Proceedings of the INMM & ESARDA Joint Virtual Annual Meeting
August 23-26 & August 30-September 1, 2021

Online Monitoring Of Plutonium Concentration Via Frequency Analysis

Alexander M. Wheeler & Ondřej Chvála

University of Tennessee, Department of Nuclear Engineering, Knoxville, TN 37996-2300
awheel13@vols.utk.edu

Abstract

Safeguarding liquid-fueled reactors has often been viewed as a difficult task. Current reactor safeguarding regimes are focused on solid fuel systems and employ item accounting. We developed a novel means of determining plutonium concentration by examining a molten salt reactor power plant frequency response. This method acts as a means of online monitoring of the plutonium concentration evolution with burnup. The frequency response shifts in a predictable manner as plutonium builds up in the fuel salt. If any plutonium were to be removed, a characteristic change in the frequency response can be easily and quickly detected. This work presented in this paper explore the design options for preforming the frequency analysis on a thermal spectrum molten salt reactor. Our results indicate that an ex-core reactivity oscillator is feasible.

1 Introduction

In the Atomic Age, the non-proliferation of nuclear weapons is a crucial part of modern diplomacy. As such, developing a robust safeguarding regime for new nuclear reactors is essential for their global deployment. However, the creation of safeguarding for new designs is often not prioritized. Given the competitive nature of the energy market, reactor designers frequently focus on how their reactors fit into the market rather than considering the details of safeguarding. While it is important to consider large picture aspects such as the market, implementing nuclear safeguards during the design phase will likely be a critical piece of reactor international marketability. When safeguards are developed and implemented post-design completion, there is potential for the safeguards to be unnecessarily complex and intrusive to the operation of the reactor. Creating safeguards after a design is complete can also contribute to more costly implementation of the safeguards. On the other hand, using the Safeguards-by-Design approach can reduce the cost of implementation while potentially giving valuable information to the operator itself [1].

Molten Salt Reactors (MSRs) are reactors that use molten salt as a primary coolant, and typically that coolant is a fuel-salt mixture. These Gen IV reactors are currently in the design phase, and the majority of research about MSRs is focused on the features of design, component technology development, and future deployment. However, it would be beneficial to focus on the safeguards needed for MSRs while they are still in the design phase by following the Safeguards-by-Design approach. There are many design features unique to MSRs that make implementing current safeguard technology a challenge, which is why it is crucial to research MSR safeguards now.

Often, when one thinks about reactor safeguards, they think about the safeguards that are currently in place for Light Water Reactors (LWRs). LWRs are considered item facilities. Current reactor safeguarding regimes in item facilities are focused on solid fuel systems and employ item accounting. The bulk nature of the liquid fuel in an MSR is not amenable for item counting. More appropriate safeguards for MSRs would be those used in bulk handling facilities, such as those that are in place at reprocessing facilities. However, the material streams at reprocessing facilities are aged (thus emitting less radiation), whereas MSR fuel is actively fissioning.

A common practice in safeguarding reprocessing facilities is using process monitoring. In process monitoring, certain state variables and how they evolve are observed. If an operator is not running the facility as declared, and possibly diverting special nuclear materials, then these chosen state variables would not evolve as expected. This is where MSRs deviate from existing bulk handling facilities, as the process of taking energy from the fissioning of nuclear material is very different from reprocessing.

The work presented in this paper considers the potential safeguards of MSRs. We developed a novel means of determining plutonium concentration by examining an MSR's frequency response. This method acts as a means of online monitoring of the plutonium concentration evolution with burnup. The frequency response shifts in a predictable manner as plutonium builds up in the fuel salt. If any plutonium were to be removed, a characteristic change in the frequency response can be easily and quickly detected. This was proved in a previously published paper [5]; the research presented here explores the question of how to properly implement a device to perform the frequency analysis mentioned above.

2 Frequency Analysis

When reactivity is introduced to an operating core, the reactor will have a power response. This response is the result of many prompt and delayed reactivity feedback effects. The power response of a reactor will differ for varying frequencies of reactivity input. For instance, if an operator were to change positions of the control rods, such as moving the rods out of the core, the decrease in neutron absorption would result in a positive reactivity transient. If this were to occur, there would be an increase in power and delayed neutron precursors. The core would also increase in temperature; given the reactor having a negative temperature feedback, it would have its own negative insertion and the reactor power would reach a new equilibrium level at a higher temperature. These effects are on different time scales. As a result, periodic reactivity insertions will have different responses, depending on their frequency.

This is the case when considering MSRs. As a wave of reactivity is inserted in the core of an MSR, corresponding temperature and delayed neutron precursor waves are created and circulated in and out of the core. These waves can either destructively or constructively interfere with each other. Measuring how the reactor responds to different frequencies of activity insertion is known as frequency analysis. For a sinusoidal reactivity insertion with a small enough amplitude, the reactor will have a corresponding sinusoidal power response of the same frequency. However, the response will have a different magnitude and phase. An example of this can be seen in Figure 1. The comparative change in magnitude is known as the gain. The differences in phases is known as phase shift. These data are typically compiled together in a single graph known as a Bode plot.

Performing frequency response tests have also been proposed for use in a molten salt

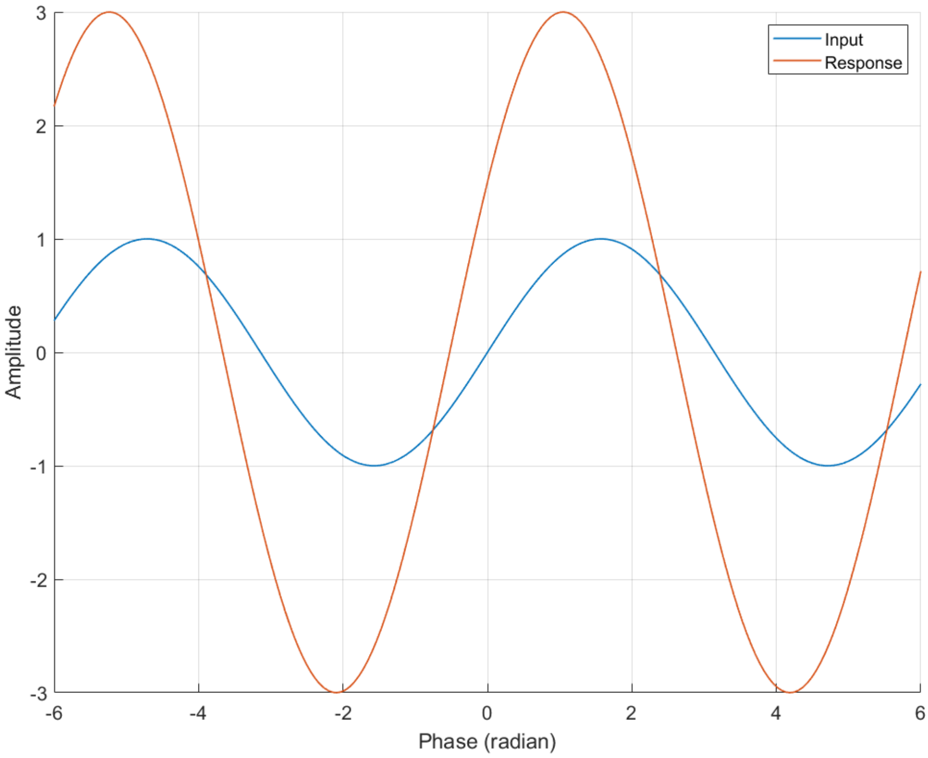

Figure 1: A frequency insertion and frequency response with a gain of 3 and phase shift of 30°.

safeguarding regime [5]. As the reactor undergoes burnup, plutonium will naturally build up in the fuel salt and become responsible for an increasing portion of the fissions. This change in fissile causes a corresponding change in the frequency response of the reactor. Additionally, if plutonium is diverted from the fuel, a characteristic shift in frequency response can be observed. Conducting these measurements would produce only minor perturbations in the temperature ($>$0.1°K given Molten Salt Reactor Experiment feedback coefficients) and produce results within minutes.

3 Instrumentation

Frequency analysis is often performed on reactor designs for many reasons, such as assessing theoretical models and determining stability margins in reactor operations [2]. Frequency analyses tests were originally performed on reactors using the oscillator method, which has a sinusoidal reactivity input. The test is typically performed using a piece of hardware that was accommodated for in the core design of a reactor. Other methods were developed to perform frequency response tests, such as the pseudo-random binary sequence done for the only power operated MSR in history: the Molten Salt Reactor Experiment (MSRE) [3]. The pseudo-random binary uses a sequence of control rod insertions and withdrawals to obtain the response of the reactor over a wide range of frequencies. This method was chosen for the MSRE for two major reasons: cost and speed. The use of a control rod instead of an oscillator reduced equipment cost, and a pseudo-random binary sequence provided data on the entire spectrum of frequency response in as little as 15 minutes. The frequency response was measured using ex-core neutron detectors observing the flux.

Equipment for use in international safeguards have different requirements. First and foremost, the reactor operator cannot have control over the equipment (cite something). Moreover, the reactor operator would not like inspectors to control instrumentation that can potentially disturb operation. These requirements rule out the use of control rods for initiating reactivity events. Instead, returning to an oscillator design would be more preferable for safeguards. In the ideal scenario, such an oscillating reactivity device could be built outside the reactor vessel. Much of the cost came from accommodating the oscillator inside the core. An ex-core oscillator will increase dependency while minimizing intrusion to core design. However, this will likely be design dependant.

3.1 Serpent Calculations

Previous work with frequency analysis for safeguards utilized 2-D infinite lattice neutronics calculations to derive dynamic parameters [5]. On-going research is extrapolating this research to 3-D core designs. Such a 3-D Low Enriched Uranium (LEU) fueled, thermal spectrum reactor is rendered in Serpent 2 [4]. A slice of the core is depicted in Figure 2. This core uses graphite slabs as the moderator and LiF-BeF$_2$-UF$_4$ (72-16-12 mole%, 99% Li-7, 1.3% U-235, 3.353 g/cm^3) as the fuel salt. The core is surrounded by a 2 cm thick SS-610 wall. For this research, the model is used to evaluate the feasibility for an ex-core reactivity oscillator.

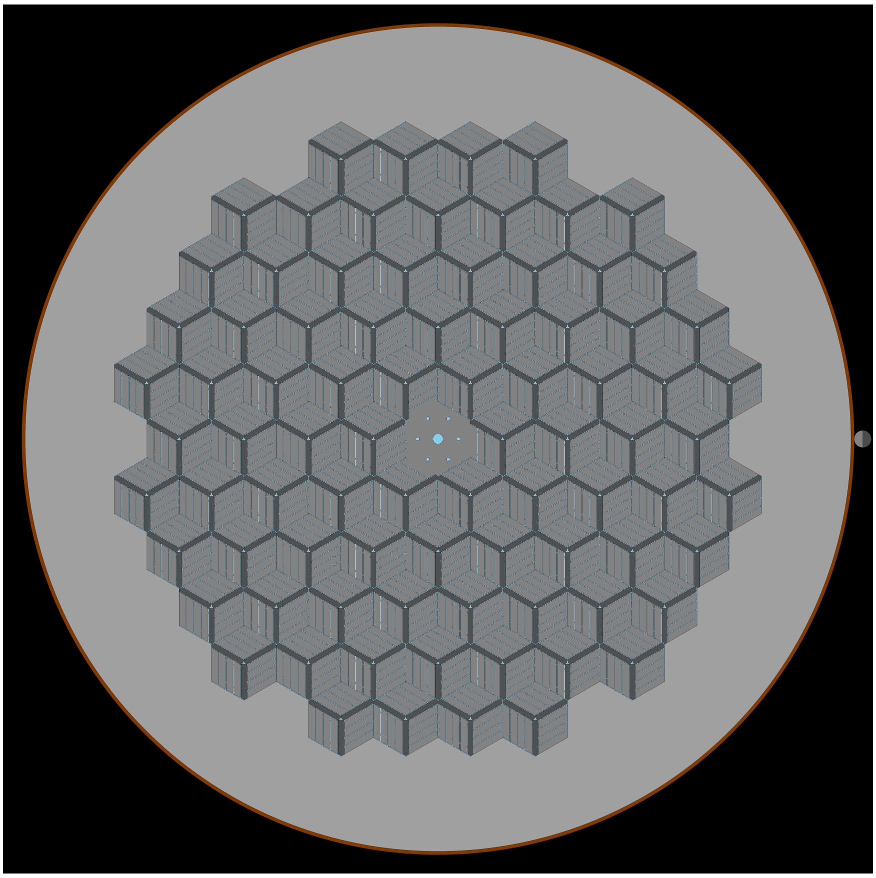

Figure 2: The thermal spectrum, LEU fueled rector core with an ex-core reactivity oscillator.

The flux of neutron extending to the annular region around the core represents 0.482 ± 0.002 % of the total. Following first order perturbation theory, a perfect absorber would need to cover only $\sim 0.4\,\%$ of the axial region to create the two pcm difference used in frequency analysis. It is possible for an oscillator could have a reflector on the other side of the absorber such as what is shown in Figure 2. This case is boron metal on one side and graphite reflector on the other side. This rod extends the height of the core (2 m) and has a radius of 4 cm (core radius of 2.5 m). The resulting difference the calculated implicit k value is $12.0 \pm 5.9\,pcm$, well above the 2 pcm need for frequency analysis.

4 Conclusions and Future Work

Given our preliminary results, it seems feasible that a thermal spectrum, LEU fueled MSR can have an ex-core reactivity oscillator. This will likely be design dependent and would have to be reevaluated for any specific reactor design. Completing an oscillator design with a specific reactor design while following the Safeguards-by-Design method would be ideal.

For future work, the model presented in this paper will be used to drive dynamic parameters of the reactor during plutonium diversion. Work on an accompanying dynamic model is underway, and this dynamic model will measure the full effects of plutonium diversion in such a reactor.

5 Acknowledgements

This work was supported by a Nuclear Energy University Programs grant sponsored by the U.S. Department of Energy, Office of Nuclear Energy, award number DE-NE0008793.

References

[1] Trond Bjornard, R Bean, S DeMuth, P Durst, M Ehinger, M Golay, D Hebditch, J Hockert, and J Morgan. Safeguards-by-design: Early integration of physical protection and safeguardability into design of nuclear facilities. Technical report, Idaho National Laboratory (INL), 2009.

[2] T Kerlin. *Frequency response testing in nuclear reactors*, volume 11. Elsevier, 2012.

[3] TW Kerlin, SJ Ball, RC Steffy, and MR Buckner. Experiences with dynamic testing methods at the molten-salt reactor experiment. *Nuclear Technology*, 10(2):103–117, 1971.

[4] Jaakko Leppänen. Serpent–a continuous-energy monte carlo reactor physics burnup calculation code. *VTT Technical Research Centre of Finland*, 4, 2013.

[5] Alexander M. Wheeler, Ondřej Chvála, and Steven Skutnik. Signatures of plutonium diversion in molten salt reactor dynamics. *Annals of Nuclear Energy*, 160:108370, 2021.