INTRODUCTION

For decades, benchmark quality measurements have been performed with a large variety of systems and types of nuclear material. The International Criticality Safety Benchmark Evaluation Project (ICSBEP) [1] is a repository containing a large number of critical and subcritical benchmarks that are used for nuclear data validation. However, there are very few benchmarks with neptunium, a rare element that is created through neutron reactions or decay processes involving special nuclear material. Even this subset of experiments are not very sensitive to neptunium cross sections [2, 3, 4, 5]. This impedes the ability of these benchmarks to validate nuclear data for neptunium isotopes.

If a subcritical benchmark can be made with a higher sensitivity to neptunium cross sections, then it will aid in the efforts to validate nuclear data. Additionally, such a measurement could aid in a better understanding of the critical mass of 237Np, the original intent behind the casting of a sphere made of the material. The experiment outlined here will be performed at the National Criticality Experiments Research Center (NCERC) at the Nevada National Security Site (NNSS), and build upon the success of previous Los Alamos subcritical benchmarks and benchmark quality measurements using the Hage-Cifarelli formalism of the Feynman Variance-to-Mean method [6, 7, 8, 9]. While the preliminary design of this experiment has been previously documented [10], this summary details the final design of the experiment, including the investigations into the issues that arose.

FINAL EXPERIMENT DESIGN

Neptunium Sphere

The Neptunium sphere (also called the “Np sphere”) was cast in 2001 to investigate the critical mass of 237Np [11]. The 6070.4 g sphere has a radius of 4.149 cm. Upon casting additional cladding and shielding was necessary to mitigate the large dose associated with a 311 keV gamma ray from 233Pa, a direct decay product of 237Np. This cladding and shielding took the form of 0.259 cm of tungsten and 0.386 cm of nickel. These layers lowered this dose rate from 2R/hr to 300mR/hr on contact.

Detailed isotopics were determined based on the sprue of the sphere (the top casting stem that was removed), but this determination may not be representative of other parts of the sphere, such as the interior or exterior surfaces. When the sphere was in a molten state, it is expected that the lower density impurities would float to the top of the sphere. Because of this, a composition based on the sprue can be expected to have more impurities relative to the rest of the sphere. The isotopics used for the design simulations, listed in Table I, are based on those listed in an ICSBEP benchmark that features the sphere [3]. Also listed is the expected spontaneous fission output based on the PANDA Manual [12].

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Mass (g)</th>
<th>S.F. yield (neutrons/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>237Np</td>
<td>6.06 x 10^3</td>
<td>6.90 x 10^-1</td>
</tr>
<tr>
<td>233U</td>
<td>2.17 x 10^-1</td>
<td>1.87 x 10^-4</td>
</tr>
<tr>
<td>234U</td>
<td>3.48 x 10^-2</td>
<td>1.75 x 10^-4</td>
</tr>
<tr>
<td>235U</td>
<td>1.66</td>
<td>4.96 x 10^-4</td>
</tr>
<tr>
<td>236U</td>
<td>9.28 x 10^-3</td>
<td>5.09 x 10^-5</td>
</tr>
<tr>
<td>238U</td>
<td>1.87 x 10^-1</td>
<td>2.54 x 10^-3</td>
</tr>
<tr>
<td>238Pu</td>
<td>9.83 x 10^-2</td>
<td>2.55 x 10^-2</td>
</tr>
<tr>
<td>239Pu</td>
<td>1.95</td>
<td>4.25 x 10^-2</td>
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<tr>
<td>240Pu</td>
<td>1.40 x 10^-1</td>
<td>1.43 x 10^-2</td>
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<tr>
<td>241Pu</td>
<td>3.77 x 10^-3</td>
<td>1.88 x 10^-4</td>
</tr>
<tr>
<td>242Pu</td>
<td>1.95 x 10^-2</td>
<td>3.35 x 10^-1</td>
</tr>
<tr>
<td>241Am</td>
<td>4.04 x 10^-4</td>
<td>4.76 x 10^-4</td>
</tr>
<tr>
<td>243Am</td>
<td>1.12 x 10^-1</td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>6.07 x 10^3</td>
<td>4.32 x 10^-2</td>
</tr>
</tbody>
</table>

Reflectors

Based on previously documented simulations with various reflector materials, the material chosen for the final design is Nickel 200, a high purity alloy of nickel. This material was chosen for both consistency with the preexisting cladding, and for the range of multiplication values that can be achieved when layers of the material are placed around the sphere.

The nickel reflection will be achieved by nesting the Np sphere inside spherical hemisheells of nickel, and the thickness will range from bare (i.e. no additional reflection) to approximately 9.14 cm (3.6 in) of nickel shells. The shells will be made to dimensions similar to those outlined...
for copper in a previous subcritical experiment [8]. This allows for easy flexibility in future experiments, as these shells could easily be interchanged or interleaved with others if an experiment calls for it. The largest thickness of reflection was chosen due to the diminishing returns of additional reflection, and due to the prohibitively heavy mass of any additional shells, as the largest hemishell already planned for this design is expected to weigh about 30 pounds.

**Detectors**

For these measurements, two Neutron Multiplicity Array Detector (NoMAD) systems will be used. These portable systems consist of 15 $^3$He tubes each embedded within high-density polyethylene, and have been used with a previous benchmark quality measurement involving a plutonium sphere reflected by copper [8]. The NoMAD records data in list mode format, meaning that each detection is time tagged, in this case to the nearest 100 nanoseconds. Such time tagging allows the data to be analyzed according to many different analysis techniques, including the Hage-Cifarelli formalism. Arranging two detectors, one on each side of the object, will provide a reasonable efficiency (~2% at 50 cm), and is again consistent with previous subcritical benchmark efforts. For these measurements, the detectors will be placed 30 cm from the center of the sphere to increase the efficiency even further.

**SIMULATED RESULTS**

Simulations were performed with MCNP® [16] using ENDF/B-VII.1 cross sections [14] in criticality eigenvalue mode in order to predict aspects of the assembled configurations such as $k_{eff}$, cross section and physical parameter sensitivities, leakage multiplication, and count rates. Each calculation used 5,000 active cycles of 10,000 neutrons each. Recent work has shown that the eigenvalue mode is able to estimate these parameters and their uncertainties, even though for some of them a fixed-source calculation may be traditionally more appropriate [15]. A plot of the predicted $k_{eff}$ as a function of nickel reflector thickness can be found in Figure 1, which shows that as expected, the multiplication factor rises with added reflector thickness. The total $^{237}$Np cross section sensitivity decreases with increasing reflector thickness, as the case with no additional reflection has a total sensitivity of 0.79, while 9.14 cm of additional reflection brings it down to 0.73. However, this range of expected sensitivities is still much higher than any existing benchmark.

Following the procedure listed in Reference 15, the leakage multiplication and singles and doubles rates can be inferred from $k_{eff}$. These rates, $R_1$ and $R_2$, represent the rate at which one or two neutrons from the same fission chain are detected, respectively. From Figure 2, it can be seen that the expected count rates from the system are quite low. This means that in order to get a small statistical uncertainty, the measurement time must be quite long. This is the primary motivation for moving the detectors from 50 to 30 cm, as the larger count rates will reduce the statistical uncertainties associated with a given counting time.

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PRELIMINARY MEASURED RESULTS

A series of preliminary measurements have been performed with the Np sphere in configurations similar to those in the final experiment design. These preliminary measurements serve to determine if the predictive simulation model can accurately represent the experiment. The most recent set of measurements, performed in June of 2018, set two NoMAD systems 50 centimeters from the center of the sphere, and collected data for 30 minutes. Table II shows the results of these preliminary measurements compared to the simulations. As can be seen, there is a large discrepancy between the simulated and measured count rates, while the leakage multiplications agree well.

TABLE II. The simulated and measured observables for two NoMADs measuring the bare Np sphere at 50 cm. The simulated uncertainty values are purely statistical.

<table>
<thead>
<tr>
<th>Case</th>
<th>Singles Rate</th>
<th>Doubles Rate</th>
<th>Leakage Multiplication</th>
</tr>
</thead>
<tbody>
<tr>
<td>Simulated</td>
<td>15.7±0.2</td>
<td>1.56±0.05</td>
<td>1.94±3E-4</td>
</tr>
<tr>
<td>Measured</td>
<td>169.5±0.3</td>
<td>16.8±0.2</td>
<td>1.95±0.02</td>
</tr>
</tbody>
</table>

Based on a detector efficiency estimated from the Monte Carlo calculations, a neutron emission rate from the sphere can be estimated. Based on the composition used for the simulation model, an emission rate of 785 neutrons per second is expected, while the experiment shows a rate of roughly 8,400 neutrons per second. This leads to the question of where these extra neutrons are coming from. Given that the leakage multiplication between the simulated and measured results matches well, it is not likely that the extra neutrons are due to greater than expected neutron multiplication in the sphere or in the surrounding environment. As an additional note, the temperature of the object not expected to rise much above that of the room, but if the benchmark measurements are performed a temperature reading will be taken during all configurations to ensure this is the case.

COMPOSITION INVESTIGATIONS

The extra neutron emission uncovered in the previous section could potentially be due to a number of different contaminants that were not revealed in the original isotopic analysis. These contaminants include more plutonium, lighter elements such as oxygen or lithium, or more exotic actinides like curium. Through various mechanisms, all of these have the potential to raise the neutron emission rate of the sphere.

If more plutonium was found to be in the sphere, that could both increase the neutron multiplication and the spontaneous fission rate inside the sphere, both of which would lead to an overall higher neutron emission rate. A Los Alamos report from 2002 estimates that there could be as much as 63 g of plutonium in the sphere [16]. This report then set the uncertainty bound in our sensitivity and uncertainty analysis for the plutonium content of the sphere. Simulations containing this much plutonium indicate that such a composition would lead to an expected count rate of around 450 counts per second, well above the experimental value of 169.5 counts per second. In adding this much plutonium, the ratios of the plutonium isotopes to each other was kept constant, and the other nuclides were reduced in a manner proportional to their preexisting mass fraction. This small amount of additional $^{239}$Pu was also not enough to raise the leakage multiplication significantly above the reference simulated and experimental values (1.96 vs 1.94 for the reference composition).

If there was a significant enough concentration of light elements such as oxygen or lithium, this would produce enough random neutrons through $(\alpha,n)$ reactions to cause the higher than expected emission rate without significantly affecting the multiplication of the system. However, this is likely not the cause. Given that the isotopic analysis was done from the top of the sphere, where any lighter impurities would have floated to, any such impurities should be overrepresented in the listed composition, not underrepresented or omitted. In addition, high precision gamma spectroscopy measurements have been performed on the sphere after the tungsten and nickel cladding. An example spectrum from these measurements is shown in Figure 3. Even though this shielding would block a significant amount of the gamma rays that would help with identifying impurities, there a number of high-energy emissions that would indicate the presence of $(\alpha,n)$ reactions. Despite the fact that such high-energy gammas would be more likely to penetrate through the shielding, no significant frequency of these events were found in the spectroscopy measurements. For these reasons, it is not likely that the extra neutrons are due to $(\alpha,n)$ emissions.
A third possible source of these extra neutrons that was investigated is a more exotic actinide such as $^{242}\text{Cm}$ or $^{244}\text{Cm}$. Given the quite high spontaneous fission rate of these isotopes, only micrograms of one would need to be present in the sphere to account for the unexpected emissions. Such small quantities may be difficult to detect in material assays, however.

CONCLUSIONS

The good agreement between the simulated and measured leakage multiplication values shows promise for a benchmark going forward. Additionally, the relative insensitivity of the leakage multiplication to the uncertainty in the plutonium content lessens the problem of not knowing the composition of the sphere well enough. However, this uncertainty in the actinide content of the sphere means that any benchmark will not be able to use the singles and doubles rates as a benchmark parameter. Despite best efforts to constrain the uncertainty in the plutonium content, it appears that determining precisely what is causing the extra neutron emissions would be quite difficult without removing a piece of the sphere itself. The experiment design as outlined in this document will provide for multiple configurations with distinct leakage multiplication values, and due to its large sensitivity to $^{237}\text{Np}$ cross sections, will be useful in validating nuclear data sets for this nuclide. The use of only the leakage multiplication as a benchmark parameter will be similar to critical experiment benchmarks, which may only use $k_{\text{eff}}$ as a benchmark parameter.

The reflectors have been ordered, with delivery expected to set execution of the experiment in fiscal year 2019. Once the experiment is complete, efforts will be made to complete a benchmark for submission to the International Criticality Safety Benchmark Evaluation Project.

ACKNOWLEDGMENTS

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REFERENCES