

Safety-Related Neutronics Parameters of a Molten Salt Actinide Recycler and Transmuter

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Abstract

This paper describes studies on reactivity coefficients and kinetics parameters in a fertile-free molten salt reactor. This reactor concept originated at RRC-KI for burning and transmutation of LWR spent fuel. The studies are performed at several institutions in the framework of a Coordinated Research Project initiated by the IAEA.

The participants investigated the influence of different codes, computation techniques and nuclear data libraries on the neutronics parameters. It is shown that the reactivity coefficients are favorable for reactor safety.

KEYWORDS: *Molten salt reactors, Reactivity coefficients, Transmutation*

1. Introduction

The IAEA has initiated a Coordinated Research Project (CRP) on "Studies of Advanced Reactor Technology Options for Effective Incineration of Radioactive Waste"[1]. About 20 institutions from 14 member states and 3 international organizations take part in the CRP. The final goal of the CRP is to deepen the understanding of the dynamics of transmutation systems, to qualify and assess the available methods, specify the range of validity of these methods, and formulate requirements for future theoretical developments.

Several systems are studied in the framework of the CRP, in particular, critical reactors with solid fuel, subcritical systems with solid fuel, critical systems with liquid (molten salt) fuel. For the abovementioned reactor types, fertile-containing and fertile-free fuels are considered. In general, the solid fuel systems show a strong deterioration of safety parameters with a significant increase of the minor actinide content that may make necessary reactor operation at subcritical conditions, i.e. in accelerator driven systems (ADS).

One of the systems studied in the CRP framework is MOSART, a molten salt "critical"

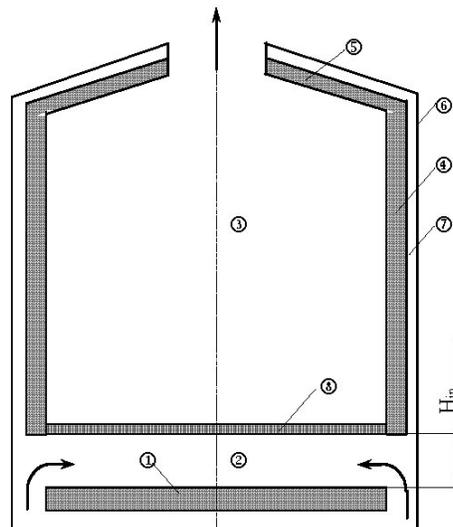
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2400MWth (1100 MWe) reactor concept with fertile-free fuel developed at RRC-KI [2]. The studies include neutronics analyses, in particular, benchmarks on computing safety parameters (reactivity coefficients, effective delayed neutron fraction, etc.) and (2) transient analyses, which are supported by neutronics studies, for simulating relevant hypothetical accidents. In the paper, some results of the MOSART neutronics analyses are presented.

2. MOSART reactor concept

MOSART is a reactor developed for burning LWR spent fuel. The fuel salt is a molten $58\text{NaF}-15\text{LiF}-27\text{BeF}_2$ (mole%) mixture with 479°C melting temperature and addition of about 1.05 mole% of $(\text{TRUF3} + \text{LnF}_3)$. MOSART contains no solid moderator in the core, thus improving its incineration and transmutation potential, that means only the molten salt, a (few) control rod(s), and a (few) distribution plate(s) for arranging the salt flow are present in the core (in the neutronics model, considered in the benchmark, the rods and plates are not taken into account). The core is cylindrical (3.4 m in diameter, 3.6 to 4.0 m high, the upper core boundary being of conical shape), it is surrounded by a reflector made of graphite or a nickel-based alloy (for neutronics studies presented in the following, only a 20 cm graphite reflector is considered). The core and loop volumes are 32.67 and 18.4 cubic meters respectively, the salt flow rate being 10000 kg/s at nominal conditions, the salt density at the core inlet being ca. 2140 kg per cubic meter.

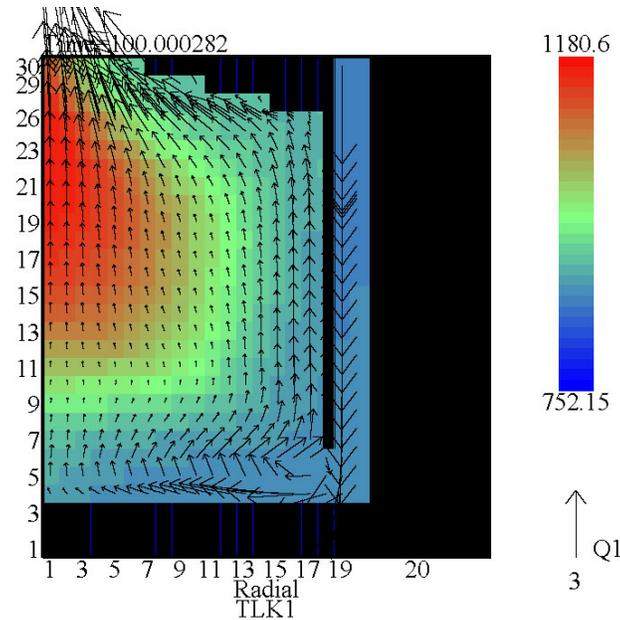
Figure 1: Sectional view of 2400MWt MOSART core (option1: the salt enters from a radial window): 1-bottom reflector, 2- radial core inlet window, 3- core, 4-radial reflector, 5- top reflector, 6-reactor vessel, 7 - radial cooling annulus, 8- distribution plate



Two options were considered in the CRP studies by now: (1) the fuel salt enters into the core through a radial window of 0.5 meter height (as shown in Figure 1, the reactor constructions outside the reflector are not considered in the following) or (2) enters from the top into the peripheral salt annulus or 20 cm thick. In both cases, the salt leaves the core through a pipe (of 1 m diameter) of the top conical reflector.

The temperature and velocity distributions for the second option (computed at steady-state by employing the SIMMER [3] code) are shown in Figure 2, the temperature (from 752.15 to 1180.6 K) and velocity (3 m/s) scales being given. This flow profile was obtained after trying several distribution plate arrangements: to avoid stagnant regions and reverse flow; otherwise the maximum salt temperature could be appreciably higher than shown in Figure 2.

Figure 2: Temperature and velocity distribution in the MOSART core (option 2: the salt enters from the top into the peripheral salt annulus)



The start-up and feed fuel material for the MOSART core, which is considered in the CRP studies, is a typical mixture of transuranics (TRUs) from the uranium oxide (UOX) spent fuel of a commercial PWR (60 GWd/t; after 1 year cooling). The benchmark results are provided in the following for the equilibrium state. At this state, a large number of TRUs contribute significantly to the neutron balance in the system, nuclides up to Cf251 being considered in the benchmark).

3. Data and codes

Several nuclear data libraries (in particular ENDF/B-VI, JEF 2.2, JEFF 3.0, JEFF 3.1, JENDL 3.3) were used for performing the analyses. Both multigroup deterministic and Monte-Carlo models (with “point-wise” nuclear data libraries) were employed. Participants from BME, NRG, RRC-KI, SCK-CEN made computations with different versions of the MCNP [4], or MCNPX [5], codes. The NRG version of MCNP includes an extension [6] for computing β -eff, the effective delayed neutron fraction.

In addition to MCNP, BME employed a 1D Sn code, XSDRNPM [7] and a corresponding 172-group library based on JEFF 3.1. This code relies on a buckling (that is computed on the basis of the core height) correction technique in 2D that may bring a significant uncertainty into computed k -eff values in the 2D case. However, the computed 172-group spectra are assumed to be accurate for computing few-group macroscopic group cross-sections.

The computed by XSDRNPM 4-group macroscopic cross-sections (for different reactor

sub-regions, at different temperatures) were employed at Polito with a 2D diffusion code [8]. This code computes k-eff & neutron flux and takes into account the effect of fuel movement on the delayed neutron precursor concentrations (if the fuel velocity distribution is known).

At FZK, a 560-group cross-section library [9] (several version of the library are available: for JEFF 3.0, JENDL 3.3, etc) was employed for (1) computing composition-dependent cross-sections and (2) producing a smaller cross-section libraries (which include, in particular, f-factors) with 172 and 9 energy groups. 2D 560-group neutron transport calculations were performed with the DANTSYS code [10]. Coupled neutronics (9-group) and thermal-hydraulics calculations were performed with the 2D SIMMER-III code in order to obtain the velocity distribution and evaluate the effect of the delayed neutron precursor movement at steady-state conditions.

In addition to MCNP, RRC-KI employed another Monte-Carlo code, MCU, which takes nuclear data from a related code library, MCUDAT [11].

4. Static neutronic analyses

The principal benchmark results are given in Table 1. The k-eff calculations were performed with the core/reflector temperature values of 900/950K.

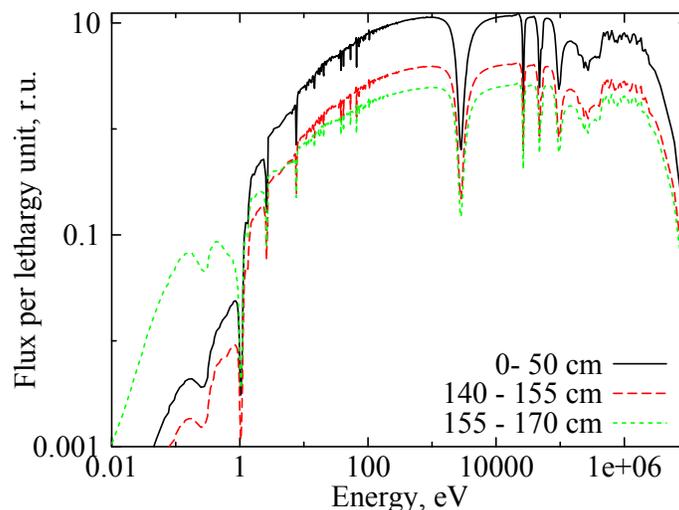
Table 1: MOSART main reactivity and kinetics parameters

	BME MCNP4C +JEFF 3.1 /1D 172 gr. +JEFF 3.1 /MCNP4C +JEF2.2	FZK 2D 560 gr. JEFF 3.0 /JENDL 3.3 / ENDF 6.8 /JEF2.2	NRG MCNP4C JEFF 3.1 /JEFF 3.0	Polito 2D 4 gr. JEFF 3.1	RRC-KI MCNP4B +ENDF 5,6 /MCU +MCUDAT	SCK-CEN MCNPX JEFF 3.1
k-eff	1.00905 /1.01984 /0.96462	0.99285 /1.01023 /0.98474 /0.96498	1.00887 /0.99335	0.99595	0.99791 /0.98930	1.00904
α -total, pcm/K	/-3.86	-3.86 /-3.82 /-3.86	-3.75	-3.78	-3.71 /-3.41	-3.66
α -Doppler, pcm/K	/-1.67	-1.52 /-1.53 /-1.46	-1.42	-1.73	-1.62 /-1.09	-1.69
α -reflector, pcm/K	/-0.05	-0.05		-0.04		
Generation time, μ s		8.3 /8.2		8.8		
β -eff, pcm (static)		/340	326			

All figures of Table 1 are obtained by assuming no inlet in the radial and top reflectors (thus bringing together the models shown in Figures 1 and 2). A simplified 2D model (a right cylinder without axial reflectors) was used at Polito. Thus, the BME (1D) and Polito k-eff results include an uncertainty that is related to the approximate geometry treatment. Assuming that the error related to the 172-group approximation applied by XSDRNPM (for discretization in energy) is minor (that aspect is discussed later), one may conclude (by comparing the k-eff results of BME and Polito with those of other participants) that the corresponding geometry approximations may have a minor influence on the computed reactivity effects and kinetics parameters.

The Monte-Carlo and 560-group based k-eff values, obtained using the JEF2.2 nuclear library, are in excellent agreement (the deviations are about 50 pcm or smaller, that is similar to the statistical uncertainty of the MCNP k-eff results) provided that the same nuclear data are used. That is in line with previously published results [8] on using the 560-group data for k-inf studies of graphite-free molten salt systems with MSRE-type [11] salts (with major fertile nuclide being either Th232 or U238). On the other hand, these studies have shown a very large sensitivity of the computed k-inf values to the group structure (e.g. the 172 group results overestimated k-inf values at high temperatures by about 3000/1000 pcm in the U238/Th232 cases compared to the 560-group and MCNP results) and to the energy threshold (the error was about 600/200 pcm if this value was lower than 30 eV), above which the upscattering effects (in particular for neutron scattering on F19) are ignored. By generating a 172-group library from the 560-group one and comparing the 560-group and 172 group k-eff results, only a minor deviation (of about 200 pcm) was observed at FZK; similarly the mentioned “upscattering” effects were much smaller. The reason for better performance of the simplified energy discretization model (with 172 groups) in the MOSART case is that there is no single fertile nuclide (like U238 and Th232) in the system. Since the neutron spectrum in Molten Salt reactors (without graphite in the core) includes a significant fraction of neutrons with energies above a few eV (see Figure 3), a quite accurate modeling of neutron interaction with nuclei (in particular heavy nuclides and F19) at energies near the major resonance peaks of the fertile nuclides is required if neutron absorption by these fertile elements contributes significantly to the overall neutron balance.

Figure 3: Axially averaged flux spectra in MOSART at different radial locations



Though the spectra shown in Figure 3 indicate a higher fraction of lower energy neutrons near the core and reflector boundary (R=170 cm) due to moderation in the graphite reflector, the neutron spectrum can be considered to be essentially a fast one, a very important feature for a transmutation reactor.

Criticality values computed with different nuclear data may differ significantly. In particular a strong difference can be seen between the JEFF 3.1 and JEF 2.2 cases. The reasons for this difference were investigated at BME by considering originally all data from JEFF 3.1 and then replacing data for particular nuclides by those from JEF 2.2. The results of this study are presented in Table 2.

Table 2: Effect of successive data replacement (JEFF-3.1 data replaced by JEF 2.2 ones)

Isotopes from JEF 2.2	k-eff	Standard deviation
None	1.00905	0.0003
Pu238	1.00576	0.00029
+ Pu239	1.00172	0.0003
+ Pu240	0.99699	0.00029
+ Pu241	1.00517	0.0003
+ Pu242	1.00438	0.0003
+ Cm245	1.01095	0.00031
+ Cm247	0.99613	0.0003
+ Be9	0.97842	0.00029
+ Li7	0.9773	0.00028
+ F19	0.96595	0.00028
+ Na23	0.96635	0.00028
+ B10	0.96661	0.00028

One may see in Table 2 that the major contributions (to the difference between the JEFF 3.1 and JEF 2.2 results) come from Cm isotopes, Be9 and F19. This result underlines importance of using new evaluated data (which are assumed here to be more accurate) for the mentioned non-heavy nuclides in the molten salt case.

All α -coefficients (total, Doppler, reflector) shown in Table 1 are related to heating up the core and reflector by 600K (from the state, at which the k-eff values were computed). The α -total coefficient is the total reactivity effect divided by 600. The α -Doppler one is determined similarly, except that the salt density is assumed to be temperature-independent. The α -reflector coefficient takes into account the temperature variation in the reflector only.

The temperature coefficients are favorable for the reactor safety (unlike the case with solid fuel transmutation reactors, which may require a subcriticality and an external neutron source for coping with safety problems), in particular due to strong density and Doppler effects. The reflector coefficient plays a minor role.

The reactivity coefficients obtained by different participants are in reasonable (for the purpose of safety analyses) agreement, except that the underestimation of the MCU results (in particular due to the fuel Doppler effect) compared to other ones.

Results of all participants show a similar trend in temperature dependence of the coefficients. Though the Doppler coefficient varies appreciably (up to 20% if a smaller temperature shift, e.g. 100K or 300K, instead of 600K, is considered), a relatively weak variation (less than 5%) of the total α -coefficient is observed. The reflector coefficient shows stronger variation (it can be lower or higher by ca. 50% at lower or higher salt and reflector

temperatures) but the absolute value remains well below 0.1 pcm/K in all considered cases.

Generation time values computed at Polito are higher than those computed at FZK (see Table 1), the first values increasing with temperature (by ca. 20% after heating up the core by 600K) while the latter ones remaining almost unchanged. This can most probably be attributed to different cross-section generation options employed in each case.

The β -eff values (shown in Table 1 and computed by assuming no delayed neutron precursor movement) were calculated at FZK and NRG. They agree reasonably well taking into account use of different nuclear data and Monte-Carlo statistical uncertainties (ca. 5 pcm). According FZK results, major nuclide contributions come from Pu241 (ca. 60%), Pu239 (ca. 17%), Cm245 (ca. 9%) and Cm247 (ca. 4%).

5. Effect of delayed neutron precursor movement at steady-state

The effect of the delayed precursor movement at steady-state was evaluated at FZK and Polito. Different salt velocity profiles and different geometry models were employed. The velocity profile (together with the precursor distributions) was computed at FZK by SIMMER (see Figure 2) for the “top inlet” option as a result of a 100s “transient” simulation that brings an initially “non-equilibrium” (determined by input code parameters for a “coarse” geometry mesh) core reasonably close to steady-state conditions (the velocity profile as well as the reactor power varied significantly during this simulation). The velocity profile employed at Polito was computed at RRC-KI for the “radial inlet” option. Both profiles show similar features: a higher salt velocity radially closer to the center. An additional difference in modeling comes due to the geometry approximation applied at Polito: simulating the flow in the right cylinder leads to higher salt residence time in the core (ca. 8 s) compared to the FZK case (ca. 7 s). Since in both cases, the “loop” time is similar (ca. 4 s), the relative fraction of the “loop” time (during which the precursors decay being outside the core) compared to the total salt circulation time differs by ca. 9% in these two cases.

We assume that the effect of the precursor movement (relative variation of the effective fraction of delayed neutrons due to salt flow, β -eff-lost/ β -eff-static) differs mainly due to the mentioned deviation in modeling. This effect is ca. 50% in the FZK case and 42% in the Polito case. These results are currently considered as preliminary and should be confirmed by future studies.

The computed “movable precursor” effects in MOSART seem to be relatively strong compared to MSRE (ca. 33% according to the experiment). Note, that the residence times in the (1) MSRE core, (2) plena below/above the core and (3) in the loop are ca. (1) 8.2, (2) 4.3, and (3) 12.4 s. Thus the MSRE “loop” time fraction is larger compared to the MOSART, but the effect in the MSRE is smaller. To get a deeper understanding of the situation, calculations with the flat velocity profile (similar to that one assumed in the MSRE case) were performed at Polito. Using the flat profile (instead of the “real” one) reduced the “movable precursor” effect to 33% (from 42%). This happened due to higher salt velocities (in the “real” flow profile) in the center where the fission source is at maximum. Thus the “effective” salt residence time in the core decreases when the “real” non-flat MOSART velocity profile is employed. One should also consider effects of the axial shape of fission source in the MOSART and MSRE cores. Preliminary evaluations show that the MSRE one is more flat in average, thus increasing the “effective” salt residence time and decreasing the “movable precursor” effect. These evaluations should be confirmed in the future.

6. Conclusions

Several nuclear data libraries, codes, and computation models were employed to compute safety-related neutronics parameters for a molten salt actinides recycler and transmuter. The results show that the parameters are favorable for reactor safety, mainly due to the strong density and fuel Doppler effect. The results are in principal agreement with respect to the major reactivity effects.

A simplified procedure - based on using few-group cross-sections obtained from the 172-group library by employing 1D spectra - was shown to be appropriate for reactivity effect and transient analyses in the considered reactor model.

560-group deterministic and Monte-Carlo k-eff results are in excellent agreement (provided that the same nuclear data are employed) giving a higher confidence to the results. The influence of different nuclear data options on the k-eff values is quite strong. Comparison of different data sets revealed a strong contribution from data differences for Cm isotopes and light (Be9, F19) elements, the latter being present in large quantities in the carrier salt.

Major kinetics parameters computed by different participants agree reasonably well taking into account data and modeling differences. Major contributions to β -eff come from Pu241 (ca. 60%), Pu239 (ca. 17%), Cm245 (ca. 9%) and Cm247 (ca. 4%).

Preliminary evaluations of the effect of delayed precursor movement at steady-state show a relatively high reduction of the effective delayed neutron fraction (by ca. 40 to 50%). This effect (as well as the temperature distribution in the core) strongly depends upon the velocity profile that in its turn depends upon the distribution plate design and may vary strongly during the transient. Additional effort should be paid to confirm the computed effect.

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