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Integral Experiments with Teflon and ^{7}LiF-BeF$_2$ Salt at LR-0 Reactor

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Abstract - *Experiments introduced in this paper are part of the research on Molten Salt Reactor (MSR) technology in Research Centre Rez. This research is aimed at validation of neutronic parameters of materials used in MSR design by integral experiments. Previous work included study on graphite and, for methodological purposes, also fluoride LiF-NaF salt. Results obtained by repeated experiments with graphite and LiF-NaF show only minor discrepancies in C/E-1 in terms of criticality. However, neutron spectrum measurement in LiF-NaF discovered quite large variations in the fast part of the spectrum. It was concluded that the variations could go on the account of the fluorine data. Therefore, other fluorine rich substances, Teflon and ^{7}LiF-BeF$_2$ salt were used in further work. ^{7}LiF-BeF$_2$ salt is the real compound used in MSR experiment in Oak Ridge National Laboratory in 60's. Presented set of integral experiments with highly fluorinated compounds show satisfactory agreement in criticality, especially in case of Teflon.*

I. INTRODUCTION

Current work follows the research of GEN IV reactors containing coolant and/or fuel in form of fluoride salts (Fluoride High Temperature Reactor, Molten Salt Reactor, FHR and MSR respectively) performed at LR-0 reactor in past years. Discrepancies in criticality between measurement and experiment were discovered during reevaluations of work done during EROS experiments [1] with core configurations containing LiF-NaF salt (60-40 mol%, natural Li, FLINA hereinafter) and combination of FLINA and graphite. It was decided to repeat some of these experiments to confirm the obtained results. Single components of graphite and FLINA were investigated during integral experiments through 2014 – 2015 with conclusions that the discrepancies between calculation and experiments with graphite are within 1 σ uncertainty interval in terms of criticality for all possible graphite insertion configurations [2]. Criticality of the core with FLINA insertion was also found within 1 σ uncertainty interval and the measurement of neutron spectra in inserted cores was performed with conclusion that the neutron spectrum in FLINA salt may not be correctly described by used nuclear data [3].

Measurements of fast neutron spectrum in case of graphite showed excellent rate of agreement with calculation in different sets of nuclear data libraries [3]. Discrepancies in C/E in case of graphite were found to be not exceeding 7 % which is better result than in case of measurement in the void, where the discrepancies can be as high as 13 %. In case of FLINA, the measurements differ by 40 % in some energy groups. It is therefore still believed that the components of FLINA can cause these deviations in neutron spectrum. The sensitivity study in [1] shows that in case of FLINA insertion, the greatest influence to criticality is introduced by ^{19}F(n,elastic) and ^{6}Li(n,t) reactions. Possible influence of fluorine has been discussed in the same work, and therefore it has been decided to continue research in this field by verification of fluorine nuclear cross section description through integral experiment with Teflon. This material has been chosen thanks to its perfect chemical stability and durability in accidental scenarios of LR-0 altogether with substantial content of bound fluorine in CF$_2$ macro-molecules. The fact that the results of experiments with graphite are correct is leading us to the conclusion that if carbon-rich insertions in the LR-0 core give correct results, possible discrepancies will be caused by other elements. Described experiments are thus designated to investigate the impact of fluorine at criticality, reactivity and spectrum.

Teflon experiment, as well as new experiments with ^{7}LiF-BeF$_2$ (66-34 mol%, referred to as FLIBE) salt obtained from past MSRE reactor operated in Oak Ridge National Laboratory, give valuable information needed for analysis of results obtained from previous measurements carried out with FLINA salt.

II. METHODS

Presented experiments, aimed at validation of core and insertions description, were carried out in the benchmark core of the LR-0 reactor and modelled in MCNP6.1 [4] code with different nuclear data libraries. Assessment of influence on reactivity is done by relative way, by comparing the experimentally determined critical states and corresponding calculations. Current results are then compared with values from benchmark calculation.

1. LR-0 Reactor

Experiments have been carried out in the zero-power light water research reactor located in Řež. The core is loaded with six fuel assemblies with nominal enrichment of 3.3 %. Fuel assemblies have hexagonal cross section with 23.6 cm lattice step, typical for VVER-1000 reactors. Design of the fuel assemblies in radial direction is equal to that used in power reactors.

The reactor configuration contains one dry experimental assembly in the center of the core. This assembly with hexagonal section (in fact it forms void channel), made of aluminum, fits directly into the lattice position. By this way, the space for different material insertions is introduced, where the parts of different structural materials for reactors can be investigated from neutronic point of view (see Fig. 1).

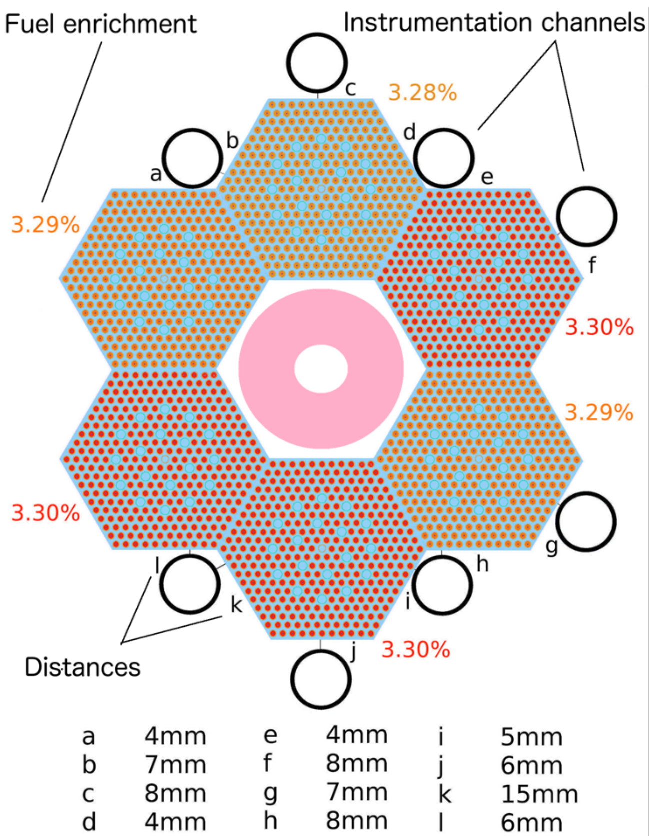

Fig. 1. Schematic view on the benchmark core in LR-0 reactor in configuration with Teflon insertion (in core center).

Described measurements are aimed at integral characteristics measurements, which are combined in critical parameter. In case of LR-0, this critical parameter is introduced by the level of moderator contained in the reactor vessel. In the reactor shut down state, the moderator is released from the reactor vessel and the absorbing clusters are inserted in bottom positions. During the start-up sequence, the clusters are withdrawn out of the core and the moderator is pumped into the reactor vessel. The core in critical state for material insertion investigation doesn't contain any boron absorber and the neutron balance is driven only by the moderator level. Thanks to the precise moderator level measurement, the criticality can be determined with uncertainty in order of few tens of pcm. Three independent critical experiments have been performed to obtain the critical parameter, H_{cr} (critical moderator level), for each studied inserted material.

Teflon insertion has simple cylindrical shape with central cavity for detectors, which are part of neutron spectrometric system. The cylinder is 64 cm high with outer diameter of 20.5 cm. The inner diameter of central cavity is 7.4 cm. Density of the material is 2.19 g/cm^3. Whole cylinder is positioned at the aluminum base, to set the beginning of the insertion into the same level as the beginning of the fuel fission column.

Fig. 2. Process of filling the FLIBE from transport cask into the canister used in irradiation channel of LR-0 reactor.

Canister containing solidified FLIBE salt (Fig. 2 and Fig. 3, right) has also cylindrical shape with central cavity for detectors and is made of nickel alloy (X5CrNi18-10). The total amount of salt, depleted in ^{6}Li, in the canister is 27.54 kg.

2. Calculations

The LR-0 model has been composed in MCNP6.1 code using data from various nuclear libraries. Description of the core components (including fuel) was fixed in ENDF/B-VII.0, only the description of the insertion material was varied. Neutron scattering in the insertion was treated by free gas treatment for both cases, Teflon and FLIBE. Previous works [5] were aimed at benchmarking of used LR-0 model which is now fully standardized. Criticality calculations with 10,000 neutrons per generation and 20,050 (50 of them inactive) generations yielded in results with standard statistical deviation of 0.00005.

Fig. 3. Photographs of reactor insertions; Teflon cylinder (left) and canister with FLIBE salt (right).

Uncertainty analysis was performed by the standard ICSBEP methodology [6]. Identified uncertainty contributors are mentioned in Tab. 1. The largest factors contributing to the total uncertainty are directly connected with fuel definition: lattice pitch and fuel cladding thickness (see Tab. 2). In case of FLIBE salt, the residual content of ^{6}Li was neglected. It is assumed that the ^{6}Li contamination is not higher than 100 ppm and therefore the uncertainty interval in case of FLIBE salt is broadened based on this assumption. Results of sensitivity study on the residual ^{6}Li content in the salt are presented in Tab 3. It is assumed that all uncertainties are uncorrelated and have rectangular distribution. Thus, the total uncertainty is calculated as a square root of sum of squares of single uncertainties.

Table I. Identified sources of uncertainties (1σ)

Type	Value
Fuel cladding thickness	0.0016 cm
Fuel density	0.0093 g/cm^3
Fuel enrichment	0.01 % variation
Fuel lattice pitch	0.15 cm
Moderator level	0.05 cm
Moderator contamination	2 ppm (BE)
Insertion density	1 %
Insertion outer diameter (OD)	0.05
Insertion inner diameter (ID)	0.05
^{6}Li contamination	100 ppm

Table II. Estimated uncertainties in terms of reactivity (pcm)

Parameter	Insertion type	
	Teflon	FLIBE
Fuel cladding	99	32
Fuel density	10	25
Fuel enrichment	44	57
Lattice pitch	102	68
Moderator level	12	24
Moderator contamination	37	13
Insertion density	12	22
Insertion OD	16	-
Insertion ID	17	-
^{6}Li contamination	-	38
Total	**156**	**110**

Table III. Sensitivity study on ^{6}Li content (ENDF/B-VII.0)

Content [ppm]	k_{ef}
0	1.00224
5	1.00202
10	1.00210
15	1.00201
30	1.00200
50	1.00184
100	1.00158

III. RESULTS

1. Criticality Calculations

Critical moderator level (k_{ef}=1) for the case with Teflon cylinder in the channel was measured to be 46.42 cm. For FLIBE salt, the critical moderator level was 54.40 cm. It can be seen that moderating properties of Teflon insertion are comparable with those of graphite insertions. Separate calculation of hypothetical case with graphite material, including S(a,b) matrix, in the body of Teflon insertion showed that the difference between graphite and Teflon in case of criticality is approximately 150 pcm. In case of FLIBE salt, the critical level is strongly influenced by canister made of nickel alloy, which acts as a neutron absorber.

Table IV. Critical levels

Case	Critical level [cm]
Case 1	55.60
Case 7	44.38
Case 8	43.22
Case 15	80.26
EROS3	49.36
Teflon	46.42
FLIBE	54.40

As to compare current experiments with previous relevant experiments, benchmark Cases 1, 7, 8, 15 and new repetition of EROS3 experiment is included in this section. Comparison of critical moderator levels shows Tab. 4 and the insertion core schemes are provided by Fig. 4.

Using benchmark MCNP model and measured critical water levels, results of calculations were obtained for different libraries. These are shown in Tab. 5 and graphically interpreted in Fig. 5 together with results of benchmark values of empty channel (Case 1), graphite insertion without central part (Case 7), graphite insertion including central graphite part (Case 8), FLINA insertion (Case 15), and graphite insertion combined with FLINA center (EROS3).

It can be seen that the C/E-1 results of experiment with Teflon are very close to the reference calculation with empty assembly (without insertion). Modelling of FLIBE experiment shows systematic overestimation, portion of which is given by the uncertainty in ^{6}Li content in depleted FLIBE salt. The sensitivity study in Table 3 shows that the variation of ^{6}Li isotope content from 0 to 100 ppm has rather small impact on reactivity in studied configuration.

Presented results show that the benchmark core in LR-0 is properly described as well as inserted materials in this core. The C/E-1 of empty channel (Case 1) and channel containing graphite insertion only (Case 7, Case 8) is very close to 0. If the Teflon insertion is used, the critical calculation provides result not much worse than the reference Case 1.

In general, the simulation of repeated experiments with FLINA salt underestimate experiments by more than 160 pcm in both cases (Case 15 and EROS3). The collective characteristic is that both configurations contain natural Li. New simulations employing Teflon and FLIBE salt tend to overestimate the experiments.

It can be stated that by introduction of pure material, containing the mixture of carbon and fluorine (CF_2) only, the C/E-1 agreement largely improves and related uncertainty decreases as well. When focusing on the nuclear data library comparison, the most variances among different experiments can be found in case of CENDL-3.1.

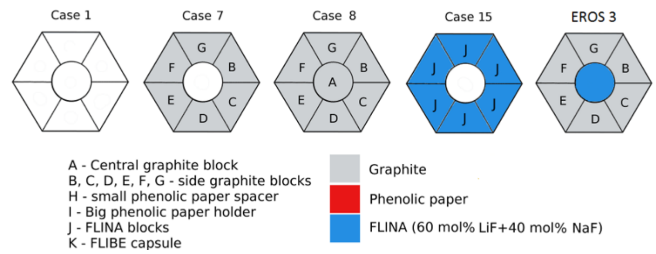

Fig. 4. Schematics of benchmark insertions. Cases 1, 7, 8, 15 and EROS3 are compared with recent results of Teflon and FLIBE.

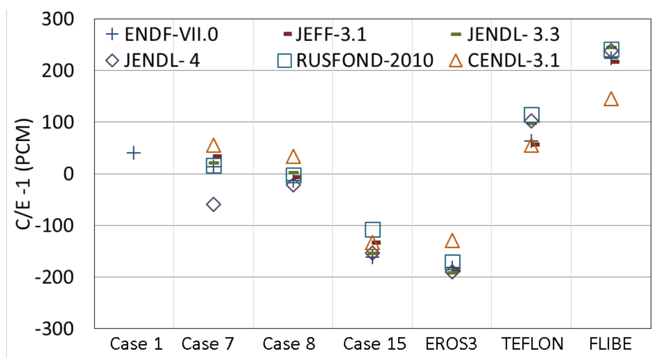

Fig. 5. Graphical comparison of C/E-1 results in different nuclear data libraries.

Table V. Results of C/E-1 in different nuclear data libraries in pcm

	Tot. uncertainty	ENDF-VII.0	JEFF-3.1	JENDL-3.3	JENDL-4	RUSFOND-2010	CENDL-3.1
Case 1	192	41					
Case 7	180	13	34	22	-59	17	56
Case 8	178	-13	-6	3	-22	-2	34
Case 15	153	-161	-133	-153	-153	-107	-133
EROS3	118	-181	-187	-191	-190	-171	-129
Teflon	156	64	57	98	103	114	55
FLIBE	108	224	217	245	239	241	146

2. Neutron Spectrum Calculations

Neutron spectrum, as well as further data for other analyses in following sub-section, were calculated by MCNP6 code. For this purpose, only ENDF/B-VII.0 library was used. Three basic cases, where also measurement by detectors can be done, were calculated and the resulting spectra are plotted in Fig. 6. Graph shows remarkable flux increase in intermediate energies in cases with fluorine-rich insertions, caused by scattering on fluorine nuclei. Characteristic dips at energies of 27, 49, 98 keV and larger perturbation at 271 keV are observable in both types of insertions, because Teflon contains very similar amount of fluorine as FLIBE (30.6 kg in Teflon, 26.2 kg in FLIBE). Larger number of lighter elements in FLIBE salt, as well as the material of the container causes bigger decrease in high energy fluxes, which results in increased intermediate flux, as compared with Teflon insertion. But still the neutron spectrum shape is subjected to fluorine cross section definition in both insertions.

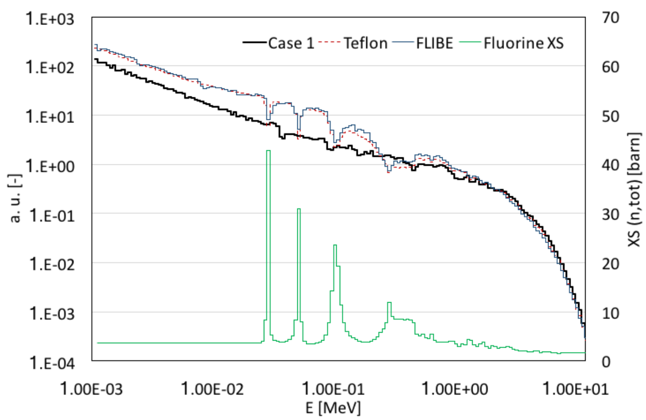

Fig. 6. Fast part of neutron spectrum calculated in Case 1 and experiments with Teflon and FLIBE.

3. Neutron Balance and Sensitivity Analysis

Calculated neutron balances and sensitivities show how important, in terms of criticality, are the inserted materials from the point of view of the whole reactor core. Balances are mentioned in Tab. 6 for Teflon insertion and Tab. 7 for FLIBE insertion.

Table VI. Shares of neutron interactions in case of experiment with Teflon

Nuclide	Collisions	Captures
H	81.6%	41.3%
O	8.2%	0.5%
^{238}U	3.5%	30.6%
F	1.5%	0.2%
^{90}Zr	0.9%	0.3%
^{235}U	0.9%	15.0%
^{56}Fe	0.8%	5.0%

It can be seen that Teflon experiment is better for representation of neutronic characteristics in fluorine, whereas the sensitivity of this nuclide is attenuated in case of FLIBE salt. This is given mainly by the fact that the salt is placed in nickel alloy canister, material which takes non-negligible part on interactions in case of FLIBE experiment. As iron appears as a structural material in fuel and reactor components, ^{56}Fe is fourth most absorbing isotope in the core. The neutronic balances show that the presence of Fe as canister alloy component increases the collision as well as absorption on this isotope and thus the importance of other isotopes contained in the salt is suppressed (See Fig 8 compared with Fig. 7 and Fig. 9) and rather larger salt samples would be needed for deeper investigation.

Sensitivity profiles in Fig. 7 and Fig. 8 show that the scattering reaction on fluorine nuclei in higher energy range (1 keV – 10 MeV) have impact on criticality, which would be visible especially in case of fast neutron systems, whereas the capture reactions on other components (Be, ^{7}Li) play only a minor role in these energies (see Fig 9).

Table VII. Shares of neutron interactions in case of experiment with FLIBE

Nuclide	Collisions	Captures
H	82.4%	41.4%
O	8.1%	0.5%
^{238}U	3.4%	30.0%
^{56}Fe	1.1%	6.3%
^{235}U	0.9%	14.7%
^{90}Zr	0.9%	0.3%
F	0.9%	0.1%
^{94}Zr	0.4%	0.1%
^{92}Zr	0.4%	0.3%
Al	0.3%	0.8%
Be	0.3%	0.0%
^{91}Zr	0.3%	0.7%
^{58}Ni	0.2%	0.9%
^{7}Li	0.1%	0.1%

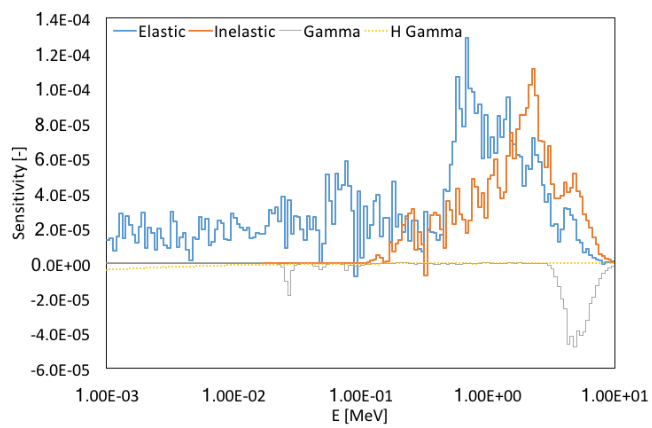

Fig. 7. Sensitivity profiles of scattering reactions on fluorine bound in Teflon in fast energy region, compared with capture reactions on fluorine and hydrogen.

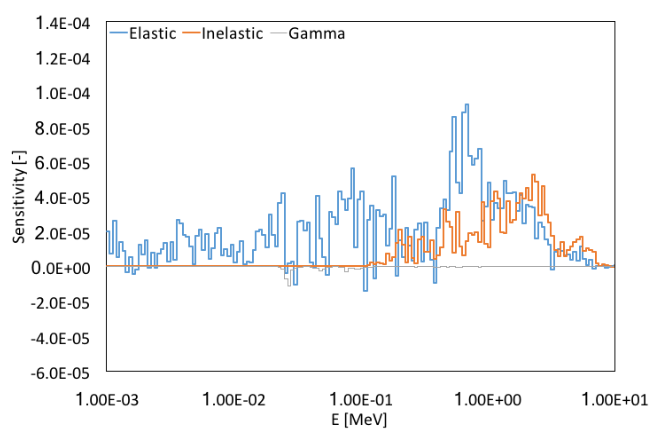

Fig. 8. Sensitivity profiles of scattering reactions on fluorine in FLIBE salt in fast energy region, compared with capture reaction.

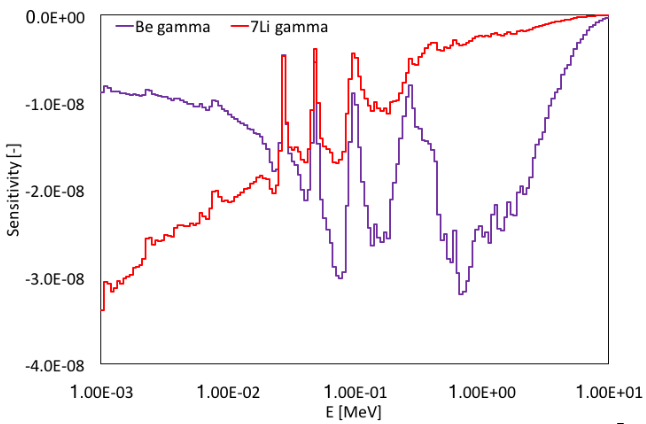

Fig. 9. Sensitivity profiles of radiative capture on Be and ^{7}Li nuclei in FLIBE salt in fast energy region.

IV. CONCLUSIONS

Since fluorine is the major component of both studied insertions, the neutron spectrum shape in fast energy range is driven by its cross section, therefore the insertion design is well suited for neutron spectrum measurement and analysis. Performed experiments showed some discrepancies, however not as large as in comparison with other experiments with FLINA salt. This can be caused by lower importance of insertions in the core, however these experiments are considered as the first steps to larger experimental cores, which will be neutronically closer to the fluoride salt based reactors.

It was observed that increased absorption in thermal part of the neutron spectrum (EROS 3, FLIBE) leads to narrowing of the experimental uncertainty interval in the benchmark configuration. Teflon acts as a good moderator, and when no absorption is introduced, the level of C/E-1 agreement reaches almost to the value obtained for empty channel without any insertion (Case 1), which is considered as a reference. No special scattering matrix was used either for Teflon or for FLIBE salt.

Concerning nuclear data libraries comparison, the RUSFOND-2010 tends to overestimate other libraries, in cases when fluorinated insertion is used. Current experiments showed that the calculations slightly overestimate when none, or almost none ^{6}Li isotope is present in studied compounds.

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