ANALYSIS OF FAST NEUTRON TRANSPORT IN CHLORIDE SALTS USING MONTE CARLO METHOD

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ABSTRACT.
The aim of this paper is to present results of fast neutron behavior analysis within the chloride salts environment using simulations based on Monte Carlo method (MCNP 6.2). Three non-fueled salts (NaCl, KCl, MgCl2) and two salts containing fissile material (UCl3, ThCl4) were studied. Results of this theoretical study will be used for design of an experimental assembly, which will serve to achieve goals of the international research project ADAR (Accelerator Driven Advanced Reactor). One of the project objectives is to investigate chloride salts as potential coolant and a dissolved fuel carrier of advanced nuclear reactor cooled by molten salts and driven by an accelerator.

KEYWORDS: Accelerator driven systems, fast neutrons, ADAR, chloride salts, MCNP, molten salt reactors, Monte Carlo simulations.

1. INTRODUCTION
Molten Salt Reactors (MSRs) have been identified as promising reactor technology with regard to thorium fuel cycle or for transmutation of spent nuclear fuel from light water reactors. There are several designs based on fuel dissolved in the molten salt coolant, such as fluorides or chlorides [1].

This article is focused on fast neutron behavior analysis within the chloride salts, since they have good nuclear, chemical and physical properties and are much easier to treat. They usually form solutions with lower melting point, and they have higher solubility for actinides. As a result, they can contain significant amounts of transuranic elements. On the other hand, the major isotope of chlorine, Cl-35 gets activated to Cl-36, long-lived energetic beta emitter, so Cl-37 is much preferable in a reactor [1].

As stated above, the goal is to perform studies with fast neutron spectrum since it is used in fast spectrum MSRs (MSFRs) and or accelerator driven systems [2]. They can have conversion ratios within range burner – converter – breeder. Furthermore, they can be optimized for burning minor actinides, for breeding plutonium from U-238, or they may be open-cycle power plants since the fast neutron spectrum allows the possibility to omit onsite processing to remove transuranic elements. Since the fission products have relatively large neutron capture cross sections in thermal energies, the capture cross sections at higher energies is much lower, resulting in much greater fission product build-up in an MSFR than in a thermal-spectrum MSR. Higher absorption rate at fission products nuclei in thermal-spectrum MSR leads to fission products transmutation and therefore they get closer to the valley of stability (after appropriate decay). Moreover, the fuel salt heavily loaded with fission products can be sent occasionally for batch processing or allowed to solidify and be disposed of in a repository. For full breeder configuration the fissile material needs to be progressively removed [1].

Recent research [3] shows significant discrepancies between evaluated nuclear data libraries in the fast neutron region. Improving fast neutron data for chloride salts is essential requirement for the ADAR project (Accelerator Driven Advanced Reactor with molten chloride salt fuel coolant)1. In order to perform such neutronic research, a theoretical study of an experimental assembly consisting of a fast neutron source in chloride salt environment was performed with the aim to optimize parameters for an experimental campaign using activation detectors.

In the following sections, the model is of experimental assembly is described, the current status of

1Detailed project information available at https://starfos.tacr.cz/en/project/LTUSA18196.
relevant nuclear data is summarized and then Monte Carlo calculations of neutron transport through chloride salts are presented.

2. DESIGN OF SALT EXPERIMENTAL ASSEMBLY

2.1. MATERIAL

In order to study neutronic properties of most commonly available chloride salts, a design of a salt experimental assembly “Solnicka” (in English “salt cellar”) has been proposed (Fig. 1). The assembly is cylindrical shape with diameter of 50 cm and height of 60 cm with coating from stainless steel (304 type, thickness of 0.6 mm on sides, 1 mm at the bottom and top cover). It contains the following channels (coated with 0.6 mm thick steel layer):

- one central channel for neutron source (22 mm in diameter) which extends from top to the middle of the height (30 cm),
- four axial experimental channels (28.1 mm in diameter) leading from to the top to the bottom located at radial distance of 5, 10, 15 and 20 cm from the vertical axis,
- three radial experimental channels (28.1 mm in diameter) at heights of 35, 45 and 55 cm above the bottom. Their length is 23 cm spanning from the outer surface of the assembly in direction to the vertical axis.

The experimental channels have been filled with vacuum for simplicity and in order to avoid influence of other materials. As a source of neutrons, the AmBe source (cylinder with diameter of 19 mm and height of 40 mm) located in the middle height (30 cm) with emission rate of $5.5615 \times 10^6$ neutrons/s was considered since it emits fast neutron field (0.1-11 MeV, see Table 1 for probability distribution function (PDF)

![Figure 1. Salt experimental assembly “Solnicka” 3D model](image)

| Salt   | Density [g/cm³] | $k_{eff}$     |
|--------|-----------------|---------------|
| PuCl₃  | 5.71            | 1.27283 ±/- 0.00006 |
| UCl₃   | 5.50            | 0.12304 ±/- 0.00001 |
| ThCl₄  | 4.59            | 0.01936 ±/- 0.00000 |
| NaCl   | 2.18            | -             |
| KCl    | 1.98            | -             |
| MgCl₂  | 2.32            | -             |

Table 1. Energy spectra for considered AmBe neutron source according to ISO-8529-1 [5]

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| KCl    | 1.98            | -             |
| MgCl₂  | 2.32            | -             |

Table 2. Results of multiplication coefficient calculation (using 250 cycles with $10^6$ particles from the source)

overview) standardized ISO-8529-1 [5] and can suitably serve for purposes of this study. To reach maximal homogeneity of the salt environment around the neutron source, a salt plug is considered to be placed above the source inside the source channel.

For purposes of this study, a model of salt experimental assembly has been created using MCNP 6.2 (see Fig. 2) [6]. Three non-fueled salts (NaCl, KCl, MgCl₂) and two salts containing fissile material (UCl₃, ThCl₄) were studied using calculation of reaction rates for set of selected reactions (see Fig. 3). The non-fueled salts serve to investigate their potential influence on transport of neutrons (as a potential ternary salt coolant for nuclear reactors [7], [8], [9]).
UCI₃ (with depleted uranium) and ThCl₄ represent a chloride salt carrying nuclear material – as a reference chemical compound which could be used in a molten salt reactor, where the fuel is dissolved in the coolant. PuCl₃ salt has also been considered, but salt assembly with abovementioned dimensions would be supercritical (see Tab. 2).

Activation foils were selected as neutron detectors. The chosen foil materials are Ni, In and Au. They are mainly mono-isotopic or with one strongly dominating isotope, therefore, the identification of the reaction channel from the observed product is expected to be unambiguous. Non-threshold and threshold reactions of these materials are shown in Fig. 3. Thanks to distribution of the threshold energies between 0.7 and 11.0 MeV it will be possible to investigate sensitivity of activation to various parts of the fast component of the neutron field and their changes in the salt environment.

3. RESULTS AND DISCUSSION
Source driven calculations were carried out with MCNP 6.2 and using the ENDF/B-VII.0 evaluated nuclear data library. Radial and axial reaction rates from the Fig. 3 have been calculated in all experimental channels (numbers 1-4 are vertically sorted by increasing distance from central source channel, 5-7 are horizontally sorted by increasing height) with 10¹⁰ neutrons from the source (resulting in relative error of results lower than 0.01), see examples of the gold activation in the following figures. The observed trends are similar for different activation foil materials (differences are mainly in the absolute values). The results are presented in a systematic comparison in three groups:

- one salt, one foil material, one reaction – different radial and axial channels (Fig. 4);
- one salt, one foil material, one radial or axial channel – different reactions (Fig. 5 and Fig. 6);
- one radial or axial channel, one foil material, one reaction – different salts (Fig. 7 and Fig. 8)

The axial as well as radial distributions of non-threshold (n,γ) reactions were found to be rather broad compared to the spatial distributions of threshold reactions, which are narrower, likely due to their higher sensitivity to the fast neutrons with energies greater than 1 MeV that dominate in the vicinity of the AmBe source. In the axial channel closest to the source, the peak factor of the threshold reactions is about an order of magnitude bigger than that of the non-threshold reactions. In the contrary, the peak
factors of both reaction types are very similar in the furthest axial channel.

In absolute terms, the reaction rates of the non-threshold reactions are about one and two orders of magnitude bigger than those of the threshold reactions at the positions in the middle part of the assembly and on its boundaries, respectively.

Comparison of the spatial distributions of reaction rates in different salt environments provide an interesting comparison, see Fig. 7 and Fig. 8. The spatial distributions of non-threshold reactions strongly differ in different salt environments in any of the axial and radial channels. The most intensive low energy neutron field causing the biggest \((n,\gamma)\) rates was found to be, as expected, in the UCl₃ salt, which contributes to the neutron field by fission. Somewhat surprisingly, the second most intensive low-energy neutron field was found in NaCl, while ThCl₄ is on the third place, which shows its rather small contribution of the fission neutrons (compare with \(k_{eff}\) in Tab. 2). The weakest low-energy neutron field is in KCl. The reaction rates in different salts differ up to a factor of 4.

The threshold reactions corresponding to the fast neutron field component show completely different trends. In the channel closest to the neutron source the reaction rates are almost the same for any of the salt environments. With increasing distance of axial channels from the source, the threshold reaction rates start differing in various salts. The strongest fast neutron field was found to be in KCl while the weakest one in UCl₃ and ThCl₄. That suggests that the down-scattering and capture prevail over the fission neutron production in these fissile materials. This behaviour is valid for any of the reactions with threshold above 1 MeV, while the inelastic scattering in \(^{115}\)In (\(E_{th} \approx 0.4\) MeV) represents a transition region between threshold and non-threshold reactions, see Fig. 9.

A strong influence of different salt composition can be also seen on specific behaviour in the radial channel furthest from the assembly centre, where the neutron leakage dominates causing a decrease in the threshold reaction rates for KCl, NaCl and MgCl₂, see Fig. 8. However, in UCl₃ and ThCl₄ fission locally prevails and causes the maximum to remain close to the assembly center axis. This will be especially interesting feature for comparison with the experiment.

4. CONCLUSIONS
Monte Carlo calculations of fast neutron transport through non-fueled salts (NaCl, KCl, MgCl₂) as well as neutron multiplication in salts containing fissile material (UCl₃, ThCl₄, PuCl₃) were performed. The
Figure 5. Axial reaction rates in gold foils in a UCl₃ salt assembly

Figure 6. Radial reaction rates in gold foils in a UCl₃ salt assembly
Figure 7. Axial reaction rates of non-threshold (left) and threshold (right) reactions in gold foils in five salt assemblies.
Figure 8. Radial reaction rates of threshold reactions in indium (left) and nickel (right) foils in five salt assemblies

Figure 9. Axial reaction rates of threshold reactions in indium (left) and nickel (right) foils in five salt assemblies
spatial distributions of threshold and non-threshold reactions in Ni, In, Au activation foils were investigated. All calculations have been so far performed using the ENDF/B-VII.0 library.

Based on presented calculation results for non-fueled salts, it can be stated, that MgCl₂ or KCl should be more convenient if neutron fast spectrum is desired. On the other hand, NaCl shows higher moderation capabilities. Concerning salts fueled with U and Th, using of ThCl₄ provides slightly higher yields of non-elastic reactions with higher threshold than the UCl₃.

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