**OpenMC Neutronics Evaluation for Fluoride Salts in Molten Salt Reactors**

Victoria Hiatt

## ABSTRACT

Molten Salt Reactors (MSR) can have many different salt carriers. Different fluoride salt carriers have been suggested from the beginning of the Molten Salt Research Experiment (MSRE) at Oak Ridge National Laboratory in 1965. While development and study of different salt carriers has been barred by economical restraints around reactor maintenance and construction, different fuel carriers can be simulated using an open source Monte Carlo code OpenMC. Through differing salt carriers in a pin cell configuration of a Molten Salt Reactor, such as NaF-$ZrF_4$, $BeF_2$, NaF-$BeF_2$, LiF-NaF-$BeF_2$, the neutronics can be studied using the tally methods of OpenMC. More specifically, we will look into k-effective values and flux spectra of the different MSR salts. The outcome of this research paper was that criticality calculations (k-eff values) were different for each pin cell geometry modeled, which suggests an optimal ratio between the size of the moderator and fuel cell exists.

## INTRODUCTION TO THE MOLTEN SALT RESEARCH EXPERIMENT

The first positive advancements of Molten Salt Reactors were demonstrated in the Aircraft Reaction Experiment (ARE) in 1954 in the Oak Ridge National Laboratory (ORNL) [1]. The main issues after the experiment largely focused on the need for materials that demonstrated qualities for low corrosion and could withstand long cycles of use during the reaction. This was in the hope of  creating a low enough cost reactor to supply economical power[2]. The advantages theorized post the ARE included the lack of radiation buildup, continuous gaseous product removal, ease of using mixtures (therefore lowering fuel production costs), a high negative coefficient of reactivity, and the ability for continuous fueling [3]. While aqueous homogeneous systems (AHS) were considered as a possibility when compared to MSRs, MSRs stood to be the more advantageous liquid fuel reactor. This includes operating at a high temperature and still maintaining a low pressure. Due to the high boiling points of salts, MSRs are able to maintain minimal gaseous product buildup whereas AHSs use of water and other similar moderators have low boiling points leading to high pressure buildup. With buildup of radioactive products during fission operation, it is important for moderators in reactors to maintain stability even with prolonged exposure to radioactivity [3]. Due to the natural composition of salts, they allow for breakdown immunity to high energy exposure. During the ORNL experiments it was theorized that fuels with $BeF_2$ were more likely to be suitable for early reactors. Due to the simple makeup of these salts, it was believed that a simple fuel cycle would lead to lower fuel cycle costs [2].

## CURRENT LANDSCAPE OF MOLTEN SALT REACTORS

The aircraft nuclear propulsion programme in the United States wound down in the end of the 1950s as the U.S. turned its focus on supplying power to the civilian grid [4]. However there were still ongoing experiments, focusing on the development of technology issues. Today, MSRs have become an important topic of discussion surrounding future energy efficiency goals. Especially in countries in Europe and others such as China, and Canada. Work on MSRs was focused by the European Union through their Fifth Framework Programme. This introduced the MOST project (2002-2005) which reviewed the safety of MSR technology. Current projects funded by the E.U. include the MIMOSA project [5], focusing on the possible abilities of recycling from light water reactors and the feasibility of current technologies, and the ENDURANCE project, focusing on the safety of MSRs [5]. In Canada, many important contributions to the furthering of nuclear research have been made by Terrestrial Energy with their Integral Molten Salt Reactor (IMSR) [6]. Due in part to their excessive contributions and also to their furthering of trying to create more economical reactors, the Canadian government awarded 20 million Canadian dollars in 2020 to Terrestrial Energy for the continuation of research toward producing a reactor that could support the public [6]. In 2011, the Chinese Academy for Science created the Thorium Molten Salt Reactor program. The goal of the TMST programme was to study its safety and economic use[6]. They have developed simulators and test reactors to further the research on creating economic power. In the US, there are many nuclear research projects underway. The U.S. Department of Energy gave 16 million USD to the future of research with TerraPower's Molten Chloride Fast Reactor Technology (MCFR) [7]. This grant went to TerraPower, Southern Company, Oak Ridge National Laboratory, the Electric Power Research Institute and Vanderbilt University in 2016 [7]. For the MCFR they have been focusing on using molten salts as both the fuel carrier and the coolant within the reactor core [7].

## BACKGROUND ON IMPORTANCE OF FLUORIDE SALTS

NaF-$ZrF_4$ was the main salt used in the Aircraft Reactor Experiment in 1954. Upon theorizing which salts could be used at carriers, one of the main studies of interest was the low vapor pressures of Fluoride salts [8]. Another high importance of fluoride salts is the high thermal conductivity as well as the small neutron cross section [9]. During 1940-1970, fluoride salts were ones of high interest due to the above mentioned reasons. After the 1970s, interest in nuclear power rapidly diminished due to anti-nuclear activism and government regulatory shifts, resulting in a corresponding decrease in focus on properties of different nuclear salts associated with the ORNL experiments. However, in the last 30 years, nuclear power as an approach to clean energy in the face of rising carbon emissions and global warming due to high dependence on fossil fuels, there has been a resurgence in the interest in nuclear salts. And with it, the studies on the importance of fluoride salts. In early studies, the main questions around the difficulties on fluoride salts included the high corrosivity of these salts. This is due to impurities of the salts. These impurities lead to breakdown of the materials in the salt carriers and the salts themselves [9].

## DIFFERENCE BETWEEN THE SALTS

In the beginning of studies in ORNL of different salts, the carrier $BeF_2$ was mentioned multiple times in its importance of its exceptionally low neutron absorption, and its ability to modify the properties of other salts (NaF, LiF) [2]. Neutron absorption is the ability for an atom to capture a neutron in the nucleus, forming a heavier atom. While this can be beneficial for a reaction if it is critical, if too many neutrons are absorbed during a fission reaction, it slows down the reaction. This low neutron absorption is why it was pushed to be a part of the salt mixture and was theorized to have a breeding ratio of around 1 [2]. NaF-$ZrF_4$ was used in the ARE with a molar ratio of 53.09–40.73–6.18 [6]. The chosen salt performed well in circulation tests and the salt mixture of NaF, which lowered the melting point and stabilized the mixture, and $ZrF_4$, which provided high solubility for the fuel use. This salt allowed for the first successful liquid fuel reactor. For every salt, a fuel of $UF_4$ was used. Both isotope U235 and U238 were used with an enrichment of 5% of U235 for every model.

## BACKGROUND ON MONTE CARLO SIMULATIONS

Monte Carlo calculations are a statistical way of mathematically predicting randomness. This way of calculating allows for a repetition of random sampling to model more complex systems. First discovered during research for the Manhattan Project, the Monte Carlo method was named after the Monte Carlo Casino, which was the source for inspiration. During the project, scientist Stanisław Ulam was attempting to find the neutron diffusion that would occur in the core of a nuclear weapon. Even with a large amount of data, he found he could not accurately predict what would happen in the core[10]. This is when, due to casino games, he proposed randomization as a way of more accurately depicting what would happen. Monte Carlo simulation uses a repetition of random sampling to conclude on a statistical outcome. Monte Carlo simulations are used in nuclear physics for simulating the interaction that individual particles have with the matter around them. It is especially useful for modeling complex geometry which goes along with a nuclear core and its individual parts. Monte Carlo methods allow for simulations without simplifications, which are oftentimes needed for deterministic methods of calculations.

## WHAT IS OPENMC?

OpenMC is an open-source Python package that simulates neutral particles, such as neutron and photon reactions. It allows for the tracking of different evaluations such as fission rates and neutron flux in what are called 'tallies'. Originally created by the Computational Reactor Physics Group at MIT, it's focused on neutron criticality calculations. It uses Monte Carlo calculations to interpret the outcome of different situations rather than tracking the neutrons deterministically. The use of OpenMC allows for 3D modeling of nuclear reactors in Euclidean space [11].

## MODELING AND SIMULATION PARAMETERS

## OPENMC GEOMETRY

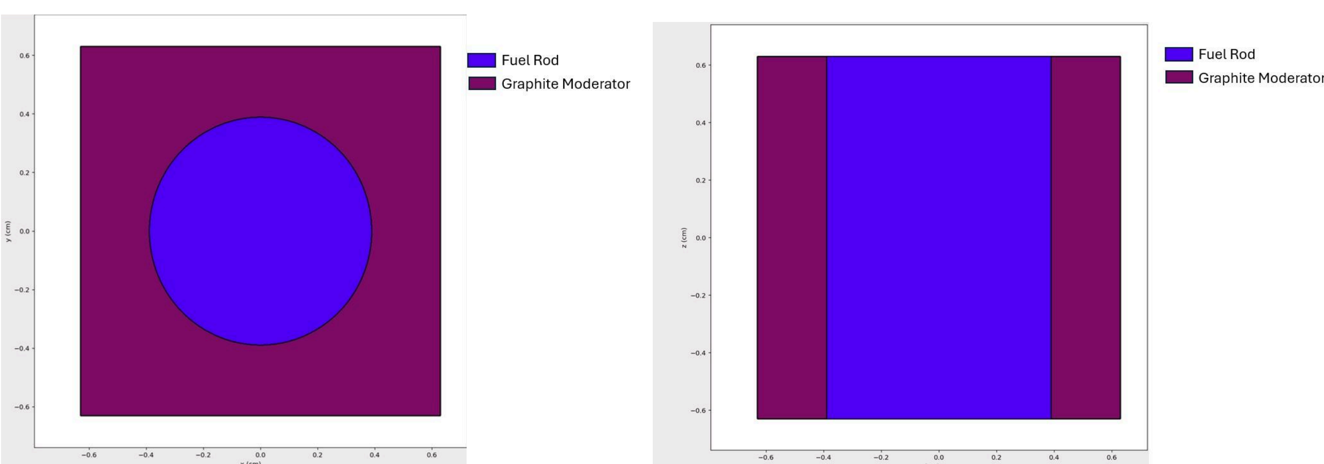


Figure 1(a). Left: OpenMC geometry plot, XY-axis.

Figure 1(b). Right: OpenMC geometry plot, XZ-axis.

In Figure 1(a), the pin cell is depicted with a xy view of the model. The circle in the middle shows the cylinder of the fuel rod in the model. The outer box depicts the moderator, made of graphite. Figure 1(b) on the right depicts a xz slice of the model, showing the fuel cell (blue) stretching the entirety of the pin cell surrounded by the moderator. This geometry makeup consists of just a single pin cell. In a typical reactor, the core is the building blocks on the lattice that makes up the core. A single pin cell is an important way of demonstrating a nuclear reaction as it simplifies calculations and simulating overly complex models of the entire core. Due to this, small changes in a pin cell's reaction create a large change in the overall energy outcome of the entire reactor.

## OPENMC MATERIALS

For the material breakdown of each model, I had a ratio of 1 to 1 for every compound within the mixture. From there, the atom percentage was calculated assuming that each molecule was 0.333 (⅓) or ¼ of the total mixture. Every salt carrier base was modeled with $UF_4$ for its fuel. Uranium was modeled with U235 and U238 with an enrichment of about 4% - 5%, so that each model had the same enrichment respectively.

| FUEL TYPE | PROS | CONS | Use-cases |
|---|---|---|---|
| $NaF\text{-}ZrF_4$ | Good chemical & thermal stability; low vapor pressure | More corrosive to metals than LiF-based salts | ARE |
| $BeF_2$ | High moderating power, low parasitic absorption | Toxic, requires careful handling | MSRE |
| $NaF\text{-}BeF_2$ | Low cost coolant high moderating power ($BeF_2$), good solubility | Highly toxic | ThorCon TMSR |
| $LiF\text{-}BeF_2$ | Excellent heat transfer, high moderating power, low parasitic absorption | Sodium can absorb neutrons | MSRE |

Table 1. Comparing the salts to be used in the reaction, and depicting the use cases.

## SETTINGS

For the simulations, fifty batches were used. In each batch 100,000 neutron particles were simulated. For a pin cell model this small, this number of particles and batches produced good enough convergence. These particles came from a neutron source located at point (0, 0, 0) in each model. The neutron source was isotropic for each model. The lack of direction bias in an isotropic spawn point allows for a more accurate simulation of the outcome of the particles. The use of random sampling allows the Monte Carlo method to capture the statistical nature of neutron behavior, and repeating this process over many batches leads to an accurate estimate of the multiplication factor K-eff.

## TALLIES

Tallies are tracking particles in simulations, following their contributions to different interactions. In nuclear physics, tallies often follow the interactions neutrons make with other particles as neutrons are central to the fission that happens inside a nuclear reaction. In our simulations, we are simulating criticality and flux. In OpenMC, tallies are assigned to regions. In our models, the tallies were assigned to the fuel rod as that is where the reactions would be happening. Tallies are averaged over many particle histories together calculated during a simulation.

We are calculating:

1. **Criticality**

In neutronics simulations, the effective multiplication factor (k-effective) quantifies the ratio of the neutrons in one generation of the reaction compared to the neutrons in the proceeding generation. For an ideal stable reactor, a K-eff of 1 is required. Anything less than 1.0 is considered subcritical and the reaction is dying. Anything above 1.0 is critical, meaning the reaction is becoming unstable and increasing.

2. **Flux**

Flux is the amount of neutrons crossing over a specific area over a given amount of time(n/(cm²·s)). Flux governs power distribution inside a nuclear reaction. In a pin cell, flux controls fuel burnup and depicts reaction rates. Since reaction rates are proportional to the neutron flux and cross section in a nuclear reaction, this means that flux distribution is essential for predicting other quantities such a fission power and absorption.

## RESULTS

For a base line model, the radius was 0.39 cm. The pitch, defining the bounds of the pin cell model, was 1.26. The highest of the k-eff are all relatively similar, yet still all are below ideal (1.0). For the second generation, keeping the ratio of the radius and pitch the same, each model's radius and pitch were increased by a factor of 100, 39 and 126 respectively. For the third generation, the radius and pitch were again increased, this time by a factor of 10 (390, 1260).

| Carrier Salt | Geometry 1<br>Radius = 0.39 cm<br>Pitch = 1.26 cm | **Geometry 2**<br>Radius =39 cm<br>Pitch = 126 cm | **Geometry 3**<br>Radius =390 cm<br>Pitch = 1260 cm |
|---|---|---|---|
| NaF-$ZrF_4$ | 0.75141 +/- 0.00057 | 0.88796 +/- 0.00046 | 0.50454 +/- 0.00575 |
| LiF-$BeF_2$ | 0.88956 +/- 0.00040 | 1.03609 +/- 0.00043 | 0.65300 +/- 0.00509 |

| | | | |
|---|---|---|---|
| $NaF\text{-}BeF_2$ | 0.87256 +/- 0.00043 | 0.97376 +/- 0.00043 | 0.58677 +/- 0.00751 |
| $BeF_2$ | 0.73307 +/- 0.00042 | 0.88275 +/- 0.00048 | 0.50431 +/- 0.00954 |

Table 2. The k-effectives of the different salt carriers for each geometry configuration.

## DISCUSSION

With the increase of the size of the model, we observed the changing k-effectives for every model. With the changing k-effectives, we attempted to observe the optimal area ratio between the size of the fuel rod in comparison to the size of the surrounding moderator. Originally, we thought the k-effective would continue to increase as the size of the fuel rod continued to increase but it became apparent that there was an optimal ratio between the size of the moderator in comparison to the fuel rod.

### Initial K-effective

As depicted in figure 2 the initial k-effectives are all significantly below 1.0. This means that the reaction within every pin cell of this model is dying, not allowing for a stable reaction. In comparison to the other models, $BeF_2$ performs substantially worse than its subparts $NaF\text{-}BeF_2$, $LiF\text{-}BeF_2$, and $NaF\text{-}ZrF_4$. Even with Berillium, which is a high neutron multiplier, the salt $BeF_2$ does not perform well. This implies that the theory that this carrier salt did best when paired with another salt is correct.

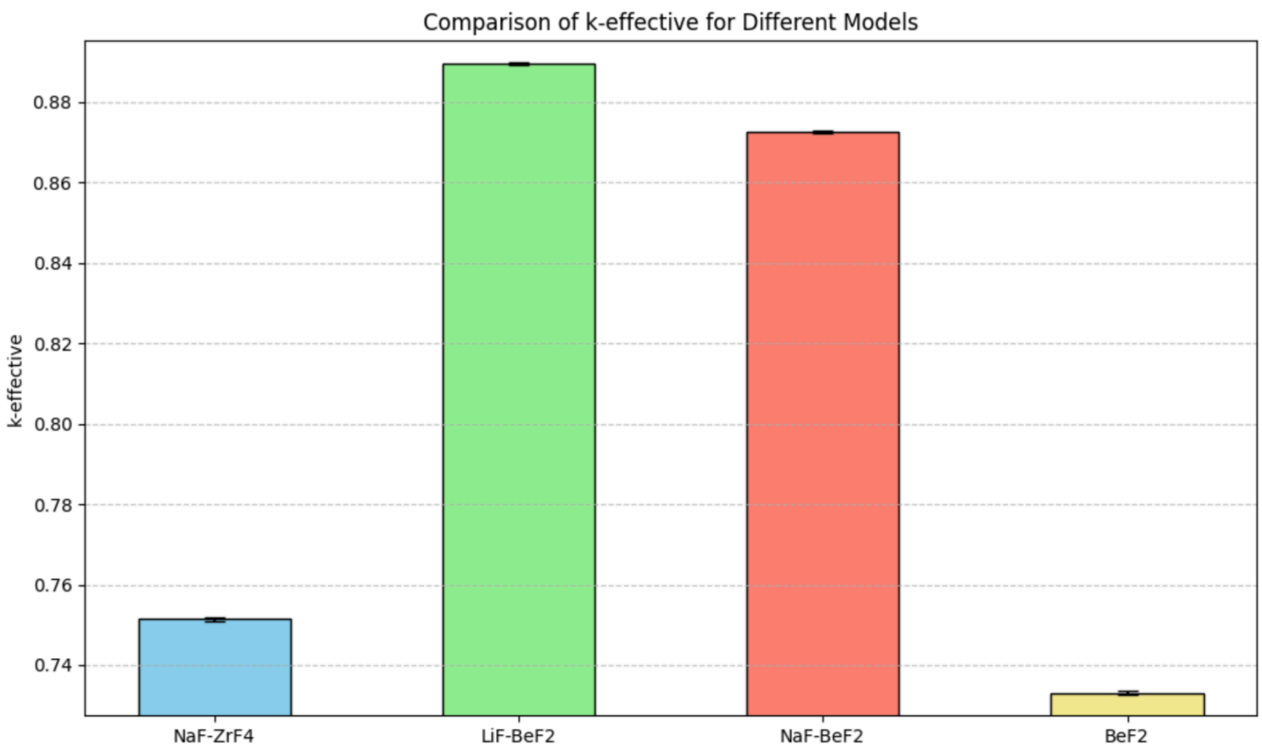

Figure 2. The initial k-effective for each carrier salt modeled against each other, each modeled with a fuel of $UF_4$ with error bars

## Second K-effective and Flux

The next depiction of the pin cells depicts that the increase in both the fuel rod and moderator lead to an increase in K-effective. As shown in figure 3 the k-effective for the salt carrier LiF-$BeF_2$ is above one (1.03609). Around 36,090 ppm above 1.0, this would make for a very critical reaction for the entirety of a reactor given that we are simply modeling pin cells. This suggests that the optimal size of a pin cell for this model is somewhere between the initial model and the second generation.

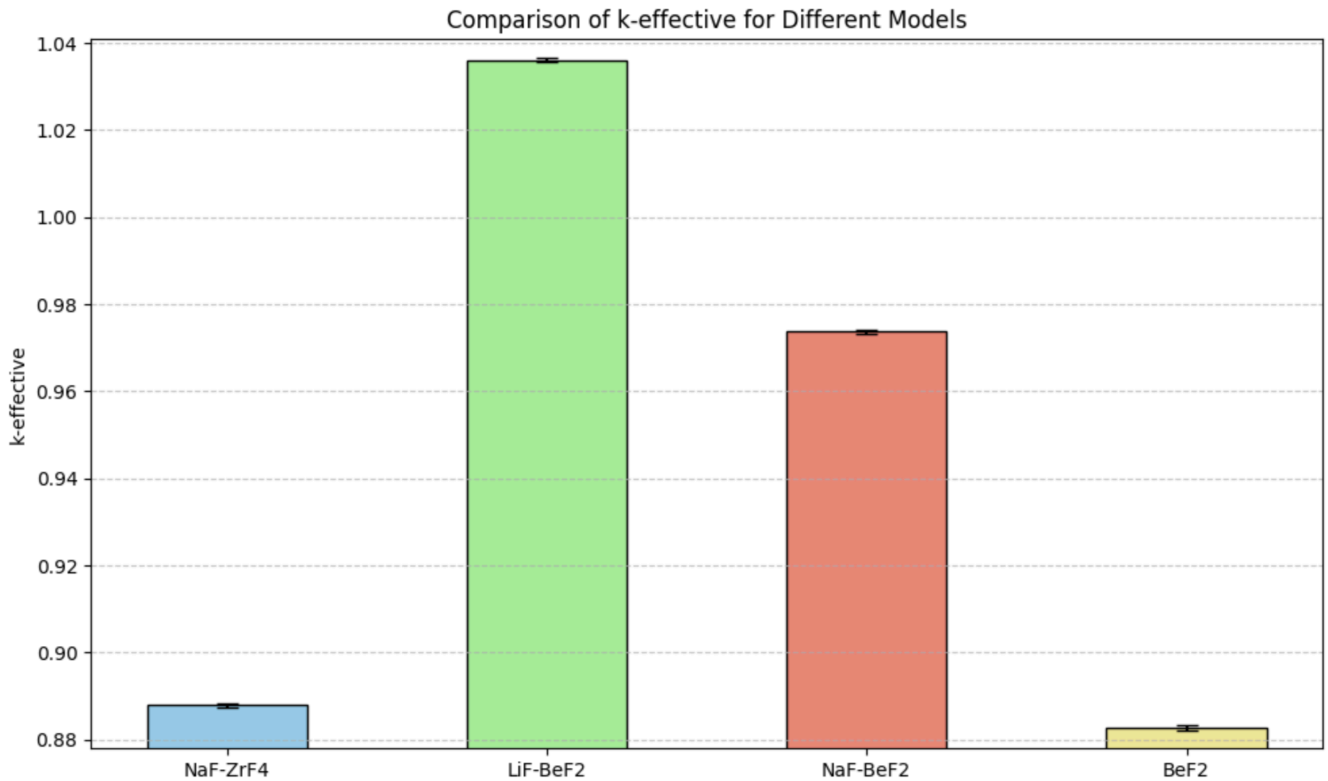


Figure 3. The second k-effective for each carrier salt modeled against each other, each modeled with a fuel of $UF_4$ with error bars

## Third K-effective

Given these first two simulations, it is easy to believe that a continual increase in the fuel rod and moderator will lead to a continuous increase in the k-effective. By observing figure 4 it becomes clear that the optimal ratio between the volume of the fuel rod and the moderator peaks somewhere before the third generation. As the volume of the moderator increases in comparison to the fuel rod, more and more particles are slowed down and absorbed by the moderator. This means that these neutrons are not contributing to the reaction.

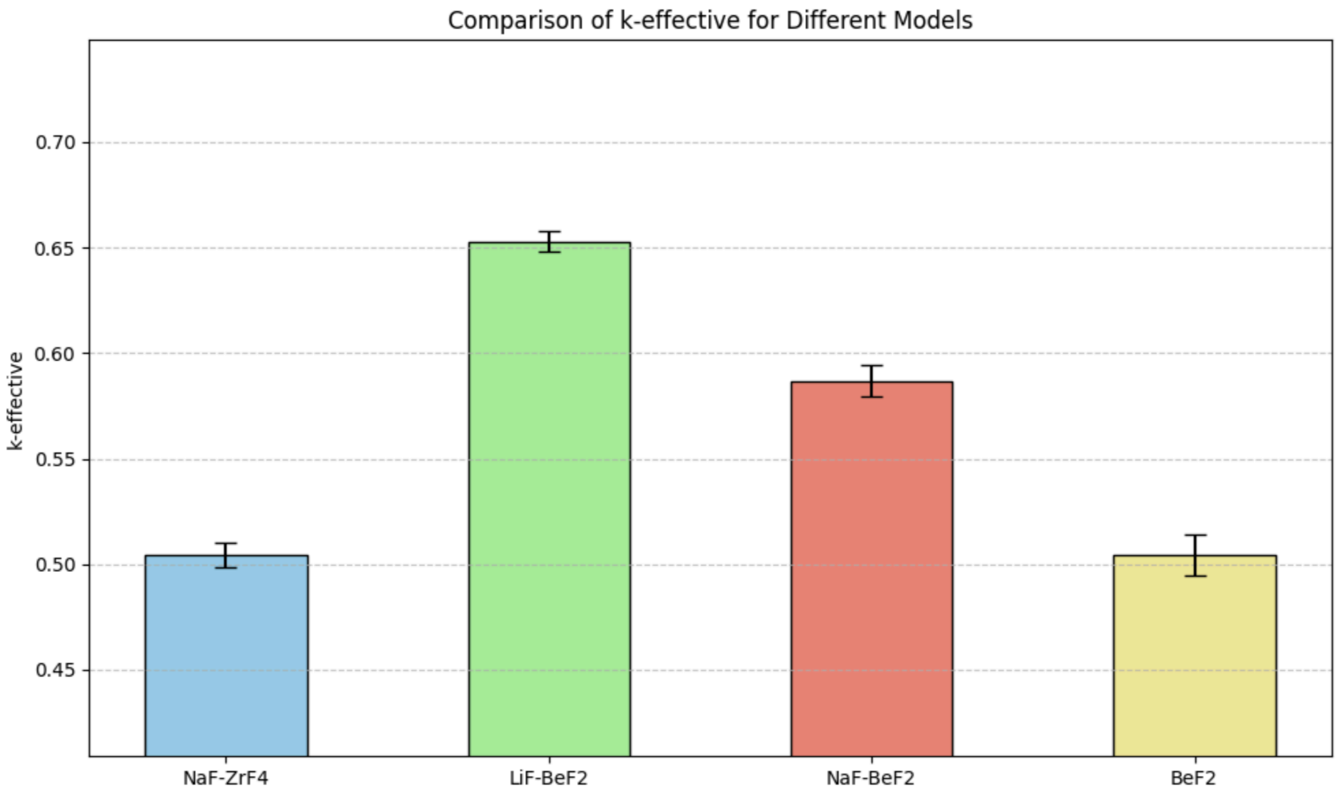


Figure 4. The final (third) k-effective for each carrier salt modeled against each other, each modeled with a fuel of $UF_4$ with error bars

## Flux

As shown in figure 5, the four different models of the geometry are showing the flux calculated based on a pitch of 126 cm and a radius of 39 cm. As the overall shape of each of the models is very similar, it reveals that their energy neutron distributions are very similar. At every point in the range, the models only different very slightly from one another. The relatively similar outcomes reveal that burnup rates in each individual pin cell would be very similar. Although NaF–$BeF_2$ shows the highest flux per source over most energies, the k-effective results favored LiF–$BeF_2$. This suggests LiF–$BeF_2$ converts neutrons to fissions more effectively (e.g., slightly better moderation or lower parasitic capture), even though its absolute flux is not the highest.

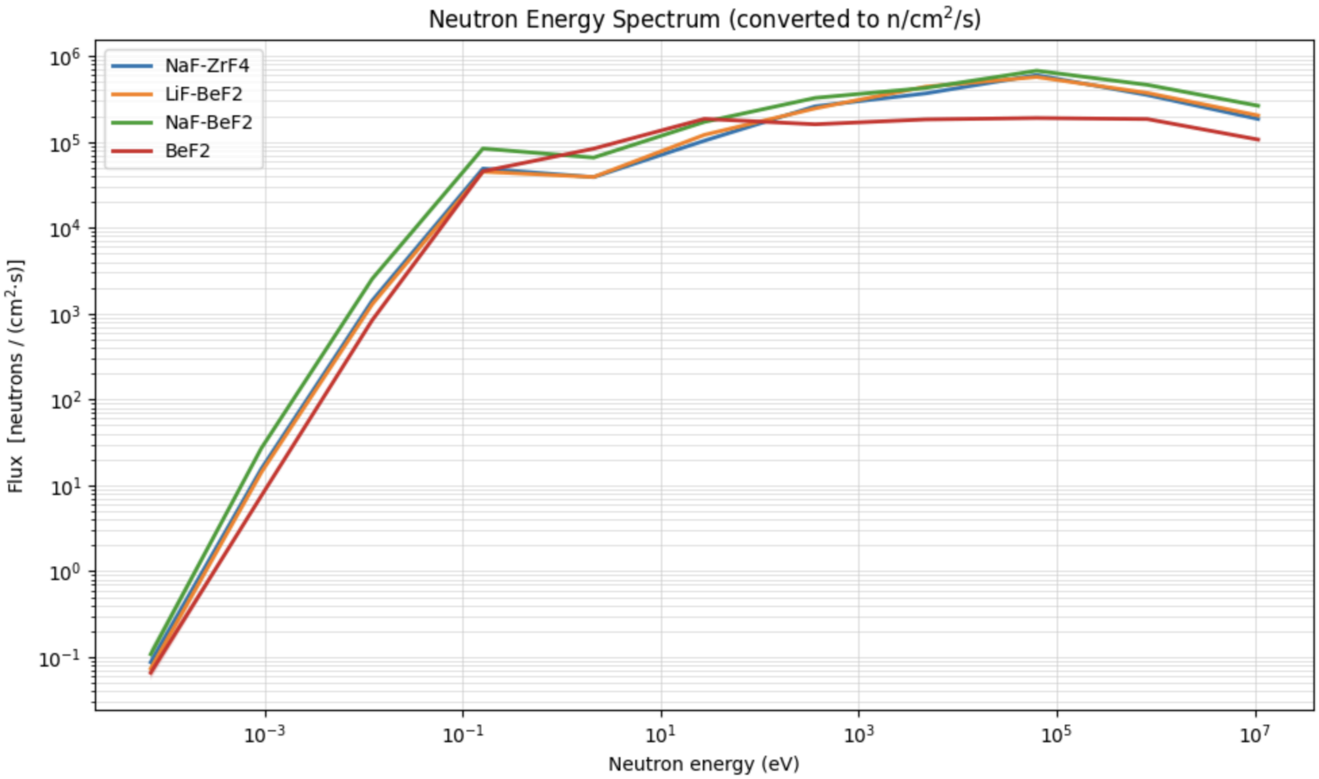


Figure 5. Neutron flux spectra of four molten-salt reactor models ($NaF$-$ZrF_4$, $LiF$-$BeF_2$, $NaF$-$BeF_2$, and $BeF_2$) showing volume-averaged flux versus energy on log–log axes for a $1 \times 10^{10}$ n $s^{-1}$ source.

## CONCLUSIONS

While the k-effectives were different for each model, the changing k-effectives throughout the simulations suggest an optimal ratio between the size of the moderator and fuel cell. The difference in k-effectives do imply a strong inequality in performance due to chemical compounds in nuclear carrier salts in MSRs. For future implications, this could imply an optimal size for every pin cell within a nuclear reactor to obtain a continuous reaction. This optimal geometry would contribute heavily in the search for a nuclear reactor that could provide economical power. The neutron flux spectra for all four salts were nearly identical in shape and magnitude, indicating that changes in salt chemistry have minimal impact on the neutron energy distribution within the pin cell. This similarity suggests that, at the reactor scale, fuel burnup and core power distribution would remain comparable across these salt options, allowing geometry and material considerations in comparisons to k-effectives to drive design choices more than spectral differences.

In the future I look to model different nuclear salts used in research today, continuing with the well studied and important $LiF$-$BeF_2$ (FLi-Be) to look at overlaps in optimal geometry set ups within pin cells.

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