

## NEUTRON FIELD MOCK-UP DEVELOPMENT FOR THE FLUORIDE SALT REACTORS NEUTRONIC RESEARCH

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### ABSTRACT

Experimental work at the LR-0 reactor was carried out to determine the possibility of the mock-up neutron field creation for the fluoride salt-based reactors. Previous calculations and experiments have shown that the fast part of the molten salt reactor (MSR) spectrum is shaped by fluorine and even the Teflon material is suitable for neutronics in that energy range. Properly selected spectrum indices can describe the neutron field of the MSR in fast thermal and intermediate parts of the spectrum. Current research has focused on a deeper study of the possibility of using the filtered thermal and intermediate neutron spectrum of the experimental light water reactor for the physics of fluoride salt-based reactor. LiF-BeF<sub>2</sub> (FLIBE) capsule and teflon cylinders are used as a spectrum filters in the LR-0 reactor. Measured results show acceptable C/E-1 agreement in the reaction rates and satisfactory agreement for usage of the FLIBE filtered neutron spectrum determined by the <sup>181</sup>Ta(n, γ) monitor as a mock-up in thermal to intermediate energy in the fluoride high-temperature reactor (FHR). Concerning MSR, intermediate spectrum can be reproduced to some extent but not as good as in the case of FHR.

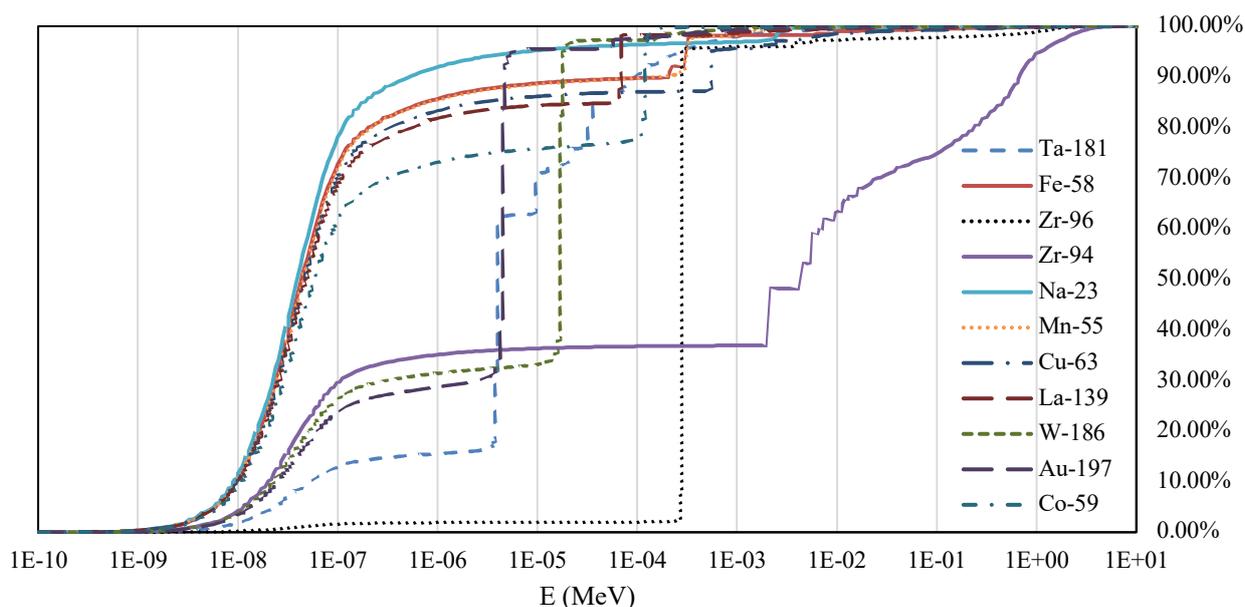
KEYWORDS: LR-0 reactor, MSR, FHR, neutron spectrum, spectral indices, reaction rates

### 1. INTRODUCTION

For reactor technology development, the good knowledge of neutronics plays a crucial role as the parameters of neutrons e.g. spectrum and flux can influence the criticality, safety parameters (feedbacks), or material aging. Previous studies have shown the idea to characterize the neutron spectrum of a molten salt reactor (MSR) by means of reaction rate ratios, the so-called spectrum indices, in the neutron field of a light-water research reactor (LWR) generated by an appropriate filter (see ref. [1]). It was concluded that the calculated spectral indices are sufficiently sensitive to changes in the fast neutron tails of different advanced reactor concepts; at the same time, the neutron spectrum of the MSR showed very good agreement with the filtered spectrum of the light water research reactor by the filter containing LiF-BeF<sub>2</sub> (FLIBE) salt. Moreover, it has been found that the fast part of the MSR spectrum can be simulated in the LWR by other compounds than FLIBE, containing substantial amounts of bound fluorine, for example with Teflon [2]. Following these findings, other general issues must be resolved, such as measuring the spectrum indices with acceptable uncertainty, the extent to which the measurements correspond with calculations, and the precision of the thermal and epithermal part of the neutron spectrum description in the filtered LWR field

and its correspondence with MSR and FHR reactors. For the aforementioned purposes, irradiation in the filtered neutron spectrum can be carried out in an experimental light water reactor.

Reaction rates measurement by means of material activation is a suitable tool for validation of the neutron field in the designed mock-up. The selection of proper material for activation monitors facilitates the neutron spectrum characterization in the thermal and intermediate range via different reaction energy sensitivities. Fig. 1 shows the energy dependence of several (n,g) reactions in terms of their cumulative contribution to the total reaction rate in the thermal and intermediate neutron spectrum. This figure demonstrates how sensitive the different materials are in the observed part of the neutron spectrum, and based on the reaction evolution, proper monitors can be selected for the experiments described in the following paragraphs.



**Figure 1. Cumulative reaction rates calculated in LR-0 core**

## 2. MATERIALS AND METHODS

### 2.1. Modules with FLIBE Salt and Teflon

Described approach used filters for neutron spectrum shaping. Filter, or irradiation module, can be inserted into the dry channel in the reactor core. The FLIBE filter consists of a stainless-steel vessel of a diameter of 20 cm with a mixture of lithium beryllium fluoride salt. It has six cavities for sample irradiation in the filtered neutron spectrum. In the center of the module, there is a larger cavity (inner diameter 6.4 cm) into which a spectrometric device can be inserted. The vessel was pre-heated and the melted FLIBE salt was poured into it to have compact form. As it is, the module is suited for experiments at room temperature. Total amount of the FLIBE salt contained in the vessel is 27.54 kg. The height of the FLIBE column in the vessel is 52 cm which corresponds to the height of 53.6 cm above the fuel column lower end. The rods for insertion of activation foils are placed in smaller cavities (see Fig. 2 left) and numbered as in Fig. 3. In axial direction, levels 3, 4, 5, 6, 7 (Table III) correspond to heights of 11.7, 16.7, 21.7, 26.7, and 31.7 cm. The values are related to the beginning of the fission column level (active part of the fuel), where 0 cm represents the lower end of the fission column.

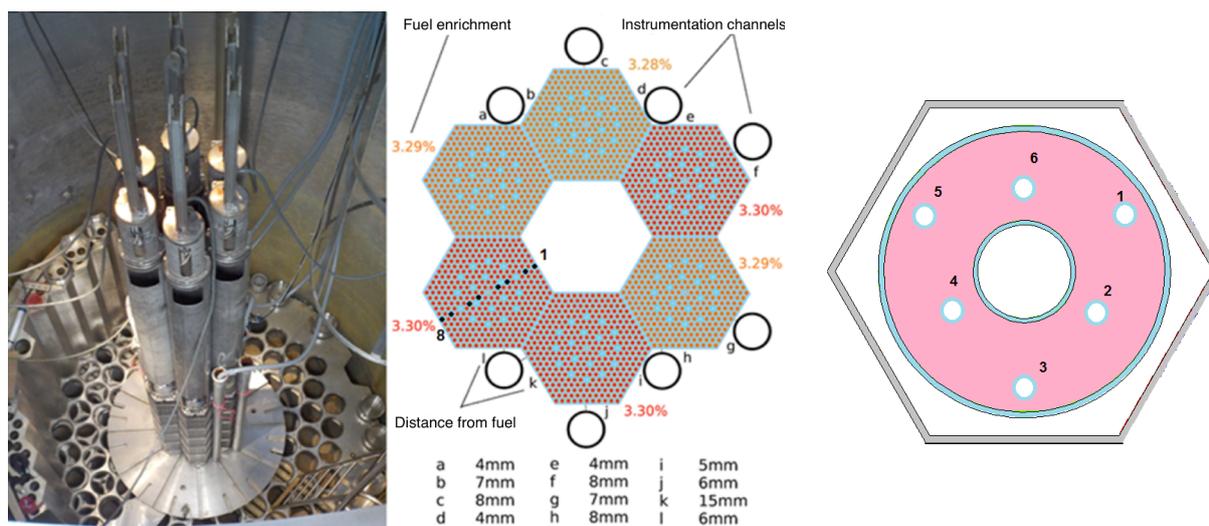
Teflon insertion has the shape of a simple cylinder with the central cavity for neutron spectrum measurement and irradiation. The cylinder outer diameter is 20.5 cm. For irradiation, the foil carrier in the form of aluminum rod with spacer is used as displayed in Fig. 2.



**Figure 2.** From the left: FLIBE module, Teflon module, activation foil carrier.

**2.2. Activation Materials**

To characterize thermal and intermediate part of the neutron spectrum, primarily materials with different  $(n,\gamma)$  reaction evolutions, as shown in Fig. 1, are used. Several materials were chosen for experimental study of reaction rate in FLIBE and Teflon modules. The FLIBE experiment was focused purely on  $(n,\gamma)$  reactions, while for Teflon  $(n,p)$ ,  $(n,\alpha)$ , and  $(n,2n)$  reactions were further studied. For the FLIBE module, materials have been selected as follows: Mn, Au, Co Ta and their placement in the module was done as shown in Tab. I. Different sets of activation foils, Au, Co, Ti, Fe, Ni, Al were used for irradiation in the Teflon module. Radioactive products of the irradiation in the filtered neutron field are detected by the semiconductor high purity germanium detector (HPGe) with verified material and geometry data.



**Figure 3.** Overhead view of the core, its scheme and scheme of the FLIBE module.

### 2.3. LR-0 Reactor

Irradiation of the module was carried out at the LR-0 reactor operated by Research Centre Řež. The LR-0 reactor is a zero-power reactor based on light water pool-type technology. Fuel burn-up is not measurable thanks to the low flux; thus, it can be considered as fresh for all calculations. However, the power is sufficient enough to cause activation of materials in the core center and its vicinity.

The core is composed of six hexagonal fuel assemblies consisting of the same number of pins and having the same face-to-face dimensions as the VVER-1000 fuel. This type of core has been benchmarked [3] in past and the reactivity [4], spatial flux distribution [5], spectrum [6], and power distribution [7] have been characterized. Thus, it acts today as a reference neutron field [8].

Specialty of the benchmark core in the LR-0 reactor is that the criticality is given by the amount of moderator while the control rods are withdrawn to upper position. Water level is then considered as a critical parameter which can be measured with low uncertainty.

### 2.4. Calculations

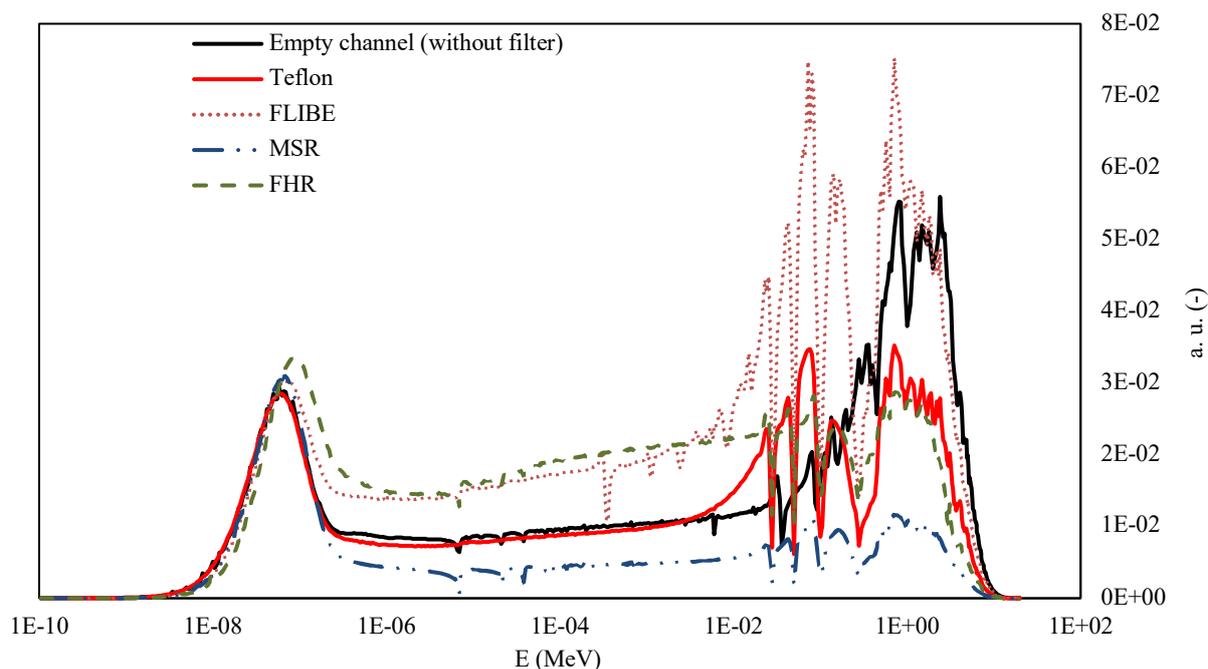
Neutron spectrum and reaction rates of the irradiated foils in the LR-0 reactor have been calculated in critical mode of the MCNP6.2 [9], where the material data characteristics have been defined in the ENDF/B-VII.0 [10] data library. Target materials were defined in other nuclear data libraries as well to observe their sensitivity on the library choice. Neutron spectrum in FHR and MSR reactors has been calculated by Serpent 2 code [11] in version 2.1.30, ENDF/B-VII.0 nuclear data library has been used for these calculations as well. Temperature of all models has been set to 294 K.

## 3. RESULTS

### 3.1. Neutron spectrum

Firstly, the comparison of the expected neutron spectra obtained from calculation models is shown in Fig. 4, where the case “Empty channel” means the central channel without neutron filter, “Teflon” and “FLIBE” are referencing the filters accordingly, “MSR” is a generic model of the single fluid molten salt reactor and “FHR” is the spectrum in the coolant of the small reactor with the plank microparticle fuel [12]. The neutron spectra were calculated in the 640 group (SAND-II) format and are normalized in energies from 0.002 eV to 0.2 eV (to the thermal energy peak at room temperature).

Calculated fluxes were integrated into 3 groups (1 meV – 1eV, 1 eV – 10 keV, and above 10 keV) representing thermal, intermediate, and fast group for the purpose of comparison. It can be seen that the largest portion of the thermal neutrons is present in the MSR system, where only fluid homogeneous fuel is used. The FHR concept is right behind the MSR due to its micro-particle TRISO fuel, which can be considered as semi-homogeneous. Ratios of the neutron share according to their energies in the given spectra are shown in Tab. I. It was found that the thermal to intermediate neutron range of the FHR reactor can be simulated by the FLIBE insertion in the LR-0 reactor because the ratio between thermal and intermediate neutrons is very similar. However, for the monitors acting in the thermal to fast energy range, the Teflon insertion is more suitable. In the case of the MSR reactor, the neutron spectrum can be simulated in the LWR only from the intermediate to fast energy range with the rate of similarity of 70 %. The rates of similarity of the three-group spectrum representation are demonstrated in Tab. II.



**Figure 4.** Comparison of the calculated neutron spectra in the LR-0 reactor with different insertions and reactor concepts using the fluoride molten salt.

**Table I.** Ratios of the group fluxes for different calculation models.

	Empty ch.	Teflon	FLIBE	MSR	FHR
Thermal/Intermediate	82%	84%	51%	163%	52%
Thermal/Fast	35%	48%	29%	131%	64%
Intermediate/Fast	43%	57%	57%	81%	123%

**Table II.** Comparison of the different insertions' similarity.

	Thermal vs. Intermediate	Thermal vs. Fast	Intermediate vs. Fast
Teflon vs. FHR	161.2%	<b>74.8%</b>	46.4%
FLIBE vs. FHR	<b>98.1%</b>	45.2%	46.1%
Teflon vs. MSR	51.3%	36.4%	<b>70.9%</b>
FLIBE vs. MSR	31.2%	22.0%	<b>70.4%</b>

### 3.2. Reaction rates

Monitors of the flux were selected according to the accessibility and response in the thermal, intermediate and fast part of the neutron spectrum. Calculations referred in previous paragraphs showed which insertions are suitable for simulating the different types of fluoride salt-based reactors.

Because the thermal/intermediate part of the spectrum in FLIBE is similar to the FHR, manganese, gold, cobalt, and tantalum were selected for more detailed description in this insertion (for their sensitivity see Fig. 1). The case where Au, Co, and Ta foils were covered by Cd sheets to suppress thermal flux was studied as well and foils placed in the Cd shrouds are denoted by (Cd) symbol in the Tab. III. Because the monitors

were placed in different radial and axial positions of the module, neutron field distribution can be assessed directly from the evaluated experimental reaction rates. According to the reaction rates on manganese (see Tab. III), the thermal flux is homogeneously distributed in the FLIBE module on axial levels between 11.7 and 21.7 cm and does not depend on the thickness of the salt filter (see the carrier positions in the Fig. 3 right). The axial flux homogeneity is confirmed by reaction rates on gold as well. The C/E-1 column shows the percentual correspondence of the measurement with the calculation. All the differences between measured reaction rates are within experimental uncertainties. Thermal neutron field homogeneity is given by the low neutron leakage from the module and its strongly scattering properties. However, the scattering is caused not only by the FLIBE salt filling, but by the structural material of the module as well, which also contributes to the spectrum shaping. Mentioned experimental uncertainties have following components: uncertainty in measured net peak area (NPA), measurement uncertainty, uncertainty in calibration, uncertainty in detection efficiency, uncertainty in sample positioning, and uncertainty in nuclear data constants.

**Table III. Measured and calculated reaction rates in the FLIBE module normalized to one target nucleus and neutron in the core.**

Position of the rod	Position on the rod	Axial level (cm)	Activation foil	Reaction	Measured reaction rate (s <sup>-1</sup> )	C/E-1	Rel. uncertainty
1	5	21.7	Mn1	<sup>55</sup> Mn(n, γ)	1.157E-26	-9.77%	4.7%
2	7	31.7	Au1(Cd)	<sup>197</sup> Au(n, γ)	6.258E-27	5.99%	4.7%
2	5	21.7	Au2	<sup>197</sup> Au(n, γ)	1.281E-26	-13.70%	4.6%
3	5	21.7	Co(A)	<sup>59</sup> Co(n, γ)	2.498E-27	1.67%	5.0%
3	7	31.7	Co(B,Cd)	<sup>59</sup> Co(n, γ)	6.400E-28	-9.02%	5.0%
4	7	31.7	Ta3(Cd)	<sup>181</sup> Ta(n, γ)	3.012E-27	1.36%	4.8%
4	5	31.7	Ta4	<sup>181</sup> Ta(n, γ)	4.302E-27	-0.06%	4.7%
5	3	11.7	Mn4	<sup>55</sup> Mn(n, γ)	1.085E-26	-16.24%	10.3%
5	5	21.7	Mn5	<sup>55</sup> Mn(n, γ)	1.125E-26	-11.02%	4.7%
5	4	16.7	Au3	<sup>197</sup> Au(n, γ)	1.348E-26	2.88%	4.7%
5	6	26.7	Au4	<sup>197</sup> Au(n, γ)	1.351E-26	-9.77%	4.7%
6	5	21.7	Mn3	<sup>55</sup> Mn(n, γ)	1.060E-26	-9.08	4.7%

**Table IV. Cadmium ratios of different thermal monitors, comparison of the reference case without insertion to FLIBE insertion.**

Reaction	LR-0 empty	LR-0 FLIBE			FHR	MSR
	Measured Cd ratio	Measured Cd ratio	Calculated Cd ratio	C/E-1	Calculated Cd ratio	
<sup>59</sup> Co(n, γ)	3.38	2.33	2.60	11.7%	4.34	13.03
<sup>197</sup> Au(n, γ)	1.39	1.23	1.13	-10.9%	2.54	6.05
<sup>181</sup> Ta(n, γ)	1.11	1.14	1.12	-1.4%	1.25	1.94

The shift in the thermal spectrum was tested by means of the cadmium ratios, which are the spectral quantities suitable for thermal spectrum assessment. Tab. IV shows that the measured and calculated Cd ratios in the FLIBE module agree very well in the case of Ta and are satisfactory for Au and Co as well, when the C/E-1 is in the order of 11 %. When compared with the empty channel (measurements without

insertion) of the LR-0 reactor, the ratios of the monitors in the more thermal part of the spectrum are smaller in case of the FLIBE insertion, which points to a slight shift of the spectrum to the intermediate energies. Cd ratio on Ta, which is moved more to resonant region, is very similar in both types of the core. This observation agrees with the theory, because neutron slow-down in FLIBE module occurs on the heavier nuclei. However, the calculated Cd ratios of  $^{59}\text{Co}(n, \gamma)$  and  $^{197}\text{Au}(n, \gamma)$  in the FHR and MSR reactors indicate that the shape of thermal part of the spectrum for the LR-0 reactor is completely different. Therefore, it is only convenient to compare ratios of total amounts of thermal, epithermal, and fast neutrons in spectrum as shown above.

**Table V. Measured reaction rates in the Teflon module normalized to one target nucleus and neutron in the core.**

Position on the rod	Axial level (cm)	Reaction	Reaction rates	C/E - 1	Rel. uncertainty
4	16.7	$^{197}\text{Au}(n, \gamma)$	1.72E-26	0.6%	2.2%
		$^{59}\text{Co}(n, \gamma)$	2.72E-27	-5.4%	2.2%
5	21.7	$^{58}\text{Ni}(n, p)$	5.12E-30	-6.1%	2.2%
		$^{54}\text{Fe}(n, p)$	3.70E-30	-3.4%	2.7%
		$^{46}\text{Ti}(n, p)$	4.83E-31	12.8%	6.6%
		$^{47}\text{Ti}(n, p)$	8.89E-31	-6.1%	2.0%
		$^{48}\text{Ti}(n, p)$	1.26E-32	-7.8%	2.4%
		$^{197}\text{Au}(n, 2n)$	1.37E-31	-8.4%	3.8%
6	26.7	$^{197}\text{Au}(n, \gamma)$	1.62E-26	-0.6%	2.2%
		$^{59}\text{Co}(n, \gamma)$	2.61E-27	-9.0%	2.0%
		$^{58}\text{Ni}(n, p)$	5.20E-30	-12.1%	2.2%
		$^{54}\text{Fe}(n, p)$	3.64E-30	-6.6%	2.7%
		$^{46}\text{Ti}(n, p)$	4.45E-31	16.1%	6.6%
		$^{47}\text{Ti}(n, p)$	8.17E-31	-3.0%	2.0%
		$^{48}\text{Ti}(n, p)$	1.15E-32	-5.9%	2.4%
7	31.7	$^{58}\text{Ni}(n, p)$	4.53E-30	-0.5%	2.3%
		$^{54}\text{Fe}(n, p)$	3.20E-30	4.9%	3.3%
		$^{46}\text{Ti}(n, p)$	4.18E-31	22.1%	5.3%
		$^{47}\text{Ti}(n, p)$	8.30E-31	-5.7%	2.0%
		$^{48}\text{Ti}(n, p)$	1.15E-32	-8.0%	3.2%
		$^{27}\text{Al}(n, \alpha)$	2.07E-32	27.3%	2.2%

It has been found that the ratio between thermal and fast neutrons in the case of the Teflon insertion is somehow similar to that in the FHR reactor. Therefore, thermal and fast monitors were used in the case of the Teflon insertion. The extended set shown in Tab. V refers to the preliminary results presented in [13] where suitable spectrum indices were proposed for neutron spectrum characterization in the MSR reactor.

#### 4. CONCLUSIONS

Calculations of the neutron spectra condensed into three groups which were used to compare ratios of the thermal, intermediate, and fast neutron spectrum in case of filtered neutron field and fields of FHR and MSR

reactors. FLIBE neutron filter in the LR-0 reactor can be used to reproduce the share of thermal to epithermal neutrons in FHR, whereas teflon has better performance in reproducing thermal to fast share in FHR. For MSR, the share of intermediate to fast neutrons can be simulated to some extent by both filters, but teflon performs better overall.

The more detailed study in the thermal and intermediate neutron spectra was carried out by measuring spectrum indices. Co, Au and Ta foils were used to study the Cd ratios. Co and Au are very sensitive since low neutron energies and therefore their Cd ratios are not corresponding neither to the FHR, nor the MSR. It seems that Ta can be used as an acceptable monitor in FLIBE filter to provide information about spectrum similar to FHR. The thermal spectrum of MSR appeared completely different than the filtered spectrum, so other spectral quantities than Cd ratio must be used for spectrum modelling.

In general, it has been shown that the reaction rates in fluorine-containing filters can be measured with satisfactory uncertainties and the mockup of the fluoride-based reactors can be developed to some extent.

### ACKNOWLEDGMENTS

Presented results were obtained with the use of the infrastructure Reactors LVR-15 and LR-0, which is financially supported by the Ministry of Education, Youth and Sports - project LM2015074 and also financially supported by the Ministry of Education, Youth and Sports Czech Republic - project LQ1603 Research for SUSEN. This work has been realized within the SUSEN Project (established in the framework of the European Regional Development Fund (ERDF) in project CZ.1.05/2.1.00/03.0108 and of the European Structural Funds and Investment Funds (ESIF) in the project CZ.02.1.01/0.0/0.0/15\_008/0000293), which is financially supported by the Ministry of Education, Youth and Sports - project LM2015093 Infrastructure SUSEN.

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