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Closing the PWR fuel cycle with a molten-salt incinerator

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We present a detailed study of a non-moderated Molten-Salt (MS) reactor, when operated as a transuranium elements burner, closing the nuclear fuel cycle of a PWR reactor. We evaluate its neutronic characteristics and main safety parameters, both in the first years of operation and in equilibrium fuel cycle. Influence of the carrier salt composition, neutron flux and feed composition on the burner performance are also studied. Our results show, that non-moderated MS burner can be operated as a critical burner reactor without fertile support in an acceptable range of parameters. The temperature feedback coefficients are relatively low, but we show that their further optimization is possible.

KEYWORDS: molten-salt, waste incineration

1 Introduction

The management of radioactive waste is becoming one of the major issues in the field of nuclear energy production. Especially in some European countries it becomes obvious that winning the public support for the construction of final repository will be more difficult than it was expected several years ago. This is one of the main reasons why an increased attention is dedicated to the research of options leading to a significant decrease of the quantity of the high-level radioactive waste (HLW). Provided that the structure of the nuclear energy production will not change in near future, this goal can be achieved only through burning the transuranium isotopes (TRU) extracted from the spent fuel in an advanced type of nuclear reactor.

Fissioning TRU isotopes in an advanced type of light water reactor (LWR) seems to be theoretically possible [1], but at the current state of knowledge repeated reprocessing of the LWR fuel seems to be excluded due to high spontaneous fission neutron rate [2]. The fast reactor technology seems to be more promising [3]. In fast spectra there is a favorable ratio of fission to capture cross section for the minor actinide (MA) isotopes. As a consequence MA do not build in the fuel to the same extent as in thermal spectra systems and therefore the radiation level of the irradiated fuel is much lower. Nevertheless considerable cooling period between the fuel outage and new pin fabrication is still inevitable. This would lead to a substantial stockpile of fissile material waiting for the reprocessing.

In view of above-mentioned problems of well established technologies to deal with the problem of TRU burning, there is a considerable renew of the interest in alternative approaches, among them in molten salt reactors (MSR). In the MSR concept the fuel is liquid, fissile material being dissolved in molten fluoride salt. This fact considerably simplifies the fuel fabrication process. The only parameter which have to be observed is the solubility limit of TRU trifluorides dissolved in the salt. It is generally admitted that the fuel reprocessing and new fuel fabrication could be done on-site, either on-line or in a batch process. It has among others a positive implication for the non-proliferation issue.

The molten salt reactor was developed in 1950's and 1960's in Oak Ridge National Laboratory with the final aim to be operated as graphite moderated breeder reactor in thorium-uranium cycle [4]. Similar reactor concept was later considered for the purpose of TRU burning [5, 6]. It was shown, that moderated MSR can burn TRU isotopes only if fertile support is added [7], which has a negative consequence on the overall efficiency of the process of TRU burning.

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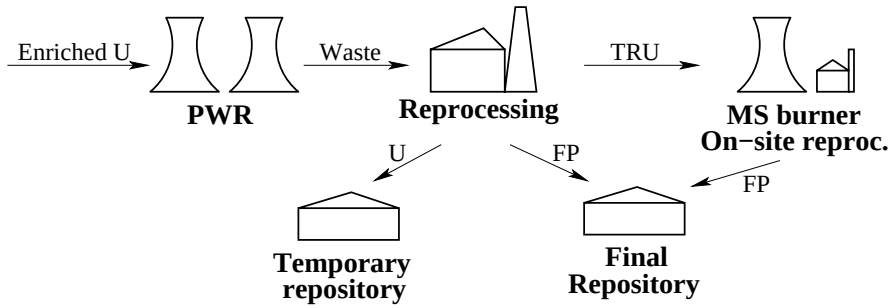


Figure 1: Simplified scheme of PWR - MS incinerator fuel cycle. Abbreviations: FP - fission products, TRU - transuranium isotopes, U - Uranium

The idea to use a non-moderated MSR for the purpose of transmutation comes probably from M. Taube [8], but in the context of TRU burning such reactor was studied only recently. Results of the first studies are encouraging, as they show, that from the point of view of neutron balance a non-moderated MSR can be operated as TRU burner without fertile support [7, 9].

In this article we pursue further this effort. We study in detail the performance of the non-moderated MSR, when operated as a burner reactor closing the nuclear fuel cycle of a pressurized water reactor (PWR). First, we present a study of a conceptual reference design of the MS burner. We present its neutronic characteristics and main safety parameters, both in the first years of operation and in equilibrium fuel cycle. Then we present results of a parametric study, where we monitor influence of the basic parameters (salt composition, neutron flux and feed composition) on the burner performance.

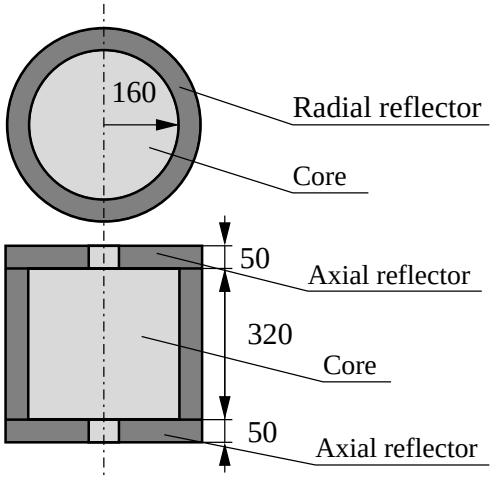
2 System description

The PWR - MS burner fuel cycle is schematically illustrated in Figure 1. PWR is fed with enriched uranium, the waste is reprocessed and split into three streams. Fission products (FP) are send to final repository. Reprocessed Uranium, as a potentially valuable material, is stored in a temporary storage. TRU isotopes are dissolved in molten fluoride salt and they are fed continuously into the MS burner, operated without fertile support. The carrier salt is reprocessed in an on-site facility, either on-line, or in small batches (compared to the core volume). FP extracted from the salt are sent to final repository, while the carrier salt with actinides is returned to the burner.

For the purpose of the simulation of salt reprocessing we divided the fission products into classes according to the method and efficiency of their extraction from the salt. Volatile FP (Xe, Kr, Rn) can be extracted very efficiently by helium bubbling. We suppose that they are extracted from the salt in 50 s. Non soluble fission products (As, Se, Zr, Nb, Mo, Tc, Ru, Rh, Pd, Ag, Sb, Te) are extracted with a characteristic time 1 hour. Soluble fission products are removed by pyrochemical methods during the salt reprocessing. In all the simulations we consider that whole core is reprocessed within 150 days with a 50% efficiency of FP extraction.

As it is impossible to separate completely fission products from TRU isotopes, a minimum quantity of TRU leaves the core with the FP stream. This fact has to be accounted for a correct evaluation of the waste toxicity. Generally it is assumed that 0.1% of actinides from the volume handled during the reprocessing is lost with the FP stream. We use this assumption in our calculations as well.

Conceptual design of reactor core is illustrated in Figure 2. It has a form of a cylinder of equal



Core characteristics	
Volume of the core [m^3]	25.7
Volume of primary circuit [m^3]	39.7
Average salt temperature [^{o}C]	650
Flow speed in core [$m\,s^{-1}$]	0.2
Graphite density [$g\,cm^{-3}$]	1.92
El. power conv. efficiency [%]	40
Annual load [days]	300
HM losses during salt reproc. [%]	0.1

Figure 2: Conceptual design of the core and its main characteristics

height and diameter, surrounded radially and axially by a graphite reflector. The core design is optimized from the point of view of neutron balance. Core is not moderated in order to obtain hard neutron spectrum which favors actinide fission. The reflector material has only a weak influence on the overall characteristics of the burner, including the neutron spectrum. Graphite was chosen in order to protect construction materials from damage caused by high energy neutrons. The 40% electric power conversion efficiency used in the fuel-cycle studies is related to the high core output temperature of approximately 700 °C.

3 Solution method

Solution of burn-up problem in the MSR is simplified if compared to the solid fuel reactor case. Due to the fact that the fuel is liquid and it flows between the core and heat exchangers, its effective mixing occurs in the reactor. We can thus consider that at a given moment the fuel composition is the same throughout the primary circuit, and note $n_i(t)$ the concentration of an isotope i at time t.

The production rate of an isotope j from an isotope i due to the inelastic scattering reaction with a macroscopic cross section Σ_{ij} is equal to $n_i(t)\Sigma_{ij}\phi_{ef}$. In this expression appears an effective neutron flux ϕ_{ef} instead of the average core neutron flux $\overline{\phi}$. This is a result of the fact that the fuel circulates out of the core where the neutron flux is negligible. The two fluxes are related through equation $\phi_{ef} = \overline{\phi}\, V_{core}/V$, where V is the total volume of the primary circuit.

Analogically the production rate of the isotope j from the isotope i due to the decay reaction with decay rate λ_{ij} is equal to $n_i(t)\,\lambda_{ij}$.

Extraction of fission products during the reprocessing is simulated by artificial decay rates ϵ_i/τ, where τ is a reprocessing period and ϵ_i is the effectiveness of extraction for given isotope. Hence isotope extracted within one minute of production "decays" with rate equal to $1/60\ s^{-1}$.

If we put all terms discussed above together and if we note q the source term (feed of TRU isotopes), it can be seen that the evolution of the inventory of MS burner is described by a set of coupled linear differential equations

$$\frac{dn_i(t)}{dt} = \sum_j \Big[\Big(\phi_{ef}\ \Sigma_{ji}(t) + \lambda_{ji}\Big)n_j(t) - \Big(\phi_{ef}\,\Sigma_{ij}(t) + \lambda_{ij}\Big)n_i(t)\Big] - \frac{\epsilon_i}{\tau_i}n_i(t) + q_i(t) \qquad (1)$$

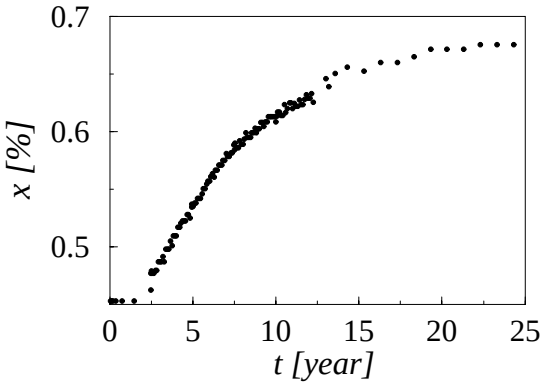


Figure 3: Evolution of the actinide molar concentration in carrier salt.

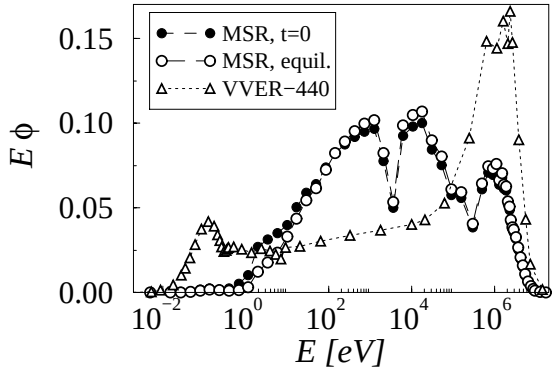


Figure 4: Average spectrum in the MS burner at the beginning of operation and at equilibrium, compared to the spectrum of a VVER-440 assembly.

For the solution of the problem given by equation (1) we have developed specialized tool OGAR (One Group AppRoximation). It incorporates numerical solver of equations (1) and code MCNP-4C [10] for periodic one-group cross sections recalculation. The integration of eq. (1) is done by the Runge-Kuta method of 4th order. The solution of time dependent burn-out problem is rather time consuming. Therefore a possibility of direct evaluation of equilibrium results ($dn_i/dt = 0$ in eq. (1)) was integrated in the code. Equation (1) is then solved iteratively, 5 to 10 recalculations of cross sections are necessary to achieve convergence.

As the libraries distributed with MCNP-4C code do not contain the data for all important TRU isotopes, new libraries were prepared from the evaluated nuclear data libraries using code NJOY-99 [11]. These data were taken from the libraries ENDF/B-VI, JENDL 3.2 and JEF 2.2 in cited order in function of the availability.

Temperature feedback coefficients were evaluated from the series of static MCNP-4C calculations done with one fuel composition for different temperatures.

4 Reference core results

4.1 Fuel inventory

As a reference case we studied MS burner fed with the TRU isotopes extracted from a VVER-440 fuel with a burn-up 40 MWd/kg and 10 years cooling period. These TRU isotopes are dissolved in a molten fluoride salt of composition 35%^{7}LiF/38%BeF_2/27%NaF. This salt composition have been chosen for its low melting point, which is 338^{o}C [12]. Chosen salt composition differs from that used in Molten Salt Reactor Experiment [4] and thermal MS reactor studies [6]. The solubility of TRU trifluorides in formerly used ^{7}LiF/Be_2F salt composition is only about 0.5 mol% at 550 ^{o}C [13], which is not sufficient for the burner. It is increased by the sodium addition. Calculations were done with a constant average neutron flux 1×10^{15} cm^{-2}s^{-1}.

The evolution of actinide molar concentration in the salt can be seen in Figure 3. It is evident that the neutron balance deteriorates due to changes in the salt composition during the beginning of operation, but ultimately an equilibrium composition is reached. The critical molar actinide concentration in salt at the beginning of operation is 0.45%, while the equilibrium critical concentration

Nuclide	MSR			PWR	FBR
	σ_f	σ_c	σ_c/σ_a	σ_c/σ_a	σ_c/σ_a
^{237}Np	0.20	20.17	0.01	0.02	0.21
^{239}Pu	14.44	9.84	0.59	0.64	0.80
^{240}Pu	0.28	14.22	0.02	0.01	0.47
^{241}Am	0.33	22.64	0.01	0.01	0.17
^{243}Am	0.20	27.42	0.01	0.01	0.21
^{242}Cm	0.12	6.63	0.02	0.09	0.39
^{243}Cm	44.62	6.56	0.87	0.90	0.92
^{244}Cm	0.58	14.11	0.04	0.06	0.38

Table 1: Average fission and capture cross sections in MS burner reactor and comparison of capture to absorption ratio for different reactor types. Data for PWR and FBR (fast breeder reactor) were taken from ORIGEN-2.1 [15] library.

Power	1340 MW_{th}
Salt density	2.10 g cm^{-3}
In reactor inventory [kg]	
Fiss. Prod.	133
TRU	3942
Np/Pu/Am/Cm	63/2827/309/710
PWR+MSR cycle waste [kg/TWh_e]	
Np/Pu/Am/Cm	0.02/1.1/0.12/0.26
PWR open cycle waste [kg/TWh_e]	
Np/Pu/Am/Cm	1.9/40.5/1.0/0.2

Table 2: Reference core characteristics in equilibrium and comparison of waste production in open and closed fuel cycles.

attains 0.75%. Due to not very efficient fission of higher actinides we observe their important build-up during the burner operation. The equilibrium burner fuel inventory amounts to about 4000 kg of actinides in primary circuit, with 2827 kg of Pu, 309 kg of Am and 710 kg of Cm. Quality of Pu vector in equilibrium is deteriorated if compared with Pu vector in the PWR burn-up fuel, only about 40% of Pu is formed from fissile isotopes. The TRU inventory is important, but it is still less than half of inventory in fast spectrum burner of equivalent power [14].

The spectrum of the burner is shown in Figure 4. We note that in spite of the fact that the core is not moderated, the proportion of neutrons of energy higher than 0.1 MeV is rather low. The carrier salt is actually composed from light-weight elements, so it acts also as a moderator. It is nevertheless seen, that the spectrum differs strongly from that of PWR reactor, especially it is noteworthy that the thermal neutrons are completely missing. The cross sections are thus lower than in the PWR case which for the same specific power allows to reach higher neutron fluxes. This facilitates the fission of fertile nuclei through their transmutation to fissile ones by neutron capture.

In Table 1 we show the ratio of fission to absorption cross section for the main TRU isotopes, which is a useful indicator of reactor properties. It is seen that this ratio is almost the same in MS burner and PWR reactor. There is not enough of high-energetic neutrons in MS burner to fission directly the fertile isotopes, which explains the important inventory of Cm in equilibrium salt composition.

In Table 2 we show the comparison of the waste produced in open PWR cycle and closed PWR-MS burner cycle for 1 TWh electric. These results were evaluated assuming losses of 0.1% of TRU from the volume handled during the reprocessing, due to imperfect FP/TRU separation. We assumed that entire volume of MS burner primary circuit is reprocessed in 150 days with 50% efficiency of fission products extraction. In this case the production of TRU isotopes from closed fuel cycle is from one to two orders of magnitude lower than in the open cycle case.

4.2 Reactivity coefficients

Values of the reactivity coefficients are summarized in Table 3. The values of the total temperature feedback coefficient α_{tot} are relatively low, especially at the beginning of the operation, where it is close to zero. For a better insight the value of α_{tot} is split into Doppler (α_{dop}) and salt expansion (α_{exp}) components. Results shown in Table 3 indicate that the value of α_{exp} grows with the molar fraction

coef.	$t = 0$	$t = 5$y	$t = 10$y	Equil.	$t = 0, +FP$
x[mol%]	0.45	0.54	0.61	0.75	0.76
α_{dop}	$-0.3(5)$	-1.3(5)	-1.3(5)	$-1.2(5)$	0.0(5)
α_{exp}	$-0.1(5)$	-0.1(5)	-0.6(5)	$-1.0(5)$	-1.8(5)
α_{tot}	$-0.4(5)$	-1.4(5)	-1.9(5)	$-2.2(5)$	-1.8(5)
β_{eff}	0.0029	0.0035	0.0034	0.0034	0.0029

Table 3: Temperature feedback coefficients α and effective delayed neutron fraction β_{eff} for the reference core during the first years of operation and in equilibrium. Last column gives the results for the core poisoned with FP. The values in parenthesis are the uncertainties of the least significant digits, based on the values of standard deviation from Monte-Carlo results. Calculations were done with the density variation of -0.0004 g cm^{-3}K^{-1}.

of TRU isotopes in the salt. This property could be used to increase the temperature feedback at the beginning of operation. To demonstrate this feature, we increased artificially the startup critical concentration of TRU isotopes by poisoning the core by fission products (column marked "$t = 0, +FP$" in the Table 3). We notice that the value α_{dop} stays unchanged, but the value of α_{exp} is increased significantly.

The values of the effective delayed neutron fraction are close to the values observed in fast reactors. The main contribution of delayed neutrons comes from ^{241}Pu, whose proportion among actinides attains 8% at $t = 0$ and then grows to 11% at equilibrium.

4.3 Tritium production

In MSR with ^{7}Li/Be/Na salt, the main part of tritium production comes from following two reaction channels [16]:

$$^9\mathrm{Be} + \mathrm{n} \longrightarrow {}^6\mathrm{Li} + \alpha + \beta^-, \quad \text{followed by} \quad {}^6\mathrm{Li} + \mathrm{n} \longrightarrow \mathrm{t} + \alpha \tag{2}$$

$$^7\mathrm{Li} + \mathrm{n} \longrightarrow \mathrm{n}' + \mathrm{t} + \alpha. \tag{3}$$

The (n, t) cross section for ^6Li in equilibrium fuel composition is 11 barns. That means that in the neutron flux 1×10^{15}cm^{-2}s^{-1} the initial fraction of ^{6}Li decays with a half-life of 2 years. After several years transition period the tritium production is thus determined by the amount of ^{9}Be and ^{7}Li in the salt. In our reference case the total tritium production is 200 g/year, 137 g/year being produced from Be and 63 g/year from ^{7}Li (1 year = 300 effective power days).

It is noteworthy that cross-sections for reaction (n, α) on ^{9}Be and reaction $(n, n't)$ on ^{7}Li have a threshold close to 1 MeV. As the fast neutron fraction is about the same for all MSR reactors, the tritium production will depend only on the salt composition and the neutron flux. Hence in well moderated cores, where the neutron flux is generally lower, the tritium production should be lower as well.

5 Parametric study

5.1 Salt composition

We have evaluated the burner properties for two alternative carrier salt compositions. The first composition is 16%^{7}LiF/28%BeF_2/56%NaF. This salt has a melting point 478 ^{o}C [12], which is higher

	16Li/28Be/56Na		50Na/50Zr		Burn-up 80MWd/kg	
coef.	$t = 0$	Equil.	$t = 0$	Equil.	$t = 0$	Equil.
x [%]	0.4514	0.8832	0.99	5.287	0.75	1.55
α_{dop}	-0.3(4)	-1.1(8)	-1.5(4)	-2.1(7)	-0.2(4)	-2.2(4)
α_{exp}	-0.8(4)	-1.6(8)	-1.1(4)	-1.2(7)	-0.8(4)	-0.7(4)
α_{tot}	-1.1(4)	-2.8(8)	-2.7(4)	-3.3(7)	-1.0(4)	-2.9(4)

Table 4: Temperature feedback coefficients α for 1) reference core with alternative salt compositions (left hand side of the table) and 2) reference core fed with TRU elements extracted from PWR fuel with higher burn-up than in the reference case (left part of the Table). Results are always given at the beginning of operation and for equilibrium composition of salt in the core.

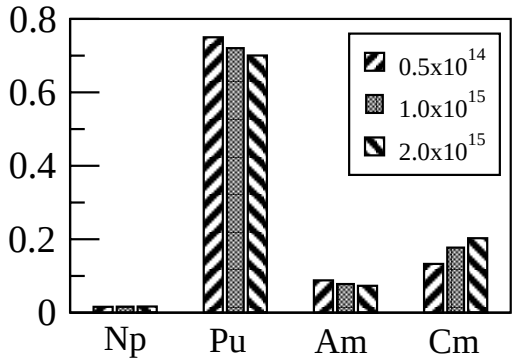


Figure 5: Fractions of the main transuranium elements at equilibrium as a function of the neutron flux

	$\overline{\phi}$ [cm^{-2}s^{-1}]	
coef.	5×10^{14}	2×10^{15}
x[%]	2.15	0.67
α_{dop}	-	-0.6(4)
α_{exp}	-	-1.1(4)
α_{tot}	-	-1.7(4)

Table 5: Molar concentration of TRU isotopes in core as a function of neutron flux

than in the reference salt case. Possible advantages of this salt composition over the reference one are lower viscosity and presumable higher solubility of TRU trifluorides: about 1.4 mol% at 550°C and 2.6 mol% at 640°C [13, 17]. The second considered carrier salt composition is 50%NaF/50%ZrF. This salt does not contain light-weight isotopes and should thus permit to reach harder spectra with good conditions for minor acitnides burning. The melting point of Na/Zr salt is 510 °C [12] and solubility of PuF_3 is about 2 mol% at 550 °C and 3.2 mol% at 750 °C [18].

Results at startup and at equilibrium are given in the Table 4. The results for the alternative ^{7}Li/Be/Na salt composition are almost the same as in the reference case (see Table 3). The critical molar concentration of TRU isotopes in the salt is slightly higher for the alternative ^{7}Li/Be/Na salt composition. It is due to the increased content of sodium, which has a higher neutron capture cross section than the other salt components. The temperature feedback coefficients are again rather low at the startup, at equilibrium their value increases. Due to lower amount of ^{7}Li and Be in the salt the production of tritium decreases of nearly 30% compared to the reference case.

In the case of Na/Zr salt the positive effect of the spectrum hardening does not balance the negative effect of the deteriorated neutron balance due to the presence of Zr, which has a higher capture cross section than ^{7}Li and Be. At equilibrium the TRU molar fraction necessary to reach criticality is above the solubility limit, which rules this salt composition out.

5.2 Flux dependence

We have studied the dependence the burner performance on the average neutron flux in core under the condition that the reprocessing period stays unchanged. The dependence of the equilibrium actinide molar concentration on the neutron flux is given in the Table 5. It appears, that in spite of the fact that the concentration of the fission products in core increases, the critical TRU molar concentration decreases with the neutron flux. The reason for this behavior is that increasing neutron flux promotes the build-up of higher TRU isotopes (see Figure 5). This results in increased number of neutrons per fission, thus better overall neutron balance.

5.3 PWR fuel burn-up

In order to assess the influence of the feed vector degradation on the burner performance, we have increased the burn-up in the PWR reactor (see fig. 1 for the fuel cycle description). The feed vector was composed of TRU isotopes extracted from VVER-1000 fuel, with 6% average enrichment and burn-up 80 MWd/kg. If compared to the 40 MWd/kg burn-up, the Pu vector is significantly degraded. It contains only 60% of fissile isotopes compared to 70% in the reference case.

Results are shown on the right-hand side of the table 4. The critical molar concentration of TRU isotopes has nearly doubled if compared to the reference case. Nevertheless we remark a positive effect on the temperature feedback coefficients at the beginning of operation, because the value of α_{exp} increases with the TRU molar fraction.

6 Conclusion

In this paper we have presented a parametric study of a non-moderated MS burner reactor. The properties of this reactor are studied in the context of closed PWR fuel cycle, where TRU elements extracted from PWR burn-up fuel were fissioned in MS burner reactor.

We have shown that as far as neutron balance is concerned, non moderated MS burner can be run as a critical reactor without fertile support in a reasonable range of parameters. The neutronic properties of ^{7}Li/Be/Na fluoride salts are acceptable and the neutron balance does not depend strongly on the precise carrier salt composition. Further composition optimization in order to decrease tritium production or increase solubility of TRU trifluorides is thus possible.

Neutron flux level has a relatively strong influence on the neutron balance. The optimum seems to be of the order of 1×10^{15} cm^{-2}s^{-1}, which corresponds to the specific power 50 W cm^{-3}. Below this value the neutron balance deteriorates rapidly.

We have also shown that MS burner can be fed with TRU having deteriorated Pu vector compared to standard 40 MWd/kg burn-up. This gives a margin for further optimization of PWR fuel cycle, hopefully leading to the lowering of the proportion of dedicated reactors needed to close the PWR fuel cycle.

The value of temperature feedback coefficient depends on the molar concentration of TRU in the salt. Its value improves with growing concentration. This gives a possibility to optimize the safety characteristics of the burner at the startup period, when the value of the temperature feedback coefficient is rather low.

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