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Neutronic experiments with fluorine rich compounds at LR-0 reactor ☆

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ABSTRACT

Research on molten salt reactor (MSR) neutronics continues in Research Centre Rez (Czech Republic) with experimental work being conducted using fluoride salt that was originally used in the Molten Salt Reactor Experiment (MSRE). Previous results identified significant variations in the neutron spectrum measured in LiF-NaF salt. These variations could originate from the fluorine description in current nuclear data sets. Subsequent experiments were performed to try to confirm this phenomenon. Therefore, another fluorine-rich compound, Teflon, was used for testing. Critical experiments showed slight discrepancies in C/E-1 for both compounds, Teflon and FLIBE, and systematic overestimation of criticality was observed in calculations. Different nuclear data libraries were used for data set testing. For Teflon, the overestimation is higher when using JENDL-3.3, JENDL-4, and RUSFOND-2010 libraries, all three of which share the same inelastic-to-elastic scattering cross section ratio. Calculations using other libraries (ENDF/B-VII.1, ENDF/B-VII.0, JEFF-3.2, JEFF-3.1, and CENDL-3.1) tend to be closer to the experimental value. Neutron spectrum measurement in both substances revealed structure similar to that seen in previous measurements using LiF-NaF salt, which indicates that the neutron spectrum seems to be strongly shaped by fluorine. Discrepancies between experimental and calculational results seem to be larger in the neutron energy range of 100–1300 keV than in higher energies. In the case of neutron spectrum calculation, none of the tested libraries gives overall better results than the others.

1. Introduction

Results presented in this work are a part of GEN IV reactor research carried out in Research Centre Rez. Since 1996, reactor physics experiments performed at LR-0 reactor have supported research of reactors with coolant and/or fuel in the form of fluoride salts (fluoride high-temperature reactors [FHRs] and molten salt reactors [MSRs]). Discrepancies between criticality calculations and experiments were discovered during re-evaluations of EROS experiments (Losa et al., 2015) in cases with core configurations containing LiF-NaF salt (40 mol% LiF and 60 mol% NaF with natural Li, referred to as *FLINA* hereinafter) and a combination of FLINA and graphite. Therefore, some of these experiments were repeated to confirm the results. The elemental parts of graphite and fluorinated salts were investigated using integral experiments during 2014 and 2015. Thorough analysis showed that the discrepancies between calculation and experiments in cases with graphite insertions were within 1 σ uncertainty interval in terms of criticality (Košťál et al., 2016). Similar results were also obtained for experiments with FLINA insertion. Experimental uncertainties were reduced by repeating experiments many times and by having a well-characterized core. These assurances enabled acceptance of the experiments as a benchmark into the IRPhEP database in 2017.

Critical experiments provide an integral measure to assess parameters of interest, so even if the comparison of calculation and measurement of this parameter gives correct results, some discrepancies might eventually be present in neutron spectrum rates or reaction rates. Effects of the fast neutron spectrum in the empty (void) experimental channel were studied and analyzed previously by Košťál (Košťál et al., 2015). For an experiment using a graphite insertion, it was found that discrepancies in calculation results

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divided by experimental results (C/E) did not exceed 7% in all energy groups. This value was better than the result obtained when the measurement was taken in the void channel with no material insertion, where the discrepancies were as high as 13%. More pronounced discrepancies are apparent from measurement in FLINA salt, where the C/E – 1 comparison can differ by 40% for specific energy regions. Influence of the FLINA salt on criticality was studied by Losa (Losa et al., 2015), with the conclusion that the ^{6}Li(n,t) reaction data can have a significant reactivity impact. Additionally, description of ^{19}F(n,elastic) reaction could have some impact.

Fluorine-rich compounds were selected for use in further study of the nuclear properties of these substances through neutron spectrum measurement and analysis. Teflon contains a substantial amount of fluorine bound in CF_2 molecules, is readily available, and is compatible with the LR-0 experimental water-moderated reactor in terms of nuclear safety. Based on well-known carbon properties in the LR-0 reactor in the form of graphite (Košťál et al., 2016; Košťál et al., 2015), it was thought that potential discrepancies using Teflon insertions could be due to the properties of fluorine bound in CF_2 molecules. Based on the analysis of this simple compound, more complex materials with fluorine can be studied. Fluorinated salt LiF-BeF_2 (FLIBE) was originally used in the coolant circuit of the Molten Salt Reactor Experiment (MSRE) operated at the Oak Ridge National Laboratory (ORNL) in the United States. FLIBE is a sample of a real material intended for use in advanced reactors and was obtained (US DOE, 2013A) within the framework of a cooperation between Czech Republic and the United States, covered by a memorandum of understanding signed in 2011 (US DOE, 2013B).

2. Methodology of experiments and calculations

The influence of fluorinated materials on reactivity has been assessed in a relative manner by comparing the experimentally determined critical states and corresponding calculations (C/E – 1). Critical experiments were carried out in the benchmark core of the LR-0 reactor (see Fig. 1) (Košťál et al., 2017), and calculations were performed in MCNP6.1 code (Goorley, 2012) using different nuclear data libraries. The cross section sensitivity analysis was performed using TSUNAMI from the SCALE 6.2 software package (Rearden and Jessee, 2016).

2.1. Experimental setup – criticality

Neutronic experiments were performed in the zero power light water research reactor LR-0, with benchmark core composition (see Fig. 1). The benchmark core for insertion material experiments consists of six fuel assemblies, each with a nominal enrichment of 3.3%. The fuel assemblies have similar construction to VVER-1000 fuel except for a major difference in the fission column length (active length 126 cm in LR-0 vs. more than 350 cm in VVER-1000). They are arranged in a hexagonal lattice with a pitch of 23.6 cm. A dry channel for material insertion experiments is located in the core centre and has a hexagonal section fitting into the lattice position.

In the shutdown state, the reactor does not contain any moderator. Criticality of the core containing the material insertion is reached by moderator addition, slowly pumping water into the reactor vessel. Differences in reactivity due to inserted materials are thus reflected by differences in the critical moderator level in different experiments. Precise moderator level measurement enables criticality to be determined with an uncertainty on the order of a few tens of pcm. All control rods are fully removed from the core during these benchmark experiments, so the control rods are only used for reactor shutdown.

FLIBE insertion occurs using a solidified melt in a stainless steel (X5CrNi18) canister. Its shape is suited for neutron spectrum and fission rate measurement in the salt. The canister (Fig. 2) is filled with 27.54 kg of salt with a density of 1.95 g/cm^{3}. The salt contains lithium that is highly depleted in ^{6}Li. The height of the salt column in the canister is 52 cm, so the salt occupies approximately 87% of the available volume in the canister.

The Teflon insertion has a cylindrical shape with a central cavity for the detectors of the neutron spectrometric system. The cylinder

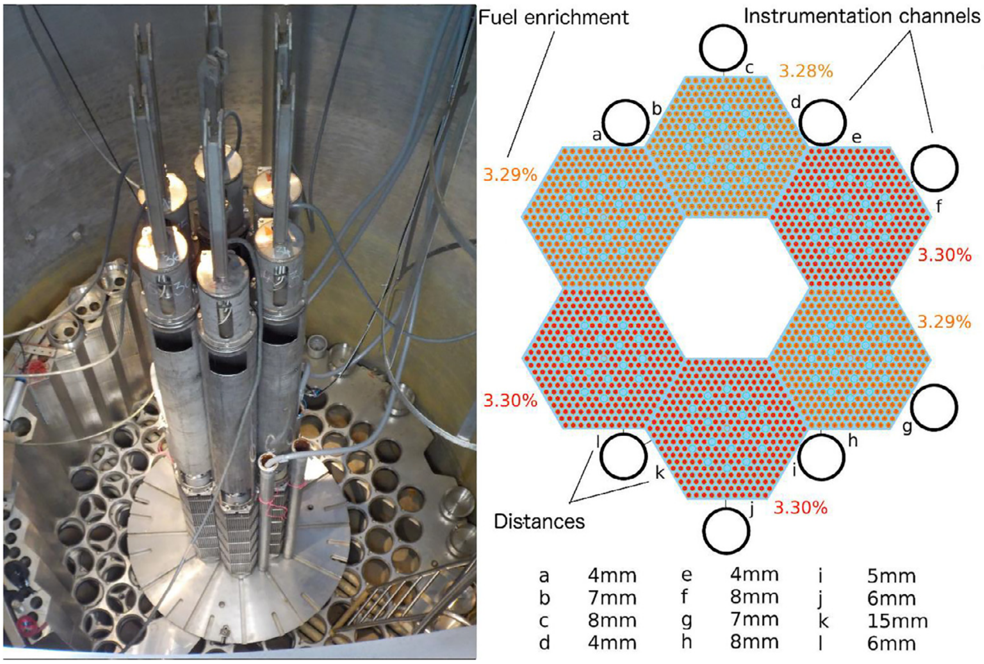

Fig. 1. Overhead view inside the LR-0 reactor special core (left) and radial plot of the core with specified enrichment for each assembly (right) and empty central experimental position (large dry channel).