suffers from major drawbacks, mainly on safety and fuel processing, as the results of these investigations (MOST, 2003) show.

The ability to chemically process the fuel during operation is the most important advantage of the MSR if the goal is breeding. Without continuous removal of some Fission Products (FP) the reactor becomes subcritical quickly.

The thorium fuel cycle produces much less TRansUranium (TRU) elements than the U-Pu cycle and it can be used both in epithermal and fast mode. Since ^{232}Th is the only isotope present in the thorium ore, enrichment of Th is not necessary for a thorium fuelled reactor.

2. Description of the study

The aim of this study is to choose a proper salt mixture and processing scheme for a graphite-moderated self-breeder molten salt reactor. To reach

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self-breeding, the molten salt of the reactor has to meet several criteria:

- The neutron capture rate in the salt has to be low enough to have excess neutrons to transmute thorium.
- The fuel salt needs to have a negative temperature feedback coefficient to compensate for the positive effect of the graphite.
- Large amounts of actinides must be soluble in the salt and the heat transfer properties are important.
- The short- and long-lived radioactive waste production of the molten salt should be minimized.

The candidate salts are binary and ternary mixtures of LiF, NaF, BeF₂, ZrF₄ and RbF. These mixtures are chemically stable, compatible with graphite and nickel-based Hastelloy alloys, are not volatile, and can withstand intense radiation. Furthermore they can dissolve the needed amount of actinides and they have good heat transfer properties (MOST, 2003; Williams, 2006).

First we investigated the breeding ratio and criticality of different salt compositions with a pre-set amount of thorium and uranium, altering the amount of graphite moderator. One salt fulfilling the criteria and the amount of moderator corresponding to Breeding Ratio (BR) slightly higher than unity are chosen for further investigations of the processing.

Several ways of processing are considered: batch-wise processing and continuous on-line processing with or without extraction of the minor actinides (MA). The possibility of removing protactinium from the salt is also investigated. The continuous removal of fission product noble gases (Xe, Kr) and noble metals (Se, Nb, Mo, Sb, Te, Ru, Rh, Pd, Ag, Tc) is assumed in every burnup calculation by a gas removal system, possibly helium bubbling. The speed of the processing is a critical parameter to achieve self-breeding so the minimum removal rate for FPs and actinides will be determined for the chosen candidate salt.

3. Methodology

3.1. Computational tools

The calculation scheme is based on the SCALE-5 code system (SCALE, 2005). Neutronic calculations with 172 and 238 group cross sections are performed assuming an infinite core made of molten salt channels surrounded by graphite moderator. In the XSDRN calculations this is realized as a cylindrical fuel channel surrounded by an annular moderator region. A white boundary condition is used on the outer surface of the moderator. For burnup analysis the ORIGEN-S burnup/decay code is used.

The CSAS/XSDRN is a static code and does not take into account the flow of the fuel. In reality, a part of the delayed neutron precursors exit with the flow and decay in the primary loop outside the reactor. These neutrons are lost for the chain reaction. The delayed neutron fraction is 0.0027 in a reactor fuelled with ²³³U. Suppose one third of the fuel inventory exists out of the core, in the primary loop. Assuming a homogeneous distribution of the delayed neutron precursors, 0.0009 must be subtracted from the calculated eigenvalues, since 33% of the delayed neutrons are emitted in the primary loop. In reality this number is even smaller since the concentration of short-lived precursors is higher in the core than in the primary loop.

Since the neutronic calculations are performed on a unit-cell its results apply to an infinite core. To give a prediction about the k_{eff} , the non-leakage probability was calculated for a non-reflected cylindrical finite core with volume 91m³ on the basis of the diffusion length L and geometrical buckling B of the core (see Eq. 1).

$$P_{NL}=(1+L^2B^2)^{-1} \quad (1)$$

Depending on the moderation ratio ($V_{\text{moderator}}/V_{\text{fuel}}$) the thermal power of the core varies from 570 to 415 MW. The non-leakage probability is very close to unity (>0.99) for all moderation ratios. Taking this into account, the minimum requirement for k_{∞} is chosen to be 1.02.

3.2. Calculation procedure

In general, the scheme is the following. First, the criticality, the temperature feedback coefficients,

the spectrum and weighted cross sections are calculated with the CSAS sequence on the basis of the initial nuclide concentrations. Then the COUPLE code is used to prepare the working library for ORIGIN from the weighted cross sections and ORIGIN master libraries. The evolution of the materials is computed using the ORIGIN-S code. The fuel is not irradiated while it is outside the core. The short irradiation and decay steps are averaged as a continuous irradiation with a reduced flux level. The removal of elements (chemical processing) can be simulated by adding a pseudo-decay constant. The burnup calculations are stopped regularly and the nuclides which are most important for the criticality calculations are collected. This means that 60 to 100 fission products and actinides are added to the list of initial nuclides. New CSAS calculations are started with the updated nuclide concentrations and a new ORIGIN working library is prepared on the basis of the new spectrum and reaction rates. The ORIGIN calculations are started again with the new working library. For each calculation an off-gas system is simulated which extracts gaseous and non-soluble metallic FP as the first step of salt processing.

3.3. Calculation of the breeding ratio

The spectrum in the core must be well tailored to achieve breeding in a moderated system. To be able to choose the proper salt, the differential breeding ratio at 100 days after startup was calculated for each salt. Since the candidate salts have different moderation effects themselves, first the needed amount of graphite moderator was calculated for each of them. The goal was to reach a breeding ratio >1. The breeding ratio is calculated in a differential way with the following simple expression (see Eq. 2),

$$BR = (R_{c(Th)} - R_{c(Pa)}) / R_{a(U)} \quad (2)$$

where R_c and R_a represent capture and absorption reaction rates. The concentrations of ^{232}Th , ^{233}Pa and ^{233}U were obtained 100 days after the startup of the reactor when the ^{233}Pa has reached its equilibrium concentration. The ORIGIN working library was updated after each 25 days. The concentration of ^{233}U is not corrected to reach criticality. The result of this calculation is the 'differential' BR of the reactor at 100 days after the start-up.

3.4. Burnup calculations

For the long-term calculations, the above scheme is performed without any other calculations added. The ORIGIN working library is updated every 5 days. Only the gas bubbling system, as processing method, is simulated.

Moving on to the more complex on-line processing, in the first schemes only FP are removed. The following step is to remove also minor actinides with the FP. As the last step, the removal of Pa is introduced. It is increasing the breeding since the Pa has relatively long half-life (27 days) and may easily capture a neutron before decaying to U. The removed Pa is stored and the U produced by decay is fed back to the core to restore criticality, if needed. The extracted amount of Pa and added amount of U are tracked to see if more U is needed than Pa was extracted.

In case of batch-wise or on-line continuous processing, besides the removal of nuclides, feeding of the reactor with molten salt is needed to keep the mass of the salt in the reactor constant. This is performed by a home-made code which changes the nuclide concentrations in the binary result file of ORIGIN while the CSAS and COUPLE code update the working library. These stops are made in every 5 days to avoid sudden increases in the concentration of the nuclides fed in the reactor. Since the concentration of the ^{233}U is decreasing during the start-up of the reactor until the equilibrium concentrations of ^{233}Pa and ^{233}U are reached, the flux is increased in each step to keep the same power level.

3.5. Salt compositions

Nine different molten salt compositions were investigated, given in Table 1. For each type of salt, 12 mol% of $^{232}\text{ThF}_4$ and 0.3 mol% $^{233}\text{UF}_4$ were added, as proposed for the MSBR. In each calculation the power density in the salt is set to 40 MW/m³. In the neutronic calculations the temperature of the fuel is assumed to be 1000 K. For each salt, the breeding ratio at 100 days after startup and the initial k_{∞} were calculated for various moderation ratios ($V_{\text{moderator}}/V_{\text{fuel}}$), ranging from 3.2 to 5.5. The density of graphite was set to 1.84 g/cm³.

Table 1
Composition of the studied molten salts

Molten salt mixture	Composition (mol%)
LiF-BeF ₂ (FLIBE)	67-33
LiF-BeF ₂ -ZrF ₄	64.5-30.5-5
LiF-NaF-BeF ₂	31-31-38
LiF-NaF-ZrF ₄	42-29-29
LiF-ZrF ₄	51-49
LiF-RbF	44-56
NaF-BeF ₂	57-43
NaF-ZrF ₄	59.5-40.5
RbF-ZrF ₄	58-42

4. Results

4.1. Breeding ratio

Figure 1 shows k_{∞} and BR as a function of moderation ratio for LiF-BeF₂. The gray arrow shows the range where the criteria for criticality and BR are fulfilled. The k_{∞} is increasing with the moderation ratio because this is still the under-moderated region. On the other hand, the BR is decreasing, showing that more moderation reduces the resonance capture rate of Th. In a harder spectrum (using a low amount of graphite), the absorption rate of thorium in the epithermal range is increasing while the fission rate of the uranium does not change. Of course the high breeding ratios (>1.2) are not physically possible since these correspond to subcritical setups. If the concentration of fissile material would be corrected to reach criticality in these cases, the BR would be significantly lower.

There is a small range of moderation ratio from 3.9 to 5.25 where the criteria for k_{∞} and BR are both fulfilled. For the other salts a very similar figure results. The LiF-BeF₂ has the best results in terms of k_{∞} and the corresponding BR. The other suitable salts are also lithium based: LiF-BeF₂-ZrF₄ and LiF-ZrF₄. Some sodium containing salts - LiF-NaF-BeF₂, LiF-NaF-ZrF₄ and NaF-BeF₂ just exceed the criteria for criticality in a breeder setup while the other salts cannot reach BR>1 in critical conditions. Our choice for the further studies is LiF-BeF₂ with

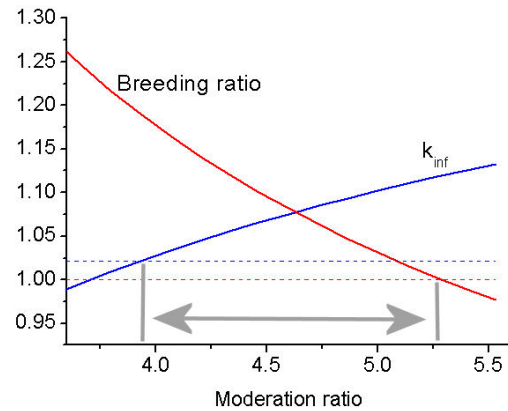


Fig 1. BR and k_{∞} as a function of moderation ratio

the amount of graphite corresponding to $V_{\text{moderator}}/V_{\text{fuel}}=5$. This composition has high k_{∞} and BR slightly higher than unity.

4.2. Temperature coefficients

The temperature coefficient of the core is an important safety parameter. It can be split into the effect due to the salt and the effect due to graphite. The temperature feedback coefficient of the salt can be positive or negative while the feedback of the graphite is always positive, ranging from 1 to 2 pcm/K depending on its amount. This effect comes from the shift of the thermal neutron spectrum due to the increase of temperature. The η (average number of fission neutrons produced per neutron absorbed in fuel), calculated for the mixture of the three most abundant actinides (²³²Th, ²³³Pa, ²³³U) is increasing above 0.2 eV (see Fig. 2) so the hardening of the thermal spectrum causes an increase of reactivity.

To achieve a core with negative temperature feedback, the feedback effect of the salt must be strongly negative to compensate for the positive contribution of the graphite. The effect of the salt is the sum of two different phenomena, the Doppler-effect and the effect of the change in the density of the salt. The Doppler-effect may be positive or negative for the nuclides in the salt. The Doppler-effect can also be split depending on the type of the corresponding interaction. The effect due to the absorption cross-section change is negative, but weak for most of the nuclides. A change in the scatter cross-section of light nuclides has a positive

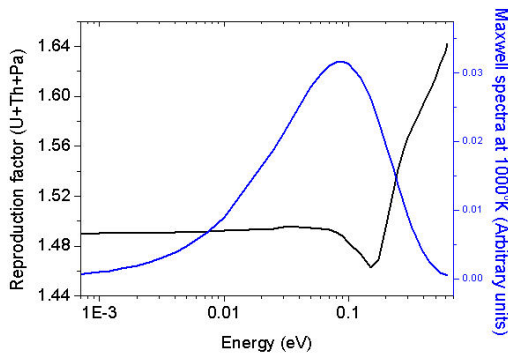


Fig 2. Neutron reproduction factor η of the ^{232}Th , ^{233}Pa , ^{233}U mixture (black) and the thermal Maxwell spectrum (blue) as a function of energy (eV).

effect. The positive effect of the graphite can be considered as a reactivity effect due to its scatter cross-section change. In case of a fissile nuclide the change of the fission cross-section implies a positive reactivity change. Summing up these effects, the light nuclides have a positive contribution to the Doppler-effect of the salt because the effect due to the change of their scatter cross-section exceeds the negative contribution of the absorption cross-section. The effect due to the fissile cross-section change makes the contribution of the fissile materials positive as well. Only the heavy nuclides like Th and Pa have a negative effect because the reactivity change implied by the change of their absorption cross-section determines their contribution. In fact, the first resonance of the thorium absorption cross sections, which implies a decrease of reactivity, makes the Doppler coefficient negative for the whole salt.

The composition of the molten salt strongly affects the temperature feedback coefficient of the fuel salt since nuclides with high absorption cross section have strong positive effect on the feedback due to the change in their atomic density. Only fissile nuclides, mainly ^{233}U in this case, have negative influence on this feedback effect. The sum of the two contributions is negative for each investigated salt mixture and strong enough to compensate for the positive effect of the graphite at a moderation ratio of 5. The total temperature coefficient increases as the moderation ratio increases and turns into positive for each salt at high moderation ratios (>6).

5. Results of long term calculations

5.1. Generalities

A simple start-up procedure was chosen: the initial load of the reactor is pure ^{233}U . The composition of the fuel salt is $\text{LiF-BeF}_2\text{-ThF}_4\text{-UF}_4$ 58.8-28.9-12-0.3 mol%. The start-up could be performed with ^{235}U or a mixture of higher actinides produced in light water reactors. The mass of FP and MA removed from the system is compensated by feeding the reactor with $\text{LiF-BeF}_2\text{-ThF}_4$ mixture or with $\text{LiF-BeF}_2\text{-ThF}_4\text{-UF}_4$ if Pa is extracted. In this case the ^{233}U comes from the decay of the removed ^{233}Pa . The composition of the salt changes quickly during the first year after start of the reactor. This is caused by the loss of initial fissile inventory during the start of the thorium fuel cycle and the build-up of FP and MA. The removal of some FP is necessary, since otherwise their high absorption rate stops the fission chain quickly.

The flux used for burnup calculations is set to be equal in each calculation to have the 40 MW/m³ power density in the salt. In the neutronic calculations the temperature of the fuel is assumed to be 1000 K.

5.2. Batch-wise processing

The first case represents a reactor with off-gas system as the only continuous FP removal process. The off-gas system removes noble gas and noble metal FP with a cycle time of 30 seconds. The FP and MA accumulated during the operation need to be removed regularly since the reactor becomes subcritical due to the elevated neutron absorption of the salt if no fissile material is fed into the core. As the concentration of FP is high, the whole inventory is processed as one batch, removing most or even all of the FP.

Fig. 3 illustrates the evolution of ^{233}U and the criticality of a $\text{LiF-BeF}_2\text{-ThF}_4\text{-UF}_4$ fuelled reactor with moderation ratio 5 for three years from startup. In the first 100 days the k_{∞} and the ^{233}U density drop quickly until the ^{233}Pa reaches its equilibrium concentration. Although the decrease of the ^{233}U amount slows down and starts to grow slowly after 500 days, the k_{∞} constantly decreases and reaches 1.02 – the minimal requirement set for k_{∞} – after 620 days. The ^{233}Pa and ^{233}U density is 1770 and 31560

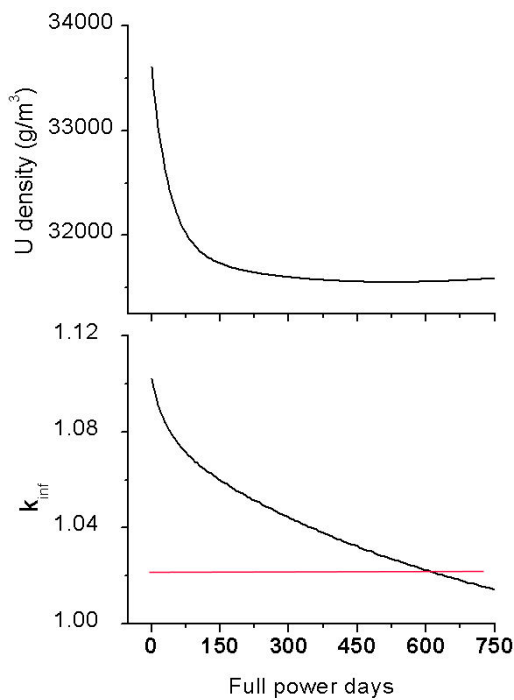


Fig 3. ^{233}U density and k_{∞} versus full power days

g/m^3 of salt, the ^{233}U density is lower with $2050 \text{ g}/\text{m}^3$ than it was at startup. At this time the chemical processing of the salt has to happen. Since the equilibrium concentration of ^{233}Pa is lower than the loss of ^{233}U , even if the chemical processing of FP and MA has 100% efficiency and the salt is completely cleaned, the initial reactivity of the next startup is lower. It is assumed that the reactor is shut down for 30 days for maintenance and processing of the salt. During this time 53.7% of the ^{233}Pa decays to ^{233}U and the rest remains in the salt. If the reactor is started up with this fuel, its reactivity is 24.5% lower than it was at the first startup. At least a part of the ^{233}U loss must be compensated or the second campaign will be shorter than the first, only 260 days. The total temperature coefficient of the core is $-2.46 \text{ pcm}/\text{K}$ at the start, which changes to $-2.12 \text{ pcm}/\text{K}$ after 620 days due to the absorption of FP accumulated in the salt.

As a possible solution the power level was changed to 20 and $80 \text{ MW}/\text{m}^3$. As other studies revealed, a lower neutron flux has numerous advantages and increases the breeding ratio while higher flux reduces breeding. In case of the lower neutron flux, the capture rate of actinides is lower, the ^{233}Pa produces less ^{234}U and more ^{233}Pa decays

to ^{233}U and less MA are formed. On the other hand the size of the core and the inventory must be doubled if the total power is kept constant.

Employing the low flux level, the reactor becomes subcritical after 1730 days. Using the same method as in the former case, the reactivity is 14.8% lower at the second startup. The temperature coefficients at the start and shutdown of the core are the same in the former case.

5.3. Continuous processing

Several processing schemes were studied for a breeder MSR but only a few describe the intended chemical processes to extract FP. All schemes include the off-gas system and the quick removal and re-introduction of U from the processed salt via fluorination. This process must be as quick as possible and the extracted U must be returned into the core immediately or else the U spends too much time out of the core and its inventory would rise. At this point the schemes diverge, some suggesting the storage of fuel for several months to let the ^{233}Pa decay to ^{233}U and employing an off-line method to remove FP and MA after a second fluorination to extract the formed ^{233}U . Other schemes, like the one designed for MSBR, use only on-line processes to extract Pa, FP and MA. During the design of the MSBR the goal was to reach the highest breeding ratio possible. The on-line chemical processing unit proposed for the MSBR is considered unfeasible nowadays mainly because it has to process the whole salt volume in 10 days with very complex processes. Recent studies (Mathieu et al., 2003) recommend to slow down the processing and treat the salt volume in months or even years.

Table 2.
Fast reprocessing scheme

Component	Cycle time
Kr, Xe, Se, Nb, Mo, Sb, Te, Ru, Rh, Pd, Ag, Tc	20 sec
Y, La, Ce, Pr, Nd, Pm, Gd	50 days
Eu	500 days
Zr, Cd, In, Sn	200 days
Br, I	60 days

Table 3.
Slow processing scheme

Component	Cycle time
Kr, Xe	50 sec
Zn, Ga, Ge, As, Se, Nb, Mo, Cd, In, Sn, Sb, Te, Ru, Rh, Pd, Ag, Tc	2.4 h
Zr, Ni, Fe, Cr, Y, La, Ce, Pr, Nd, Pm, Gd, Tb, Dy, Ho, Er, Sm, Eu	1 or 3 years

In our study we consider two types of processing, one scheme close to the one proposed for the Molten Salt Actinide Recycler & Transmuter (MOSART) (Ignatiev et al., 2007) and the MSBR processing scheme (see Table 2.) for comparison. We choose the basis of the MOSART scheme (see Table 3), because it is detailed and feasible from a chemical point of view. The first two groups of nuclides are treated by the off-gas system while the last group (Lanthanides & Zr, Ni, Fe, Cr, Y) is extracted by different chemical processes depending on their chemical properties.

Calculations with both schemes were performed where two cases considered for the slow processing scheme. The first case uses a 1 year cycle time for the last group (case 1) while the second is even slower with a 3 year cycle time (case 2). Pa and the MA are not removed in these schemes. As the results show (see Fig. 4) even the faster removal of the slow processing scheme (case 1) is not effective enough to stabilize the k_{∞} . The piling up of FP stops the chain reaction for both cases after 1550 and 765 full power days, respectively. The total temperature coefficients drop from -2.46 pcm/K to -2.23 and -2.13 pcm/K at the shutdown of the reactor. The fast processing scheme shows its potential even without the removal of Pa and MA, the decrease of k_{∞} stops after 2000 days and stabilizes at 1.032. The fast FP extraction would be enough to sustain a self-breeder reactor if such a scheme would be feasible.

The extraction of TRU elements (Np, Pu, Am, Cf) with 1 year cycle time was also considered with the faster Lanthanide removal case of the slow reprocessing scheme (case 1). The lower concentration of TRU is the only effect of this extraction; the calculated eigenvalues, the U density or the temperature coefficients do not change during the first five year operation of the investigated reactor.

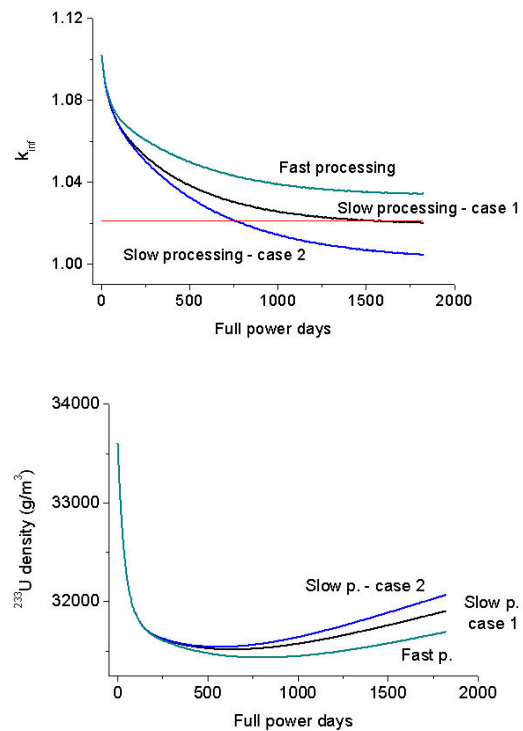


Fig. 4. k_{∞} and ^{233}U inventory as a function of irradiation time for case one (black) and case two (blue) and fast processing (green) schemes

All these results lead to the conclusion: it is difficult to operate a self-breeder reactor without the extraction of. The Pa removal is a key point in a breeder MSR. To reach its surpassing 24 years doubling time the MSBR was supposed to operate with 3 days Pa cycle time. This can be strongly reduced in case of a self-breeder reactor. In our code the Pa is continuously removed from the core and a part of the formed U is re-inserted to keep the calculated k_{∞} between 1.028 and 1.03, if necessary. Other nuclides are removed as in case 1.

Because of the chosen start-up method of the fuel cycle and the feeding, the ^{233}U stock increases rapidly after the start of the reactor. On the other hand the ^{233}U density in the core decreases and the k_{∞} drops quickly since a large part of Pa decays outside the core. As k_{∞} reaches 1.03 a part of ^{233}U stock is re-introduced regularly into the core to maintain the criticality. From this point the ^{233}U stock starts to decrease until the ^{233}U density in the core reaches its initial value and approaches its new equilibrium value. As the ^{233}U density increases, less ^{233}U is needed to be re-inserted to sustain criticality. If the

stock of the removed ^{233}Pa is large enough, the amount of the ^{233}U formed exceeds the amount fed back to the core. In this case the reactor is a breeder, otherwise the reactor uses the whole ^{233}U stock and additional U is needed to keep the reactor critical. The amount of stock Pa depends on its extraction speed.

Calculations were performed with four different Pa cycle times (see Table 4.). The results show that this parameter has a strong influence on the breeding capability of the reactor. The 6 months Pa cycle time results a reactor close to a self-regenerating system.

Table 4.
Doubling times for different Pa cycle times

Pa cycle time	Produced ^{233}U per year (g/m^3 salt)	Doubling time (years)
27 days	908	40
4 months	287	123
6 months	258	146
1 year	-287	-

6. Conclusions

In this paper we investigated the breeding capability of a moderated MSR operating on the thorium fuel cycle. The impact of different molten salt compositions on the breeding ratio, moderation ratios, power density and different salt processing strategies were studied. The main results obtained are as follows:

- From the numerous binary and ternary LiF, NaF, BeF_2 , ZrF_4 and RbF molten salt mixtures only a few can be used in a self-breeder MSR. The most promising is LiF- BeF_2 (FLIBE) because of its low parasitic neutron capture. This molten salt was also chosen for the MSBR.
- The moderation ratio has a strong influence on the criticality, breeding ratio and total temperature coefficient.
- It is possible to have a negative total temperature coefficient in a moderated MSR without adding any burnable poisons if the moderation ratio is low.
- Employing batch-wise processing, a high conversion ratio is achievable in the MSR

but more FP must be extracted from the core to obtain a self-regenerating system.

- A slow removal of FP and MA is not sufficient to sustain a self-breeder reactor. The extraction of Pa and the re-introduction of the formed U is necessary.
- The Pa extraction rate has a strong influence on the doubling time of the reactor. 1 year cycle time of soluble FP and 6 months Pa cycle time is enough to sustain a self-breeder MSR.

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