

New breeding gain definitions and their application to the optimization of a molten salt reactor design

K. Nagy*, J.L. Kloosterman, D. Lathouwers, T.H.J.J. van der Hagen

Delft University of Technology, Department of Radiation, Radionuclides and Reactors, Physics of Nuclear Reactors, Mekelweg 15, 2629JB Delft, Netherlands

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ABSTRACT

The molten salt reactor (MSR) is an attractive breeder reactor. A graphite-moderated MSR can reach breeding because of the online salt processing and refueling. These features give difficulties when the breeding gain (BG) of the MSR is evaluated. The inventory of the core and external stockpiles have to be treated separately in order to quantify the breeding performance of the reactor. In this paper, an improved BG definition is given and it is compared with definitions used earlier. The improved definition was used in an optimization study of the graphite – salt lattice of the core. The aim of the optimization is a passively safe, self-breeder reactor. The fuel channel diameter, graphite volume and thorium concentration were varied while the temperature feedback coefficient of the core, BG – as defined in the paper – and the lifetime of the graphite were calculated. There is a small range of lattices which provide both negative temperature feedback and breeding. Furthermore, breeding is possible only at low power densities in case of the salt processing efficiencies set in this study. In this range of power the lifetime of the graphite is between 12 and 20 years.

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1. Introduction

In the molten salt reactor (MSR) the fuel – a fluorine salt of actinides – is dissolved in a molten salt carrier. This mixture of salts acts as coolant as well. The fluid fuel is a unique feature of the reactor which provides exceptional transmutation and breeding capabilities because the composition of the salt can be continuously altered by chemical processes during operation. This way the concentrations of various nuclei – actinides and fission products – can be tailored to optimize transmutation and breeding. The MSR has other advantages on fuel fabrication, neutron economy and high temperature operation as well due to the low absorption, high boiling point and low vapor pressure of the salt. This study focuses on an MSR which is a possible breeder utilizing the thorium fuel cycle.

The fertile isotope of the thorium fuel cycle is ^{232}Th which forms ^{233}U after a neutron capture and 2 beta decays. The intermediate products are ^{233}Th and ^{233}Pa . The half-life of the two isotopes is 22.3 min and 27 days, respectively. The dynamics of this fuel cycle differs from the uranium cycle because of the relatively long half-life of ^{233}Pa , while ^{239}Np , the isotope in the same position in the uranium fuel cycle has a half-life of 2.35 days. This long half-life of ^{233}Pa results in a high probability of capturing a neutron before the protactinium decays to uranium. If ^{233}Pa captures a neutron

before it decays, the formed isotope will be ^{234}U , a non-fissile isotope. To avoid this, it is possible in a breeder MSR to extract part of the protactinium from the salt during operation and to store it outside the core (Robertson, 1978). All the removed ^{233}Pa will decay to ^{233}U . In this way the ^{233}U production can be enhanced. Moreover, external stockpiles of protactinium and uranium are formed which are coupled to the reactor. The protactinium stockpile obtains its feed from the reactor while a part of the ^{233}U formed in the stockpile can be sent back into the core to maintain criticality if needed.

The history of the molten salt reactor dates back to the 1950s. The design was first proposed as the propulsion system of a nuclear-powered aircraft at the Oak Ridge National Laboratory (MacPherson, 1985). After the program finished, the emphasis was put on the research of an MSR running on the thorium fuel cycle. In the 1960s the project focused on the breeding possibilities, resulting in the design of the Molten Salt Breeder Reactor (MSBR) (Robertson, 1978). An experiment, called molten salt reactor experiment (MSRE) (Prince et al., 1968) was carried out from 1965 to 1969. The reactor was operated with ^{233}U in early 1969. This was the first time ^{233}U was used as reactor fuel. The salt of this reactor did not contain any thorium because it was intended to simulate only the fuel stream of a two-fluid breeder reactor. The MSR program was terminated in 1976 although the results of the experiment were promising. The MSR without the complicated chemical removal processes is a converter reactor with high conversion ratio if thorium is added to the fuel (Perry, 1975). The ORNL studies were used as a basis for the molten salt reactor design FUJI

* Corresponding author.

E-mail address: k.nagy@tudelft.nl (K. Nagy).

in Japan in 1985. From the numerous variations of the design, the 200MWe FUJI-U3 (Mitachi et al., 2007) can operate on the thorium fuel cycle with a high conversion ratio. The simplification of the original MSBR design was the direction of the research in France as well (Vergnes and Lecarpentier, 2002) and as a recent study showed (Renault et al., 2005), the simplification of the salt purification processes is a key issue in the development of the MSR. An extensive study is carried out in France at Laboratoire de Physique Subatomique et de Cosmologie (LPSC). First, a design of a moderated reactor was developed on the basis of the MSBR (Mathieu et al., 2006, 2009); nowadays the project focuses on a fast breeder reactor (Merle-Lucotte et al., 2008a, 2009).

Many breeding ratio (BR) and breeding gain (BG) definitions have been used for fast breeder reactors (Ott and Borg, 1980). Some of these may give misleading results if they are applied to an MSR employing protactinium extraction. In this paper an improved description of the dynamics of the fissile fuel of such a system is proposed. This description is related to the mass growth rate of the fuel in the core and in the external fuel stockpile. Breeding gain definitions – separately for the core and the stockpile – are introduced on the basis of the mass growth rates as it was originally done for fast breeder reactors (Harms, 1978). Separate BG have not been defined for the stock and the stockpile of the MSR before, although it is necessary for the proper discussion of the fuel evolution (Nagy et al., 2010). The breeding gain definitions are used as one of the performance parameters of an optimization study which is intended to result in a design of a self-breeder moderated MSR with as high power density as possible at set processing rates. In this study the core geometry (amount of graphite and the diameter of fuel channels) is altered rather than the removal rates of various elements. This option was investigated by several authors (Mathieu et al., 2006; Merle-Lucotte et al., 2008b).

2. General description of the reactor and computational methods

2.1. Choice of the molten salt

In a breeder molten salt reactor the fuel consists of UF_4 as fuel and ThF_4 as fertile isotope. The fluorides of these actinides are dissolved in a mixture of LiF , BeF_2 , NaF , KF or ZrF_4 . These fluorides of metals act as diluents and the main reason to use their mixture is to keep the melting point of the whole salt low enough. The salt is also the heat transport medium and flows to the heat exchangers. The fluoride salts are chemically stable and can withstand intense radiation. Their vapor pressure is low at the operating temperatures of the reactor and well-chosen salt mixtures have sufficient solubility for actinides. Furthermore, fluoride salts are chemically compatible with graphite and with nickel-based alloys. For the moderated self-breeder MSR in the scope of this study the 2–1 mixture of LiF and BeF_2 (FLiBe) was chosen because of its low neutron capture cross-section. To reach this good neutron economy the lithium has to be enriched to 99.995% in ^7Li . In the optimization study the concentration of ThF_4 is varied between 12 mol% and 30 mol%. In the range between 22 and 30 mol% the melting point of the LiF – ThF_4 mixture is sufficiently low (850 K) (Benes and Konings, 2009). Substituting the beryllium with lithium does not significantly change the results of this study.

2.2. Moderator

Graphite is a suitable moderator for the MSR; its low absorption helps to achieve good neutron economy. No metals are used in the core because the salt does not wet the graphite and does not penetrate its pores. Graphite is subject to radiation damage, it

Table 1

The applied mean residence times of different elements.

Gaseous	Non-soluble	Soluble	Protactinium
50 s	2.4 h	1 year	3 months

undergoes significant volume changes, at first shrinking and later on expanding. For fast neutron fluences greater than $3 \cdot 10^{22}$ n/cm² of energy higher than 50 keV the rate of expansion becomes rapid and this is the upper limit is used since the ORNL studies (Robertson, 1978) till our days (Mitachi et al., 2007).

2.3. Chemical purification processes and actinide feed

In the MSR, fission products and actinides can be removed from the salt during operation. The solubility of noble gases and noble metals is very low. The major part of these elements is continuously removed from the salt by sparging with helium. The removal of soluble fission products and actinides is accomplished in a coupled processing plant. Removal of lanthanides is necessary to reach breeding in the reactor and protactinium removal and storage further increases the amount of produced fissile material. The mean residence time of different elements were chosen according to recent studies (Ignatiev et al., 2007; Mathieu et al., 2003; Delpech et al., 2009). The mean residence times are listed in Table 1. In the calculations all the uranium used to refuel the reactor is assumed to be extracted from the external stockpile and all the uranium produced by the decay of the extracted protactinium is placed in the stockpile. Furthermore, the thorium mass is kept constant by online feed in each calculation.

The efficiency of the purification processes is not changed during the study. Different efficiencies would have an impact on the presented results because quicker removal of both soluble FP (lanthanides in this case) and protactinium increases the amount of produced fissile material. The effect of varying the mean residence times of FP and Pa was investigated by several authors (Mathieu et al., 2006; Merle-Lucotte et al., 2008b).

2.4. Reactor core

The reactor core consists of hexagonal graphite blocks with fuel channels at the center of each block. Both the size of the graphite blocks and the diameter of the fuel channels are altered in the optimization process. This way the amount of graphite in the core and the diameter of the fuel channels can be changed independently. The core is 5 m in diameter and in height which makes a volume of 98 m³. The volume of the fuel salt in the loop is assumed to be equal to that in the core. During the optimization the overall size of the core is kept constant. The mean operating temperature of the reactor is 1000 K.

2.5. Computational tools

The calculations presented in this paper were performed with the XSDRN and KENO neutron transport codes of the SCALE code package (SCALE, 2005) and an in-house developed burnup code which is able to take into account continuous removal and feed processes as well as the material evolution of stockpiles outside the core.

3. Evolution of fissile fuel in the MSR

3.1. Distinction between converter and breeder MSR

Long-term reactivity control is realized without burnable poisons because it is possible to add fissile nuclides to the core of

the MSR during operation. To compensate for the build-up of absorption caused by fission products, non-fissile uranium isotopes and trans-uranium elements, fissile fuel is added regularly to the salt. First consider a reactor without protactinium extraction. The reactor is a converter if uranium has to be added to maintain criticality and is a breeder if reactivity is increasing in the core. In the latter case some uranium has to be removed by fluorination to control reactivity. This removed uranium is the bred fuel.

If a part of protactinium is removed from the salt, uranium is produced both in the core and in the stockpile by decay. The ^{233}U produced in the core may not be enough to compensate for the increased absorption. This produced uranium has to be left in the core and more needs to be added from the stockpile. If the amount of the uranium produced in the stockpile exceeds the needs to maintain criticality, the remainder is the gain which is considered as bred fuel. On the other hand, if the amount of uranium produced in the core and the stockpile together is not enough to maintain criticality, addition of fuel from an independent source is necessary. Although the reactor produces more fissile material than it consumes, it is a converter in this case because it needs more fissile nuclides to compensate for FP and MA build-up.

3.2. Burnup calculations

As an illustration, the time variation of the mass of ^{233}U was calculated for three different cases, a converter with high conversion ratio, a self-breeder and a breeder reactor. In order to realize this different behavior, only the fuel channel diameter was changed. The results presented here – using three different diameters, 40, 1, and 8 cm, respectively – were chosen from the set of the results presented in Fig. 5.2. Each case was calculated using FLiBe as carrier salt with 12 mol% ThF₄. The reactor was started up with ^{233}U and UF₄ was added continuously to maintain criticality. In all cases the volume of graphite was 75% of the core volume. This resulted in 44, 1.1, and 8.8 cm hexagons sizes. The number of channels was different in each case – 39, 54, 154, and 979, respectively – but the total volume of the salt was kept the same. The efficiency of the removal processes, thus the mean residence time of fission products and protactinium, was the same as well. The total thermal power of the core was 490 MW.

3.3. Results

The results are shown in Fig. 1 for the three reactor types (converter, self-breeder, breeder). The mass of ^{233}U is continuously increasing in the core to keep the reactor critical. This increase is necessary because of the build-up of absorbers in the salt. The build-up of FP is slow because of the slow removal process (see Table 1) and the actinides take decades to reach equilibrium in the core even at higher power densities (Nuttin et al., 2005). The amplitude of the flux decreases because the total power production is kept constant. This decreases the ^{232}Th capture rate while the hardening of the spectrum increases it. The net effect is still negative, the capture rate is around 14% less after 50 years of full power operation compared to the initial core for all the three cases. Different diameters of the fuel channel result in spectrum changes which cause the differences in the mass of the initial load and all the subsequent loadings. The amount of ^{233}U in the stockpile shows different behavior in the three cases but all can be separated in three stages. At the start-up, there is no ^{233}Pa thus no uranium production in the reactor.

First, the ^{233}Pa in the core has to reach its equilibrium concentration in order to produce ^{233}U in the core. This takes around 3 months depending on the efficiency of the protactinium removal. During this time the reactor needs high feeding rate compared to the later stages. Second, because the residence time of protactinium

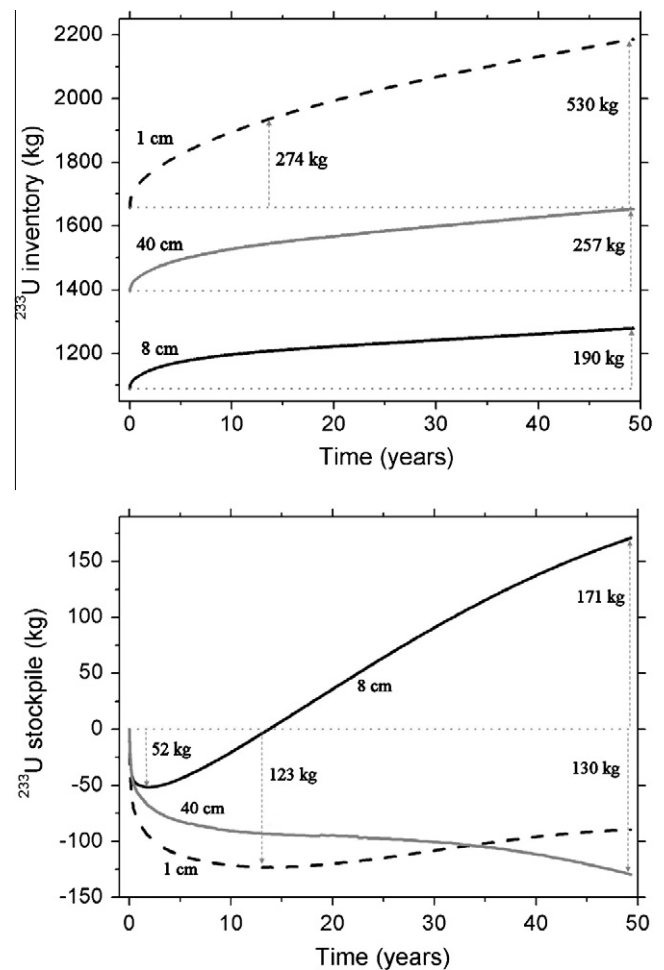


Fig. 1. Evolution of the mass of ^{233}U in the core (above) and in the stockpile (below) for the three studied cases. The inventory is determined by criticality control. Black, gray and dashed lines show the breeder, converter and self-breeder cases, respectively.

is longer than its half-life, the protactinium reaches its equilibrium mass in the external stockpile much slower than in the core. During this time only the core produces ^{233}U at the maximal possible rate. This takes around 9 months with the applied residence time. Once the protactinium reaches equilibrium in the stockpile, the reactor (the core and the stockpile together) is producing ^{233}U at its maximal capacity. After this point, depending on the amount of ^{233}U needed to maintain criticality, the reactor may turn to a breeder while it was a converter before. As the necessary feeding rate decreases with time the transition from a converter to a breeder may happen after a long time of operation.

The mass of ^{233}U in the stockpile is set to a starting value of 0. As the fuel loadings during operation are assumed to be taken from this pile and the ^{233}U produced by the decay of the extracted protactinium is placed here, negative values note that more uranium was extracted from the stockpile than produced. Similarly, positive values show the excess fissile material. The important points of Fig. 1 are compared in Table 2. The case with 1 cm channel diameter is close to self-breeding. Although the reactor does not produce enough ^{233}U in 50 years to replace the uranium used in the first 13 years of operation but reproduces it in a longer time. The reactor with the channel diameter of 40 cm is a converter, the mass in the stockpile is monotonously decreasing. In the case of 8 cm channel diameter the reactor is a breeder. It turns to breeding after 2 years and it reproduces the uranium used in the converter phase

Table 2
Comparison of the three cases presented in Fig. 1.

Channel diameter (cm)	1	8	40
Initial load (kg)	1658	1089	1396
Load after 50 years (kg)	2188	1279	1653
Minimum of the stock (kg)	–123	–52	–130
Stock after 50 years (kg)	–90	171	–130

in 14 years after the start-up. After this time the reactor produces excess fissile fuel.

4. New breeding gain definitions

4.1. From mass balance equations to breeding gain definitions

First the mass change rates in the core and the stockpile are defined. These help to understand the time variation of the fuel mass in both volumes. Two mass change rates are defined for the core, one for the increase needed for criticality and one for the actual production of the core. The difference between them is important because usually the two expressions are not the same due to the continuous uranium feed. This way three different mass change rates are introduced (two for the core and one for the stockpile). Breeding gains are defined on the basis of each of them using the consumption rate of the fissile isotopes in the reactor.

4.2. Mass change rates of the core

If the reactor is started up with ^{233}U the two significant fissile isotopes are ^{233}U and ^{235}U . During operation the uranium vector changes, after 50 years the composition is 0.678–0.212–0.076–0.034 ^{233}U – ^{236}U by weight in the case of the reactor with 8 cm channel diameter presented in Section 3.3. The rate of consumption of the fissile isotopes in a reactor can be defined as, with the reaction rate given below:

$$C(t) = R_a^{23}(t) + R_a^{25}(t) \quad (1)$$

$$R_i^j(t) = Vw^j N^j(t) \int \sigma_i^j(E) \phi(E, t) dE \quad (2)$$

In these equations the two digit superscripts denote the actinides with the last digit of their atomic and mass number ($90 \leq A < 100$), respectively while the subscript denotes type of interaction, a for absorption in this case. N represents the atomic density of isotope j . The correct treatment of the problem would be to integrate over the volume of the reactor and the stockpile. The simple multiplication with the volume (denoted as V in Eq. (2)) is sufficient since the flowing fuel continuously mixes in the primary loop and the plenums of the core. Correspondingly volume-averaged cross-sections are used. The flux is averaged over the volume of the core and weighted according to the residence times in the core and the loop. There are more ways to define the weights, w^j , in the reaction rate, depending on the choice the rate can count number of atoms, mass or ^{233}U equivalent mass of fissile isotope. In the first case the weight is 1 for every isotope, while in the second and third case the weight equals the mass of the isotope and the mass multiplied with some rate between ^{233}U and ^{235}U . The weight of the product is used for any production term. In this paper the third approach is used and the weights are defined by the following formula

$$w^j = \frac{\sigma^j - \sigma^{02}}{\sigma^{23} - \sigma^{02}} \cdot M \quad (3)$$

where M denotes the mass of the isotope. The σ^j for a certain nuclide is defined as the traditional reactivity weight (Salvatore, 1986):

$$\sigma^j = v^j \sigma_f^j - \sigma_a^j \quad (4)$$

The mass change rate of fissile uranium (^{233}U and ^{235}U) in a critical core is given as:

$$\left(\frac{dm(t)}{dt} \right)_{\text{critical}} = R_d^{13}(t) + R_c^{24}(t) + R_{fe}^{23}(t) - C(t) \quad (5)$$

where the subscript d denotes decay and the integral term in Eq. 2 is replaced by the decay constant of the isotope. The ^{233}U is produced by the decay of ^{233}Pa and a part of the ^{233}U produced in the stockpile is sent back to maintain criticality. This “feed” from the stockpile to the core is noted as fe in the subscripts. ^{235}U is produced by the neutron capture rate of ^{234}U . This expression counts all the production processes occurring in the core and an extra one, namely the mass change due to the feeding of the core from the uranium stockpile. This term does not explicitly depend on the amplitude of the flux, only on the decrease rate of reactivity in the core and it is set to reach the necessary mass growth rate to keep the reactor critical. Without this term the mass change rate counts only the production and consumption processes in the core:

$$\left(\frac{dm(t)}{dt} \right)_{\text{core}} = R_d^{13}(t) + R_c^{24}(t) - C(t) \quad (6)$$

The concentration of ^{232}Th is kept constant during the operation of the reactor and ^{233}Pa reaches equilibrium quickly both in the core and the stockpile. This takes about 4 months in the core and 1 year in the stockpile. After this time the Pa concentration changes slowly due to the slight shift of the spectrum of the reactor. The equations of Pa are introduced and used in Appendix A.

4.3. Limitations of the mass change rates defined for the core

These definitions are not useful for direct comparison of completely different cases of MSR because the efficiency of the chemical removal processes is incorporated in them. For a simple demonstration two identical MSRs with different efficiency of the removal processes will be considered.

If the removal of protactinium is quicker in one reactor, less ^{233}U will be produced in the core by decay; thus, the right hand side of Eq. 6 is smaller than in the case of slow removal. On the contrary the value of Eq. 5 does not differ for the two reactors because the decay of ^{233}Pa and the ^{233}U feed is supposed to cover the needed increase of uranium mass in order to maintain criticality. The mass of the uranium stockpile is different for the two reactors but it is not included in these definitions.

The removal efficiency of fission products can be different as well. In this case the value of Eq. 6 does not differ for the two reactors but the right hand side of Eq. 5 does. If the efficiency is low, the higher amount of absorbers in the salt has to be compensated by uranium, thus the feed rate is higher. This results in lower uranium stockpile as well but the stockpile is not included in the definition.

Based on these considerations we can conclude that these definitions can be used for direct comparison only if the efficiencies of the removal processes are the same for each case. Furthermore, these definitions need to be extended to overcome this drawback.

4.4. Mass change rate of the stockpile

It is an obvious choice to extend the growth definitions to the stockpile, defining the net gain in the uranium stockpile as:

$$\left(\frac{dm(t)}{dt} \right)_{\text{stock}} = R_d^{13\text{stock}}(t) - R_{fe}^{23}(t) \quad (7)$$

where the first term denotes the decay rate of ^{233}Pa in the stockpile while the latter term is the mass transfer from the stockpile to the

core. This expression contains the effect of different removal processes. The decay rate of ^{233}Pa in the stockpile is connected with its removal efficiency, and the mass transfer depends in the increase of absorption in the core thus the removal efficiency of fission products.

4.5. Breeding gain definitions

Differential breeding gain calculations count the balance of nuclear reactions increasing and decreasing the amount of fissile material in the reactor. These definitions, compared to those based on the ratio of weighted masses of fissile isotopes after subsequent irradiation cycles, suit the MSR since there are no inherent irradiation cycles in this type of reactor. Based on the mass change rates defined in Eqs. (5)–(7), breeding gains can be defined using the consumption rate as described in Eq. 1. The stockpile is considered to be a part of the reactor where the flux is negligible. This way the consumption rate is the sum of the consumption in the reactor core and in the stockpile. This assumption allows to use the same consumption rate for the core and the stockpile. The breeding gains are defined as the mass growth rate over the consumption rate:

$$BG_{critical} = \frac{R_d^{13}(t) + R_c^{24}(t) + R_{fe}^{23}(t)}{C(t)} - 1 \quad (8)$$

$$BG_{core} = \frac{R_d^{13}(t) + R_c^{24}(t)}{C(t)} - 1 \quad (9)$$

$$BG_{stock} = \frac{R_d^{13stock}(t) - R_{fe}^{23}(t)}{C(t)} \quad (10)$$

The first definition contains the feed term, and this term is always set to keep the reactor critical. Therefore Eq. 8 is the BG in the core needed to maintain criticality. Eq. 9 describes the amount of fuel the reactor core produces. This can not be directly observed in a real reactor. The last definition is the BG connected to the available fuel as all the excess from the core and the stockpile is supposed to be placed here. The mass change rates defined above can be expressed as the product of the corresponding breeding gain and the consumption rate.

Based on the BG definitions the doubling time of the reactor can be estimated. The reactor (core and stockpile together) needs a feed of fissile fuel until the stockpile reaches a minimum value; thus, the doubling time is time dependent (Harms, 1978). The first identical reactor can be started up when the amount of ^{233}U in the stockpile equals the initial ^{233}U mass in the core:

$$m = \int_0^{DT} \left(\frac{dm(t)}{dt} \right)_{stock} dt = \int_0^{DT} BG_{stock}(t)C(t)dt, \quad (11)$$

where m is the initial load of the reactor and DT denotes the doubling time. The integral is necessary because of the time variation of the BG after start-up. After the initial period the BG may be considered as constant and the equation can be simplified to calculate the equilibrium doubling time.

Fig. 2 shows the time behavior of the three BG definitions at start-up and at the transition from converter stage to breeder operation for the reactor described in Section 3.3 with 8 cm channel diameter. The fissile uranium isotopes are weighted to get ^{233}U -equivalent masses as described in 4.2. The $BG_{critical}$, Eq. 8, describing the growth rate of the mass in the core, is always positive as fissile fuel is continuously added to balance the decrease of reactivity due to the initial build-up of FP and the build-up of absorber actinides. On the other hand, this definition does not distinguish between the growth due to breeding or feeding while the BG_{core} , Eq. 9, counts only the fuel production in the core. This is negative during the whole time of operation meaning that the core alone is a converter. The BG of the stock starts from large negative values

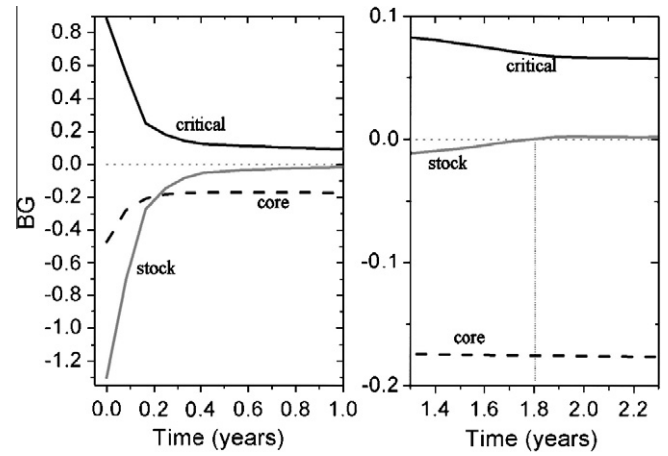


Fig. 2. Time variation of the three BG at start-up (left) and during the transition from converter to breeder operation (right).

since the large amount of ^{233}U needed to maintain criticality at the start-up of the reactor is assumed to be extracted from the stockpile. After the protactinium reaches its equilibrium level both in the core and in the stockpile, more uranium is produced than consumed and the stockpile starts to increase. As the reactor shifts to breeding from conversion the value of BG_{stock} turns to positive.

The presented BG in Eqs. (8)–(10) are differential BG definitions and not suitable to support the comparison of different cores. Integral definitions are more useful in this sense but the ones used for fast breeder (solid fuel) reactors cannot be used in this case (Nagy et al., 2010). The proposed BG definitions averaged over the operation time give integral BG definitions. As it was shown (Nagy et al., 2010), these averages directly relate to the change of fissile mass in the core or in the stockpile through the consumption rate Eq. (1) of the reactor.

As the breeding gain of the stockpile describes the evolution of the available fissile fuel, this definition is used in the following optimization study. In order to form an integral value which describes the whole lifetime of the reactor, the time average of Eq. 10 is calculated in each case. The relation between the BG definitions presented here and the BR definition used in earlier papers is discussed in Appendix A.

5. Optimization

5.1. Design conditions

The power density of the reactor investigated in Section 3 is low compared to other MSR designs (MSBR, FUJI, TMSR) and other reactor types. Therefore, an optimization study was carried out to maximize the power density of the reactor. In order to reach this goal, the graphite-salt lattice is optimized. Several constraints were applied: the core has to achieve self-breeding, long graphite lifetime and passive safety. The reactor core was assumed to have only one region. The parameters calculated for each case are the breeding gain defined in Eq. 10 averaged over 50 years of full power operation, the graphite lifetime and the temperature feedback coefficients for the graphite and the salt; while the fuel channel diameter, the volume ratio of the core (volume of graphite over the volume of the salt) and the amount of thorium in the salt were altered. Two limits were set on these parameters: $\overline{BG}_{stock} = 0$ and -1.5 pcm/K total temperature feedback coefficient. The total volume of the core was kept constant. The number of fuel channels was changed to fill the volume of the core as the parameters varied. The residence times of FP and Pa are listed in Table 1.

5.2. Effect of varying channel diameter and volume ratios

First, the effect of the channel diameter and the graphite volume on the BG of the reactor is demonstrated here. As the channel diameter was varied (Fig. 3) the volume ratio (volume of graphite over the volume of salt in the lattice) was set constant at 3. In the other case the volume ratio was altered (Fig. 4) while the channel diameter was kept at 6 cm. In both cases the concentration of the ThF₄ was 12 mol% in the salt. Each case was calculated for a core of 98 m³ and the thermal power of the core was set to 490 MW. The parameter used for the evaluation of both cases is the breeding gain of the stockpile, averaged over 50 years of full power operation. Each case has one maximum and the position of the maximum depends on the value of the parameter kept constant. The difference in the BG is caused by the changes in the spectrum, the production of uranium and the needed feeding rate changes as the channel diameter or the volume ratio is altered. Both maxima are in the region where the reactor has near zero or positive feedback coefficient (see Figs. 3 and 4). This observation is true in general, the best breeder lattices for different channel diameter or volume ratio have temperature feedback which is positive or close to zero. The possible channel diameter - volume ratio combinations, which are breeders and have sufficient negative feedback, are limited.

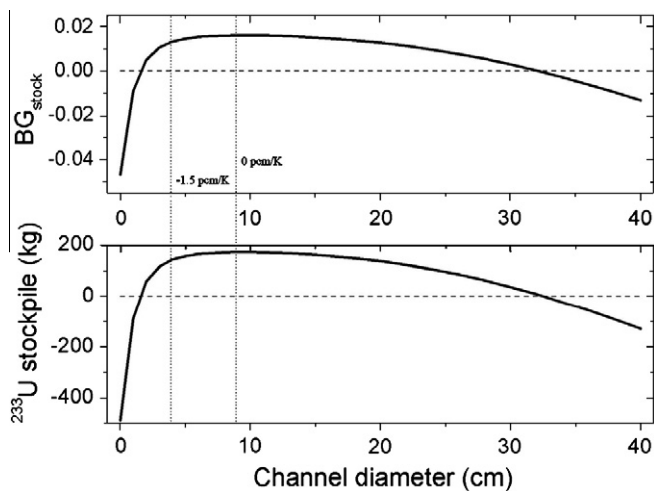


Fig. 3. Effect of the variation of the channel diameter on the breeding gain. Th concentration: 12 mol%, power density: 5 MW/m³, volume ratio: 3.

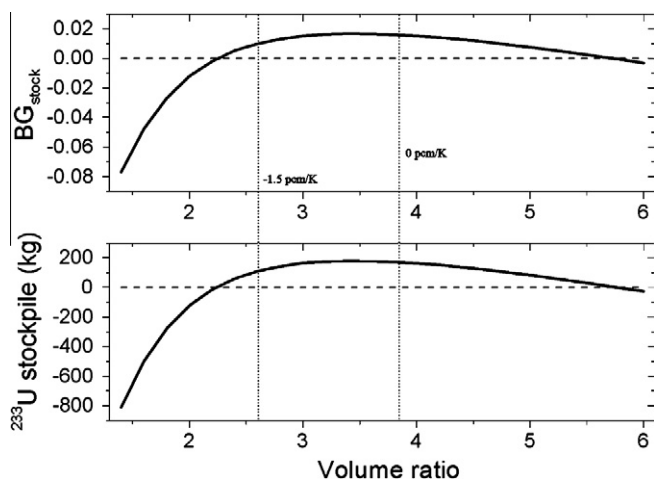


Fig. 4. Effect of the variation of the volume ratio on the breeding gain. Th concentration: 12 mol%, power density: 5 MW/m³, channel diameter: 6 cm.

5.3. Calculation procedure

The search for optimal self-breeder lattices is done in three steps. The first step is to find the graphite-salt lattices which have a feedback coefficient of -1.5 pcm/K at start-up. This is needed because the reactor cores which provide good breeding performance have unacceptable feedback coefficients. In the next step, the graphite lifetime and the breeding gain is calculated for these cores at an average power density of 5 MW/m³. This makes the total thermal power production of the core 490 MW. Finally, the power density is increased for each lattice to suppress breeding to self-breeding, resulting in higher power density. This is possible because the absorption rate of ²³³Pa is proportional to the flux while its decay rate does not depend on the flux. The lifespan of the graphite is re-calculated as well.

5.4. Temperature feedback

The first step in the optimization is to determine the range of lattices providing safety. In order to do this, the temperature feedback coefficient of different lattices has to be calculated. This is the sum of the effect of the salt and the graphite. The salt itself has negative feedback and this has to compensate for the positive feedback of the graphite. The cause of the positive feedback are the low energy resonances of the ²³³U fission cross-section (Nuttin et al., 2005; Mathieu et al., 2006). The main reason of the negative feedback of the salt are the ²³²Th capture resonances at 20 eV.

Temperature feedback coefficient of different critical lattices were calculated. The varied parameters were the volume ratio, channel diameter and Th concentration. First the k_{eff} for a temperature of 1000 K was calculated then it was calculated again at a higher (1100 K) reactor temperature. The thermal expansion of the salt was taken into account. Densities at different temperatures were calculated based on the temperature-dependent molar volume of the components assuming an ideal mixture. Thus, the dilatation coefficient differs for every salt mixture. This approach gives a dilatation coefficient of $6 \cdot 10^{-4}/K$ for FLiBe which contains 12 mol% ThF₄ and 0.24% UF₄. This value corresponds with the literature (Robertson, 1978). Dilatation of the graphite was neglected.

Fig. 5 shows the results with the margin of -1.5 pcm/K for three different Th concentration. This margin limits the possible channel diameter and volume ratio combinations. All the varied parameters, channel diameter, volume ratio and thorium concentration have effect on the temperature feedback. More homogenized cores – small fuel channels or less graphite volume – provide stronger negative feedback coefficients, as it was shown in the literature (Mathieu et al., 2009). Higher thorium concentration results in stronger negative coefficients in the same graphite structure. Changing the thorium concentration in the core changes the amount of berillium as well; thus, the spectrum hardens in the core if the thorium concentration is increased. This results in a less positive contribution of the graphite because the thermal peak is closer to the resonances causing this effect. The feedback of the salt becomes more negative as well. This is the sum of the Doppler and the dilatation effect and the decrease of the first to stronger negative values overrules the more positive effect of the thermal expansion. During the burnup the feedback coefficient changes, first it increases to -1.2 pcm/K and then starts to slowly decrease to stronger negative values.

5.5. Calculations with constant power density

Unfortunately, the best breeder configurations do not meet the safety criteria. As it was shown on Figs. 3 and 4, the lattice configurations on the -1.5 pcm/K margin give the highest possible BG. In

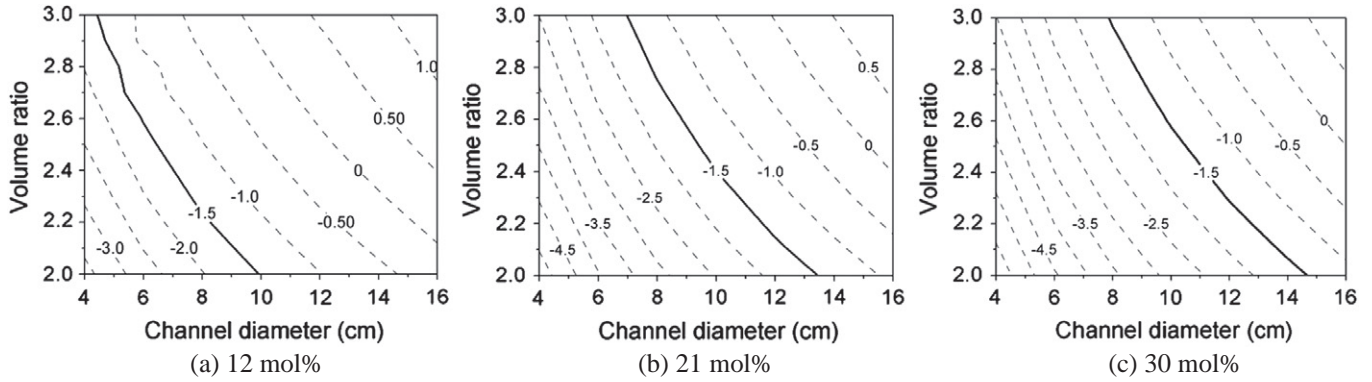


Fig. 5. Temperature feedback coefficient (pcm/K) as a function of channel diameter, graphite volume and thorium concentration.

the second step of the optimization, the volume ratio resulting -1.5 pcm/K feedback coefficient at start-up was determined for every channel diameter between 1 and 9 cm. Then, the graphite lifetime and the breeding gain of these cores was calculated with a constant 5 MW/m^3 average power density. Because there is only one region in the reactor, the peak power density is significantly higher than the average. The graphite blocks in the center have higher fluence than the ones at the side of the core. This can be overcome if the graphite blocks are reshuffled when the reactor is shut down for maintenance. This way the whole volume of

graphite reaches the irradiation limit at the same time. The lifetime of the graphite is calculated for both cases.

Table 3 shows the results of the calculations. All the studied cases are breeders at the power level of 5 MW/m^3 . Small channel diameters offer a gain on breeding performance and graphite lifetime due to their more thermalized spectrum. As the thorium concentration increases, the amount of graphite allowed by the safety margin increases as well thus compensating for the loss of the moderating components, Li and Be, in the salt. Higher fertile material content results in better breeding performance. The lifetime of

Table 3
Results with constant power density of 5 MW/m^3 .

Th conc (mol%)		Channel diameter (cm)								
		1	2	3	4	5	6	7	8	9
12	Volume ratio	4.7	4	3.5	3.1	2.8	2.6	2.4	2.25	2.1
	Initial load (kg)	955	977	1067	1172	1273	1347	1445	1527	1628
	BG ($\cdot 10^{-2}$)	1.960	1.842	1.663	1.433	1.186	1.001	0.732	0.505	0.203
	Lifetime (PP)	22.5	22.3	22	21.9	21.8	21.6	21.5	21.3	21.2
	Lifetime (AP)	36	35.5	35.2	35	34.8	34.6	34.4	34.2	34
15	Volume ratio	5.5	4.6	4	3.5	3.15	2.85	2.65	2.45	2.3
	Initial load (kg)	918	1011	1112	1242	1356	1484	1580	1702	1805
	BG ($\cdot 10^{-2}$)	2.562	2.414	2.244	1.99	1.733	1.401	1.17	0.838	0.569
	Lifetime (PP)	22.1	21.8	21.5	21.4	21.2	21.1	21	20.8	20.6
	Lifetime (AP)	35.3	34.7	34.4	34.1	33.9	33.8	33.5	33.3	33
18	Volume ratio	6.35	5.25	4.4	3.8	3.4	3.05	2.8	2.6	2.45
	Initial load (kg)	908	1006	1159	1317	1451	1608	1742	1869	1976
	BG ($\cdot 10^{-2}$)	2.966	2.861	2.65	2.346	2.059	1.664	1.33	1.014	0.762
	Lifetime (PP)	21.8	21.3	21.1	21	20.9	20.8	20.6	20.5	20.2
	Lifetime (AP)	34.7	33.9	33.7	33.5	33.3	33.2	33	32.7	32.3
21	Volume ratio	7.05	5.75	4.7	4.1	3.6	3.25	3	2.75	2.6
	Initial load (kg)	891	1021	1215	1364	1541	1696	1826	1995	2101
	BG ($\cdot 10^{-2}$)	3.231	3.186	2.929	2.665	2.281	1.92	1.646	1.22	1.025
	Lifetime (PP)	21.4	21	20.9	20.6	20.5	20.4	20.1	20	19.7
	Lifetime (AP)	34	33.4	33.3	33	32.8	32.5	32.2	32	31.6
24	Volume ratio	7.7	6.1	5	4.3	3.8	3.4	3.1	2.9	2.7
	Initial load (kg)	898	1055	1251	1432	1605	1790	1961	2088	2247
	BG ($\cdot 10^{-2}$)	3.497	3.425	3.173	2.853	2.497	2.085	1.703	1.468	1.131
	Lifetime (PP)	21.2	20.8	20.6	20.5	20.3	20.1	20	19.6	19.5
	Lifetime (AP)	33.8	33	32.8	32.5	32.3	32.1	31.9	31.4	31
27	Volume ratio	8.3	6.5	5.3	4.5	3.95	3.55	3.25	3	2.8
	Initial load (kg)	894	1065	1272	1481	1677	1860	2029	2202	2364
	BG ($\cdot 10^{-2}$)	3.684	3.634	3.394	3.032	2.635	2.254	1.918	1.571	1.27
	Lifetime (PP)	21	20.5	20.4	20.2	20.1	19.9	19.6	19.4	19.2
	Lifetime (AP)	33.5	32.6	32.4	32.2	32	31.7	31.4	31	30.7
30	Volume ratio	8.8	6.75	5.55	4.7	4.1	3.65	3.35	3.1	2.85
	Initial load (kg)	897	1098	1298	1516	1733	1948	2120	2293	2516
	BG ($\cdot 10^{-2}$)	3.835	3.788	3.574	3.204	2.785	2.332	2.019	1.699	1.238
	Lifetime (PP)	20.9	20.4	20.1	20	19.8	19.7	19.5	19.2	19.1
	Lifetime (AP)	33.1	32.5	32.1	31.8	31.6	31.4	31.1	30.7	30.4

PP – peak power.
AP – average power.
Lifetimes are given in full power years.

Table 4
Power density and corresponding graphite lifetime for self-breeder cores.

Th conc (mol%)		Channel diameter (cm)						
		3	4	5	6	7	8	9
12	Volume ratio	3.5	3.1	2.8	2.6	2.4	2.25	2.1
	Initial load (kg)	1067	1172	1273	1347	1445	1527	1628
	Power density (MW/m ³)	8.52	8.22	7.84	7.55	7.04	6.55	5.8
	Flux level ($\cdot 10^{14}/\text{cm}^2 \text{ s}$)	2.02	1.89	1.75	1.65	1.51	1.38	1.20
	Lifetime (PP)	13.1	13.5	14.1	14.5	15.4	16.4	18.3
	Lifetime (AP)	20.9	21.5	22.4	23.1	24.6	26.2	29.4
15	Volume ratio	4	3.5	3.15	2.85	2.65	2.45	2.3
	Initial load (kg)	1112	1242	1356	1484	1580	1702	1805
	Power density (MW/m ³)	9.75	9.5	9.19	8.71	8.36	7.71	7.11
	Flux level ($\cdot 10^{14}/\text{cm}^2 \text{ s}$)	2.28	2.13	2.00	1.84	1.74	1.56	1.42
	Lifetime (PP)	11.2	11.4	11.7	12.2	12.7	13.6	14.6
	Lifetime (AP)	17.8	18.2	18.7	19.6	20.2	21.8	23.3

PP – peak power.

AP – average power.

Lifetimes are given in full power years.

the graphite is comparable in every case, ranging from 19 to 22 years. On the other hand, the lifetime of the power plant is expected to be longer, thus the replacement of the moderator is necessary once or twice during the lifetime of the reactor.

5.6. Self-breeder reactors

The aim of this study is to find a core configuration which combines self-breeding, long graphite lifetime and passive safety. As it was seen in the last section, all the different reactors are breeders at 5 MW/m³ average power density. In the last step of the optimization, the breeder reactors from the last section were shifted to self-breeders. In this paper a reactor is considered self-breeder if it reproduces all the fuel used to keep the reactor critical after start-up. Numerically this means that the lifetime-average of the BG_{stock} has to equal 0. This shift was realized by increasing the power density in the core. The power density resulting in a self-breeder core was calculated for some cases in Table 3. The lifetime of the graphite is shown in Table 4 for the cases with low thorium concentration. The lifetime drops as the power density increases. At higher concentrations the lifetime is too short for the scope of this study. Ten to twenty years lifetime of the graphite can be achieved in the range of 5–10 MW/m³ power density. This is true for one region throughout the core. Further increase of the lifetime can be attained by dividing the core into more regions. This way the fast flux peak can be reduced in the center of the reactor. The limit for this is the flat power distribution in the core, and the corresponding lifetime is presented in the tables as well. Although the lifetimes with average power are significantly longer, these still do not meet the lifetime of a power plant.

6. Conclusion

In this paper the optimization of the graphite-salt lattice of a channel type, self-breeder MSR is shown. Various parameters were taken into account, such as the temperature feedback coefficient of the core, the breeding gain of the reactor, and the lifetime of the moderator graphite. The BG of the reactor was evaluated using a new definition, which is discussed in the paper as well.

In the first step of the optimization, both the fuel channel diameter and the graphite to salt volume ratio were altered. It was found that the temperature feedback coefficient of the core depends strongly on these variables and that the possible lattice configurations are limited by the restrictions on negative feedback. Furthermore, the lattices providing the best breeding performances have positive feedback and there is a small range of lattice

configurations which have both breeding and negative feedback. These lattices were compared for a wide range of thorium concentration in the salt at a set power density. The results show that breeding is possible both at low and high Th concentration. The lifetime of the graphite moderator – around 20 years – is shorter than the expected lifetime of the reactor; thus, the replacement of the graphite is necessary even at low power operation. Self-breeding was achieved by increasing the power density of the breeder configurations. This reduces the graphite lifetime further more. The graphite – salt lattices found in the optimization study combine passive safety, self-breeding and 10–20 years of graphite lifetime in the range of 5–10 MW/m³ average power density. These cores consist of only one region of graphite and an increase of graphite lifetime can be achieved by core designs with two or more regions. This is left for future work.

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Appendix A. Relation of the proposed BG definitions to the old ones

The breeding ratio of MSR was calculated by several authors (Nuttin et al., 2005; Mathieu et al., 2006; Nagy et al., 2008; Mathieu et al., 2009) in a simple form:

$$BR = \frac{R_c^{02} - R_c^{13}}{R_a^{23}} \quad (\text{A.1})$$

This definition is true only after the thorium and protactinium reached their equilibrium concentration so the time dependence is omitted. Breeding gain can be easily formed from this definition by subtracting 1 since the consumption rate is in the denominator. The extension to accompany ²³⁵U is straightforward:

$$BG = \frac{R_c^{02} - R_c^{13} + R_c^{24}}{R_a^{23} + R_a^{25}} - 1 \quad (\text{A.2})$$

This form can be compared with the BG definitions suggested in this paper. The main difference is the lack of the decay term of protactinium. Hereby will be proven that the numerator of Eq. A.1 equals the total decay rate of ²³³Pa in the core and in the stockpile together and that Eq. (A.2) (and Eq. A.1) gives the uranium production in the core and the stockpile together at equilibrium. In

order to do this, the Bateman equation of the ^{233}Pa is supplemented with a pseudo-decay describing the removal rate of protactinium through its mean residence time.

$$\frac{dN^{13}(t)}{dt} = \sigma_c^{02} N^{02}(t) \phi(t) - \sigma_c^{13} N^{13}(t) \phi(t) - \lambda N^{13}(t) - \frac{1}{\tau^{13}} N^{13}(t) \quad (\text{A.3})$$

In the equation direct production of ^{233}Pa by capture of ^{232}Th is assumed and the residence time is denoted as τ . At equilibrium, the numerator of Eq. A.1 equals the total decay of ^{233}Pa in the core:

$$\sigma_c^{02} N^{02} \phi - \sigma_c^{13} N^{13} \phi = \lambda N^{13} - \frac{1}{\tau^{13}} N^{13} \quad (\text{A.4})$$

Similarly a Bateman equation can be written for the ^{233}Pa extracted to the stockpile in which the only production term is the extraction from the core and the loss term is the decay of protactinium. At equilibrium, the two terms equal:

$$\lambda N^{13,stock} = \frac{1}{\tau^{13}} N^{13} \quad (\text{A.5})$$

Combining Eqs. (A.4) and (A.5) gives that the decay rate of ^{233}Pa in the core and in the stockpile together equals the numerator of Eq. A.1:

$$\sigma_c^{02} N^{02} \phi - \sigma_c^{13} N^{13} \phi = \lambda N^{13} + \lambda N^{13,stock} \quad (\text{A.6})$$

The total decay rate can not be found in the BG definitions in Eqs. (8)–(10) since those are defined separately for the core and the stock. BG describing the core and the stockpile has to be summed to obtain the right hand side of Eq. A.6:

$$\begin{aligned} BG &= BG_{critical} + BG_{stock} \\ &= \frac{R_d^{13}(t) + R_c^{24}(t) + R_{fe}^{23}(t)}{R_a^{23}(t) + R_a^{25}(t)} - 1 + \frac{R_d^{13,stock}(t) - R_{fe}^{23}(t)}{R_a^{23}(t) + R_a^{25}(t)} \\ &= \frac{R_d^{13}(t) + R_d^{13,stock}(t) + R_c^{24}(t)}{R_a^{23}(t) + R_a^{25}(t)} - 1 \cong \frac{R_c^{02} - R_c^{13} + R_c^{24}}{R_a^{23} + R_a^{25}} - 1 \quad (\text{A.7}) \end{aligned}$$

Eq. A.7 is the total increase of fissile material in the core and the stockpile together. It is concluded that the sum of Eqs. (8) and (10) equals Eq. A.2 as Eq. A.6 is applied as an approximation. This expression (Eq. A.7) does not distinguish between the increase needed to run the reactor and the increase of the excess material. Thus, Eqs. (A.1) and (A.2) describe the total increase of ^{233}U and the total increase of fissile uranium isotopes, respectively. In case of a reactor which employs Pa extraction, these definitions do not describe the evolution of the excess ^{233}U .

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