MCNP Neutronic Design Optimization of an Accelerator-Driven Subcritical Assembly for Molybdenum-99 Production

Andrew Conant[1], Noel Nelson[1], Jorge Navarro[1]
Chad Denbrock[2], Robert Whalen[2], Terry Grimm[2*]

[1]Oak Ridge National Laboratory
PO Box 2008, MS6172, Oak Ridge, TN 37831

[2]Niowave Inc.
1012 N. Walnut St., Lansing, MI 48906

conantaj@ornl.gov, nelsonnb@ornl.gov, navarroj@ornl.gov,
denbrock@niowaveinc.com, wahlen@niowaveinc.com, grimm@niowaveinc.com

ABSTRACT

Molybdenum-99 for medical applications has been in short supply in the recent decades. The most common production mechanism, irradiation in a reactor fueled by highly enriched uranium (HEU), raises proliferation concerns. Novel efforts are being pursued to meet the demand for ^{99}Mo without HEU irradiation, relying on the processing of low-enriched uranium (LEU) or natural uranium (NU). One concept is to use an electron linear accelerator to generate bremsstrahlung that, through a photonuclear reaction, can help multiply neutrons in a subcritical assembly. The goal of this work was to neutronically optimize the subcritical assembly for k_{eff} and ^{99}Mo production. This optimization is unique due to the mixed LEU and NU core combined with the processing of solely NU rods. The design was optimized in terms of reflector type, pitch, and LEU and NU mass. Results of the paper show the effects of fuel mass, pitch, and reflector material on k_{eff} and the fission fraction in the NU rods. k_{eff} was found to be mostly driven by LEU mass, and the NU fission fraction was dependent on several parameters, primarily pitch.

KEYWORDS: subcritical assembly, Molybedenum, neutronics, optimization, MCNP

1. INTRODUCTION

Technetium-99 is the daughter of medical isotope molybdenum-99 and is routinely used for medical diagnostics. Molybdenum-99 has traditionally been produced inside highly enriched uranium (HEU) reactors outside the United States. The National Nuclear Security Administration's Office of Material Management and Minimization (M^3) aims to support ^{99}Moproduction that does not include HEU as fuel. Many proposed designs use either a lower enrichment or molybdenum targets.

Niowave Inc. (Niowave) is designing a linear accelerator coupled with a subcritical uranium target assembly (UTA). Preliminary studies suggest that this design will likely be sufficient to produce enough ^{99}Mo to meet a significant portion of the US demand [1]. The UTA has three design iterations: UTA-1, UTA-2, and UTA-3. The design parameters of the three UTA stages are shown in Table 1. Calculations for the second iteration design [2] and shielding [3] of UTA-2 have been performed. The third design iteration, UTA-3, has the goal of producing 2 kCi of ^{99}Moper week at the end of bombardment, or about 5% of the US demand [4]. Oak Ridge National Laboratory is providing support for the design and shielding of UTA-3. The UTA-3 core optimization studies took a staged approach to isolate parameters of interest.

Table 1: Niowave UTA Design Phases. Note that some design parameters are updated from Ref. [2].

Characteristic	UTA-1	UTA-2	UTA-3
Electron Beam	10 MeV, 10 kW	20 MeV, 10.4 kW	40 MeV, 200 kW
Thermal Power (W)	2.3 W	230 W	330 kW
k_{eff}	0.43	0.63	0.95
LBE Neutron Source (n/s)	7.2×10^{11}	7.0×10^{12}	2.0×10^{15}
Mass of LEU (kgU)	1.6	4.5	33
LEU enrichment (%)	6.4	9.75	9.75
Mass of NU (kgU)	4.6	13.5	60.0
^{99}Mo Production (Ci/week)	0.1	10	2,000

2. MATERIALS AND METHODS

2.1. Subcritical Assembly Design

The accelerator-driven subcritical assembly (ADSA) core is relatively compact and contains low-enriched uranium (LEU) and natural uranium (NU) rods in a subcritical configuration. The inner lattice contains LEU rods, and outer rings of NU rods surround the LEU lattice. Radially outward of the fuel lattice is a reflector, although water may still be selected as a reflector. The UTA-3 core is based on the design of a previous iteration, UTA-2 [3]. The UTA-2 design aimed to have a $k_{eff} \leq 0.65$. The UTA-3 aims to have a $k_{eff} \approx 0.95$. An overview of the ADSA system and current parameters is shown in Figure 1. The UTA-3 neutron source will be bremsstrahlung photon irradiation from electrons incident upon a lead bismuth eutectic (LBE) target.

2.2. UTA-3 Optimization and Staged Approach

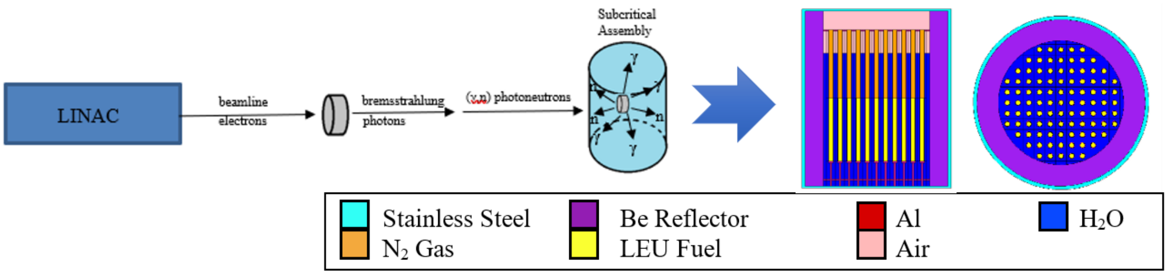
The neutronic design and optimization of UTA-3 is unique from traditional core optimization because of a combination of considerations from regulation, cost, and operation. As stated, UTA-3 differs from traditional methods of generating ^{99}Mo by utilizing LEU and NU instead of HEU. Additionally, Niowave currently plans to only process the ^{99}Mo from NU rods to simplify the regulatory pathway. The NU rods can be considered target material. The LEU fuel rods are unique in that they are high-assay low enriched uranium (HALEU) with 9.75% and both the LEU and NU rods have a relatively low density of 2.5 g/cm^3. All of these factors contribute to the requirement of a specialized approach to core design. The goal of the optimization is to analyze the effects LEU and NU fuel parameters on the k_{eff} and NU fission fraction.

The core optimization studies took a staged approach developed by Niowave. The purpose of the staged approach is to break the study down into a digestable subset of parameters that avoids a large number of parameters to explore all at once. A diagram of the stages is shown in Figure 2. This paper will present results from Stages 1–3. Note that the masses in some optimization stages are not the same as current parameters listed in Table 1 because of updates to Niowave's design.

2.3. Modeling and Simulation Method

UTA-3 is modeled with the MCNP code [5]. Prior work was done on optimization and shielding of the UTA-2 [2,3]. Its parameter study module PSTUDY [6] can be used to rapidly iterate over many parameters with parallel executions of input files. Table 2 lists the input parameters in the optimization study and metrics to optimize. Figure 3 shows the top and side view of one example configuration of the UTA-3 Stage 2 core. Material and geometry specifications were obtained from Niowave or from Ref. [7]. Note that statistical error on the MCNP simulations was around 50–100 pcm for k_{eff}, and fission tallies were $\leq 1\%$.

The optimization of k_{eff} is to be as close to 0.95 while still under that limit. The second optimization metric is the relationship of the fission rate in the NU to the total fission rate (i.e., f_{NU}/f_{total}). The primary difference



Accelerator Driven Neutron Source	
Electron Beam Power	200 kW = 40 MeV $\times$ 5 mA
Neutron Source Intensity	2.0×10^{15} n/s
Uranium Target Assembly-3	
k_{eff}	≤ 0.95
Fuel form	U_3O_8
LEU target mass	33 kgU, 9.75%
NU target mass	60 kgU
LEU fission power	280 kW
NU fission power	50 kW
^{99}Mo activity produced	13 kCi/week

Figure 1: UTA-3 design schematic (top) and system parameter overview (bottom). Note that the schematic shows LEU fuel, but the core will have LEU and NU rods.

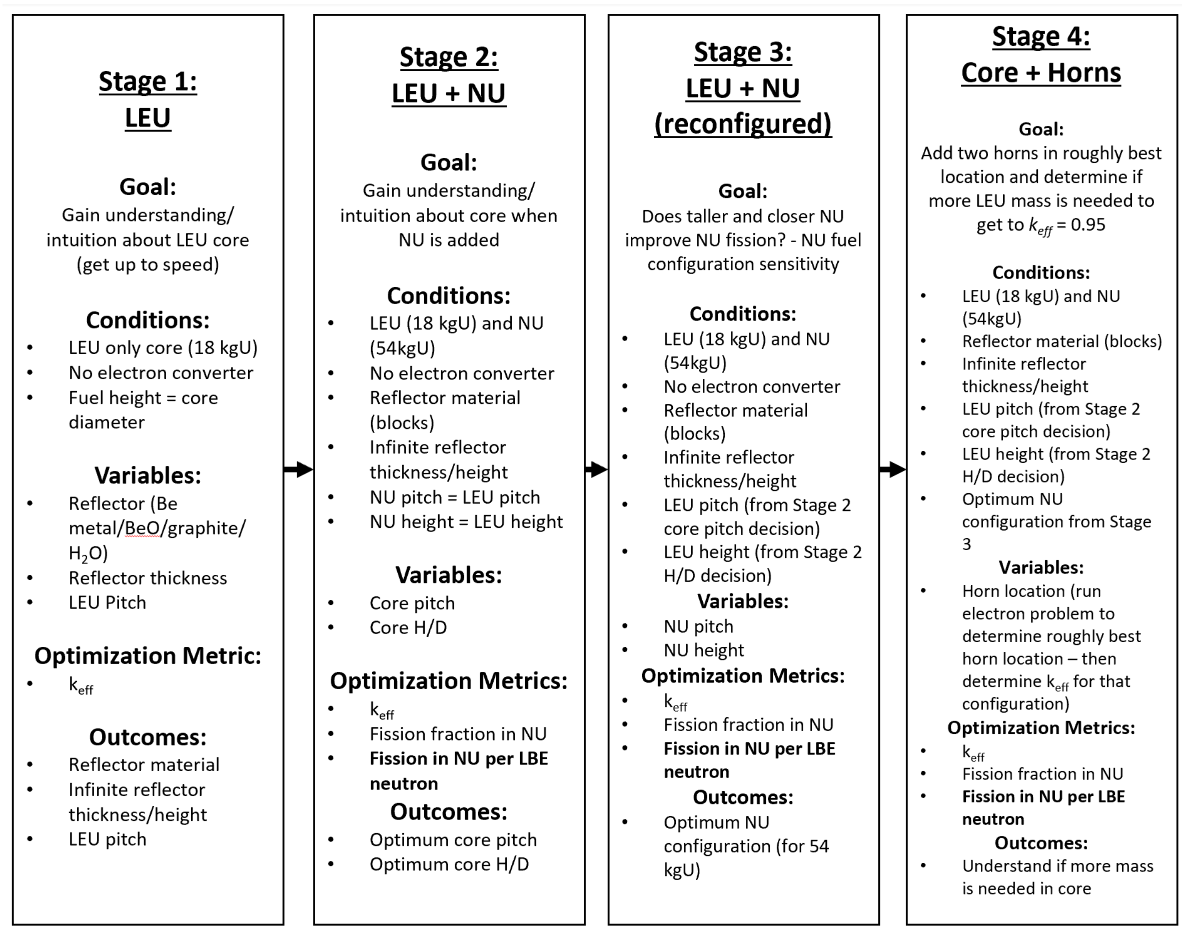


Figure 2: UTA-3 core optimization design stages.

Table 2: UTA-3 inputs and metrics for the optimization study.

Inputs	Metrics
Fuel* Mass	k_{eff}
Fuel* Pitch	NU fission fraction
Fuel* Height	
Fuel* Loading Pattern	
Core H/D	
Reflector Type	

*Fuel parameters for LEU and NU were different for some studies

between this study and the UTA-2 studies [3,2] is that the primary goal of the system is to maximize isotope production in the NU while maintaining $k_{eff} \leq 0.95$.

3. RESULTS

3.1. Stage 1

The Stage 1 optimization focuses on the core configuration with 18 kgU of LEU only and no LBE neutron source. Parameters to vary include the reflector material, reflector thickness, and LEU pitch. Figure 4 shows the effects of these parameters on k_{eff}. A commercial production facility requires a reflector that is cost effective and maintains neutronic and safety characteristics. The two beryllium reflectors—beryllium metal and beryllium oxide (BeO)—showed the strongest neutronic performance. k_{eff} was found to saturate around 25 cm for all reflector types, except for light water which was near 10 cm. The results for the lattice pitch show that the optimal pitch for the LEU configuration was 95% of the nominal UTA-3 pitch.

3.2. Stage 2

The Stage 2 optimization focuses on the addition of the NU rods into the lattice. To achieve the highest neutron multiplication factor, the LEU rods are located in the center, and the NU rods are located on the periphery. This stage includes 18 kgU of LEU and 54 kgU of NU. As part of Stage 2, simulations were performed to find combinations of LEU and NU mass given constant rod geometry that would satisfy $k_{eff} \geq 0.95$. Figure 5 shows the k_{eff}value that was calculated for each of these configurations. From the plot, it is evident that a minimum amount of LEU is required to achieve the target k_{eff} value and that the NU rods negligibly affect the multiplication factor. The points with $k_{eff} > 0.95$ are circled in red to indicate anticipated favorable configurations given the expected reactivity penalty that will occur once the neutron source is incorporated into the design.

3.3. Stage 3

The Stage 3 optimization focuses on the optimization of the added NU rods in the lattice. Stage 2 found that adding NU rods negligibly influenced the neutron multiplication of the inner LEU portion of the core. The selected value for Niowave's pitch was found to be near optimal for the LEU configuration, but an optimization is needed for a combination of LEU and NU rods. The current plan is to process the ^{99}Mo from the NU rods, so decreasing their pitch to make them closer to the main neutron-multiplying region of the core will maximize the ^{99}Moproduction in the NU. Additionally, a multi-parameter optimization study was also performed to check for cross-correlation effects between several design parameters.

3.3.1. NU Pitch

Separate MCNP models were developed to test a variety of different NU pitches for a given LEU pitch. Provided a fixed square lattice for the LEU, a separate value for the NU pitch requires a separate lattice.

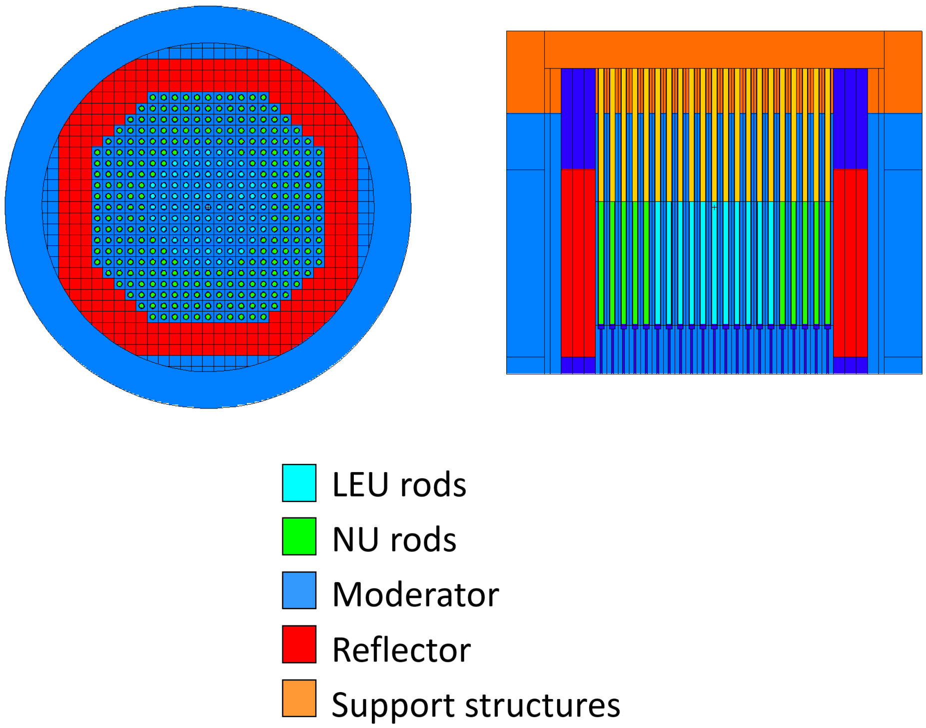


Figure 3: An XY slice (left) and a YZ slice (right) of the MCNP model of one iteration of the UTA-3 Stage 2 design.

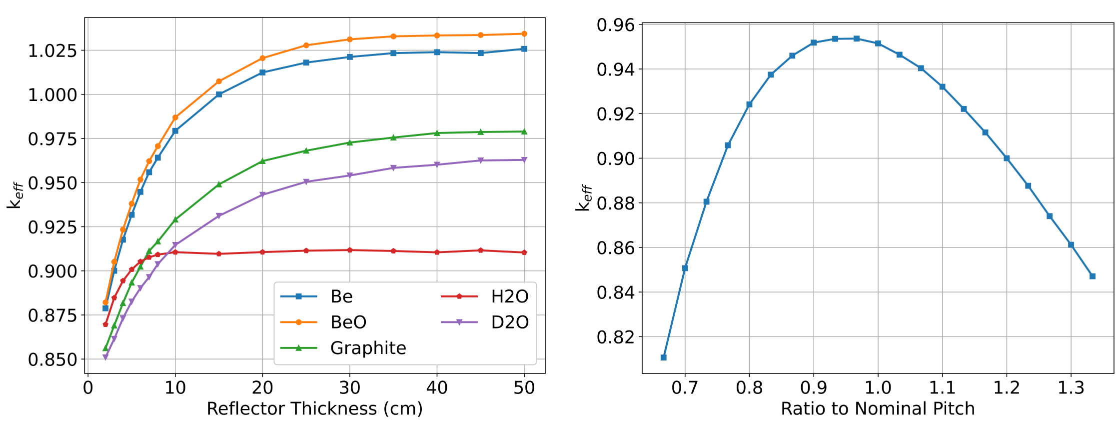


Figure 4: Stage 1 k_{eff} results for various reflector materials and thicknesses (left) and k_{eff} for different pitches of LEU-only cores (right).

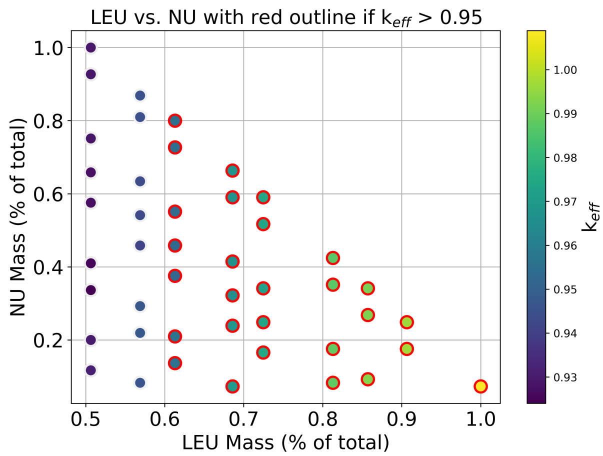


Figure 5: $k_{eff}$for various configurations of LEU and NU masses. The red outlines indicate configurations with $k_{eff} \geq$ 0.95

It is possible to consider geometries for the NU rods that are not a square lattice, but a square lattice was chosen for this study because of the simplicity of combining with the central LEU lattice. There will be some offset between the lattices, leaving a small gap between them, creating incongruous geometries. Although a circular core would be more efficient, some sensitivity studies showed that the shape of the NU lattice was negligible compared to the effect of the NU pitch value itself.

Figure 6 shows an example of a test case with separate LEU and NU pitches, as well as the results of k_{eff} and NU fission fraction for various NU pitch values. The gap between the LEU and NU checkerboards is visible, which we call an "offset." In other words, the offset is the distance between the LEU and NU lattices. The results for k_{eff} show a jagged pattern due to the differing offset values. The values of this offset were less than the LEU pitch, ranging from 0.0 to 2.2 cm. The sizes of the markers in Figure 6 are proportional to the offset value. For k_{eff}, the trend is nearly monotonously decreasing, with the exception of the larger offset values. The results show that a smaller value for the NU pitch is ideal to maximize k_{eff}. The NU fission fraction shows a behavior that is not as monotonically decreasing (despite offset size) as it is for k_{eff}. Decreasing the NU pitch can increase the NU fission fraction to as high as 22%. Although outside the scope of this paper, alternative fuel geometries or loading configurations could be analyzed to optimize k_{eff}.

3.4. Simultaneous Parameter Study

Niowave plans to only reprocess the ^{99}Mofrom the irradiated NU rods. Therefore, maximizing the fission fraction in the NU rods is ideal in addition to k_{eff}. At this stage, Niowave had updated its design to include 36 kg of LEU and 60 kg NU and plans to fix the mass for a UTA system. A parametric study was performed to simultaneously analyze multiple parameters from this stage and previous stages given the fixed masses. The list of parameters to vary for the fuel include: pitch, radius, height, and density. MCNP models were created for a spread of values for the these parameters. The variation in the fuel radius and height dictate a fixed rod volume and mass. The number of rods for the LEU and NU can be calculated as

$$N_{\text{rods}} = \frac{M_{\text{U,total}}}{w_f \rho \pi R^2 H},$$

where $M_{\text{U,total}}$ is the total mass of either the LEU or NU, w_f is the weight fraction of U in the fuel form (e.g., U_3O_8), ρ is the fuel density, R is the fuel radius, and H is the fuel height. For this study, the fuel height and radius for the LEU and NU are the same, but future studies can examine different values as was explored previously.

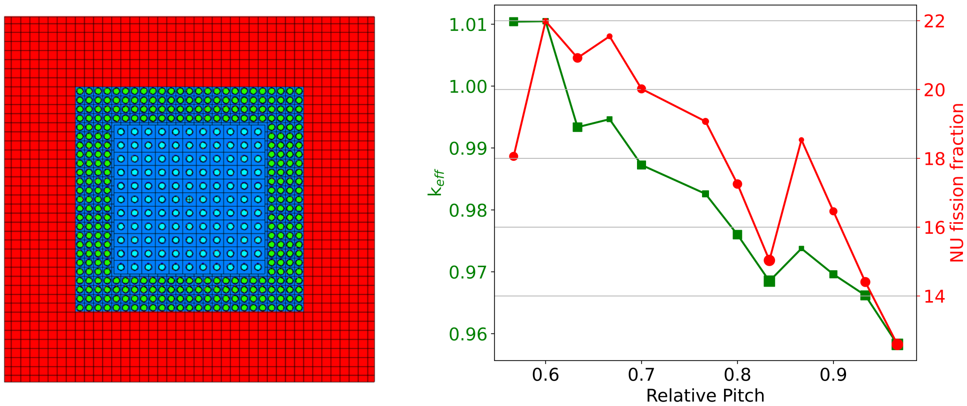


Figure 6: Example of a test case with different LEU and NU pitches (left) and results for k_{eff} and NU fission fraction for various NU pitches with a fixed LEU pitch (right). The sizes of the markers are proportional to the offset distance between the LEU and NU lattices.

Because N_{rods} will vary with different fuel geometries, a core loading mechanism was developed to fill the rods within the core. The LEU rods will be placed in the center, and the NU rods will placed in the periphery surrounding the LEU.

A core loading method is developed to incrementally fill the rods within the lattice to minimize the placement of each rod according to the Pythagorean theorem. The goal is to optimize for a circular core loading pattern by minimizing the distance D of each successively loaded pin from the central pin:

$$D_{\text{P}}(i,j) = \sqrt{x_i^2 + y_i^2},$$

where x_i is the x-position of the pin and y_i is the y-position of the pin. As an example, the first few cases will be discussed. Following the central pin at (0,0), the next pins loaded will be the four pins at (0,$\pm$1) and ($\pm$1,0) with a distance P from the central pin. The next four pins loaded would be at ($\pm$1,$\pm$1) at a distance of $P\sqrt{2}$. This pattern is repeated until N_{rods} for LEU is matched. Then, the pattern is repeated until the N_{rods} for NU is matched. Two different core loadings are shown in Figure 7.

Figure 8 shows the trade-off of k_{eff} and the NU fission fraction for 2,600 cases given fixed masses for the LEU and NU. Note that these cases do not incorporate the geometry of the neutron source, so their future incorporation in Stage 4 will decrease the k_{eff}. The figure shows a peak in k_{eff} around an NU fission fraction of 10%. After this peak, there is a nearly linear decreasing relationship between the two.

4. CONCLUSIONS

This work focused on the optimization of a nontraditional subcritical assembly for ^{99}Mo production. The UTA-3 assembly consists of LEU and NU fuel rods with 9.75% enrichment and NU fuel rods but both with a lower density as compared to traditional oxide fuels, which presents a unique optimization case. The optimization metrics included $k_{eff} \leq 0.95$ and maximization of the NU fission fraction becuase Niowave plans to only reprocess NU rods. A critical mass number was determined for the LEU mass required for $k_{eff} \approx 0.95$. Optimization of the NU proved more complex, but its pitch was a primary driver. As a target material, the NU served best when closest to the higher-multiplying LEU region of the core. Given fixed LEU and NU mass values, the NU and k_{eff} had an inverse relationship, and a k_{eff} = 0.95 equated to a maximum NU fission fraction of nearly 20%. Future studies aim to examine a larger parameter space of the NU fuel rod geometry, incorporate LBE neutron source into the model, and calculate ^{99}Mo production rates which plan to include depletion calculations.

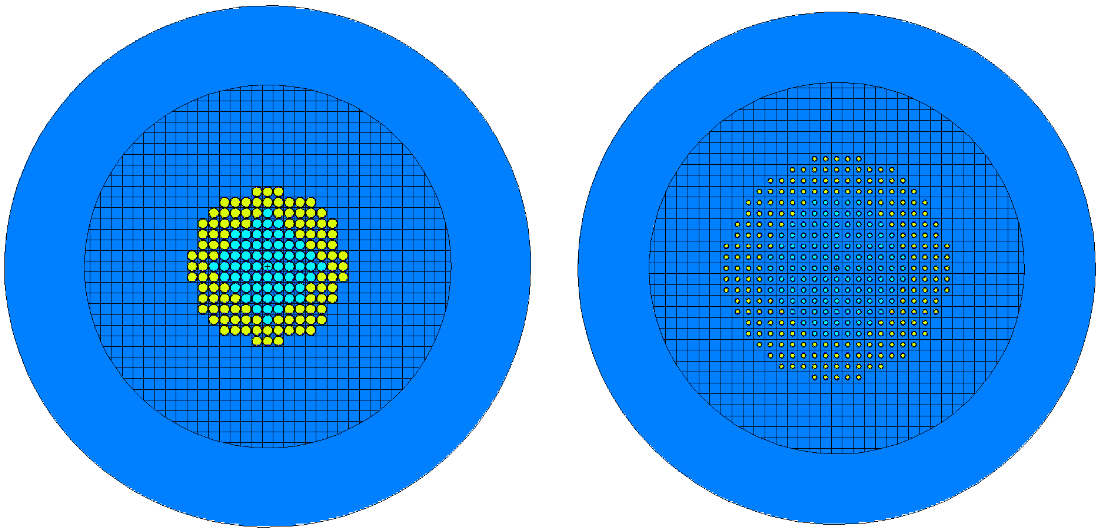


Figure 7: Example top-down view of two cases generated form the simultaneous parameter study, the first with a smaller height, larger radius, and smaller pitch (left) and the second with a larger height, smaller radius, and larger pitch.

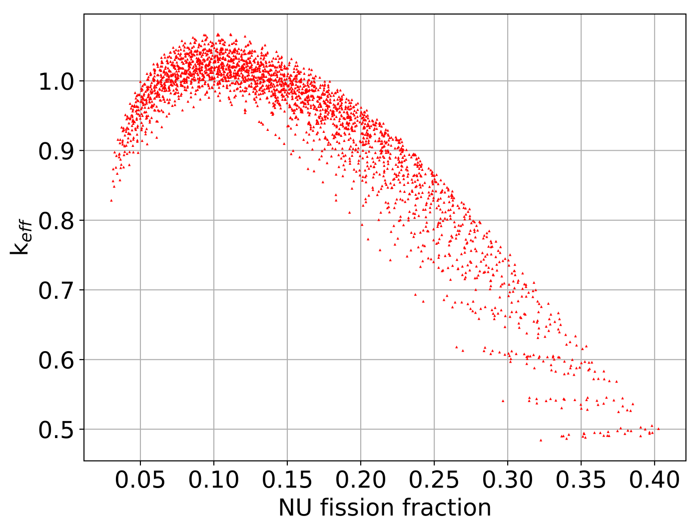


Figure 8: $k_{eff}$and the NU fission fraction for 2,600 MCNP cases varying fuel radius, height, pitch, and density

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