Measurements of Fission Gas Release from Nuclear Fuel in Support of BISON Fuel Performance Analysis

Nuclear Energy Advanced Modeling and Simulation

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Project Title: MEASUREMENTS OF FISSION GAS RELEASE FROM NUCLEAR FUEL IN SUPPORT OF BISON FUEL PERFORMANCE ANALYSIS

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Project Objective: This project established a fuel irradiation and fission gas release (FGR) loop that has been designed for implementation at the PULSTAR reactor of North Carolina State University (NCSU). The FGR loop is capable of analyzing various types of fuel samples including, e.g., uranium dioxide fuel (UO$_2$) and uranium silicide (U$_3$Si$_2$) samples. The fuel samples were produced and characterized at Idaho National Laboratory (INL) and shipped NCSU. The samples are ready for irradiation in the FGR loop to a given total thermal neutron fluence and under controlled temperature conditions. An innovative high resolution gamma-ray spectrometry approach and instrumentation have been established to monitor the loop’s sweep gas and perform high accuracy measurements of the gas release rates relative to predicted birth rates to establish the release-to-birth ratio (R/B) for a given radionuclide. In addition, uncertainty minimization is achieved through the formulation of the measurement observables into relative release-to-birth indicators for selected radionuclides. The experimental findings will be utilized to enhance and validate the fission gas release models that are currently implemented in the BISON fuel performance code. The experimentally deduced physical parameters will be incorporated in BISON’s fission gas release models. Atomistic simulations of fuel behavior have been performed to investigate the mechanisms of fission gas migration in the fuel. At this stage, performance of the fuel irradiation experiments awaits licensing approval by the US NRC.

Background: In the NEAMS program, BISON is a computer code that operates within the MOOSE framework. It is currently under development as the principal nuclear fuel performance prediction tool. It utilizes the finite element method and is
capable of analyzing 1-D spherical, 2-D axisymmetric and 3-D geometries under steady state and transient conditions. It applies to a variety of fuel forms including light water reactor (LWR) fuel, TRISO particle fuel, and metallic fuel in both rod and plate geometries. It is known that the prediction of macroscopic fuel behavior, e.g., fuel gaseous swelling and pellet cladding mechanical interaction (PCMI), is highly sensitive to understanding microscopic phenomena (e.g., fission gas diffusion). Consequently, a physics based model describing fission gas behavior and release has been included in BISON. However, this model is known to be inaccurate and is UO$_2$ specific (i.e., no U$_3$Si$_2$ data is included). Previous studies identified the uncertainties associated with the intra-granular fission gas diffusion coefficient as most limiting for the accuracy of fission gas release calculations. Also, the accurate characterization of microstructural properties such as the fuel’s grain radius and porosity was shown to be critical in terms of the proper modeling of FGR. While the predictions of BISON are considered useful, experimental exercises that extend the capability to other fuel types such as U$_3$Si$_2$ (an ATF candidate), and that are designed to access the underlying microscopic phenomena directly, accurately and under a wide range of irradiation and temperature conditions would be desirable for model validation and to enhance its predictive ability.

In particular, this project established an experimental facility at the PULSTAR reactor that is ideally suited for exploring the aspects of fuel behavior listed above and identified as priority in terms of improving fission gas release predictions, as well as being the source of great sensitivity for understanding the underlying physics of the phenomenon. The established FGR loop is capable of performing experiments that are designed to isolate (in a separate effects fashion) and correlate the various parameters (e.g., the fuel sample’s fission rate, temperature, and microstructure) with the measured gas release rates and the deduced gas diffusion coefficient. The measurements and findings of the FGR experiments will be used to test and validate the current release models of the BISON code.

Below is a conceptual diagram that illustrates the overall approach for FGR measurements and data analysis developed in this project:

Outline of the developed approach for in-pile measurements of UO$_2$ and U$_3$Si$_2$ fuel samples and the integration of the information in BISON.
1. Design and Implementation of the FGR Measurement Facility

1.1 Conceptual design overview of the facility

The FGR measurement facility should enable the irradiation of fuel samples under accurately controlled conditions, especially controlled temperatures and calibrated neutron flux. The distinct advantage of this concept over previous studies is that the temperature is controlled \textit{in-situ} and monitored within a compact high-temperature furnace inside the beam tube during fuel irradiation, so that the fission gas diffusion behavior can be measured under well-defined conditions. Small crystalline fuel particles are used to minimize any temperature gradient within the fuel. Another advantage of using single crystalline fuel particles is that the diffusion behavior of the fission gases is isotropic and not affected by grain boundaries, hence a simple diffusion model can be used to interpret the measurement results. In addition, the total sample mass is chosen to be small so that the heat generated from the fission process can be largely ignored.

A schematic of the conceptual design of the main structure of the FGR measurement facility is shown in Fig. 1 below. The fuel particles are represented by an array of spheres placed inside a furnace. The released fission gases will be carried by a stream of Helium sweep gas to the detector, where the concentration of radioisotopes is detected and analyzed. The fission gases then pass through a series of filters and a decay volume to meet safety requirements before being released to the environment. Since the travel time from the furnace chamber to the detector cannot be ignored, in order to accurately extract the release rate at the fuel, a well-controlled gas flow is also needed.

![Figure 1. A concept design of the FGR measurement open loop facility. The Helium gas is swept through the furnace where the fuel sample particles are heated. The flow rate of the Helium is controlled and monitored so that the decay of the fission products enroute to the detector can be determined.](image)

To take the conceptual design above one step further, several functional blocks need to be specified:

1. The temperature control and monitoring unit, which directly affects the accuracy of the system temperature and consequently the diffusion coefficient of the fission gases. A safety feature is also needed to guarantee that the reactor beam tube will not overheat and adversely affect the operation of the reactor.
2. The fission gas flow rate to the detector should be determined by accurately controlling the flow rate of the sweep gas. In addition, the sweep gas should be preheated so that it would not perturb the fuel temperature. The transportation of the fission gases needs to be controlled and...
efficient, and the decay of the fission gases during transportation needs to be accounted for. Leakage of the fission gases should be avoided.

3. The irradiation flux needs to be accurately measured. This can be done by a combination of modeling and actual measurement with standard samples. However, the \textit{in-situ} neutron flux cannot be measured directly because of the high temperature environment, so will be deduced by the activation of Argon gas flown through the sample position. A background calibration is also needed.

4. The decay of the fission gases needs to be detectable with minimized uncertainties.

A schematic of the system functional blocks is shown in Fig. 2. The gas flow loop is presented as blue blocks. The Helium sweep gas and the Argon calibration gas are connected to the same gas loop. At the returning side of the gas route, a delay branch can also selectively filter out the short-lived isotopes before reaching the detector, so that the long-lived isotopes can be measured with less background. The data acquisition system is marked in green. The pre-amplifier signals from the HPGe detector will be digitized and processed by a digital spectroscopy unit before sending to the main computer. A NaI detector (not shown in the block diagram) can also be implemented to monitor the real time count rate of the fission gases and confirm that a steady state is maintained during data acquisition.

![Figure 2. Functional blocks of the FGR facility control, monitoring, and data acquisition systems.](image)
1.2 Detectability Analysis

It is crucial to estimate the detectability of the series of fission gas isotopes that will be produced to provide guidance for the system design.

**Diffusion of the fission gases within the fuel material and the release rates**

The release rate ($R$) of the fission gases is one of the primary observables in the experiment utilizing this FGR measurement facility. In the measurement, the detectability of certain fission gas isotopes are determined by a number of limiting factors. For example, the diffusion coefficient inside the material is dramatically affected by the system temperature; the production rate of the isotope is determined by the neutron flux at the location of the fuel sample; As a result, the release rate is affected by both of the above factors, in addition to geometric and morphological aspects of the fuel samples. Note that without any actual measurement, we can only estimate the release rate or the detectability based on empirical formula. The release to birth ratio ($R/B$) of a certain radionuclide in a sphere can be calculated and solved under the steady state condition,

$$\frac{R}{B} = 3 \sqrt{\frac{D}{\lambda a^2}} \left[ \coth \left( a \frac{\lambda}{D} \right) - \frac{D}{\sqrt{\lambda a^2}} \right]$$

and the reduced diffusion coefficient, $D'$, is defined as $D/a^2$,

$$D' = \frac{D}{a^2} = \left( \frac{5000}{d} \right)^2 10^{-7.97-\left(1920/T\right)} \quad T < 1173K$$

$$D' = \frac{D}{a^2} = \left( \frac{5000}{d} \right)^2 10^{-2.60-\left(8220/T\right)} \quad T > 1173K$$

where $D$ is the diffusion constant, $a$ is the spherical radius, $\lambda$ is the decay rate, $d$ is the diameter in μm, and $T$ is the temperature in Kelvin. Fig. 3 shows the empirical relationship between the reduced diffusion coefficients, $D'$, as a function of the temperature, and it is clear that the diffusion constant is quite sensitive to temperature. Over a range of 1000K, the value of $D'$ could increase by more than three orders of magnitude.

![Figure 3. Empirical reduced diffusion coefficient $D'$ of fission gas as a function of sample temperature assuming a spherical sample geometry with a diameter of 100μm.](image)
Neutron flux in the beam tube #1

The neutron source term can be estimated using the data calculated from MCNP simulations. An MCNP model has been developed to study the neutron flux at the front end (facing the reactor core) of beam tube #1. The model included the full geometric details of the reactor core and beam tube #1, with a four-plate activation foil array placed inside the front end of the beam tube (see Fig. 4).

Figure 4. The MCNP model of beam tube #1 and the reactor core. Left) top view. Right) side view.

Figure 5 shows the MCNP simulation results of the thermal neutron flux inside beam tube #1. It is observed that at the very end of the beam tube adjacent to the reactor core, the neutron flux is significantly higher and slightly uneven spatially. At the end corner of the beam tube, the maximum thermal neutron flux is approximately $5 \times 10^{12}$ n/cm$^2$·s.

Figure 5. MCNP result of the neutron flux in beam tube #1.
The neutron flux distributions along the beam tube cross sectional area at 4 different locations are shown in Fig. 6. These places correspond to the location of the four thin plates shown in the model, and can be experimentally tested. It is shown that the neutron field becomes more evenly distributed at distances further from the reactor core. The neutron flux information is not only useful for providing guidance on the maximum allowed sample mass to meet safety requirements, but is also critical for evaluating the birth rate of the fission products in the samples. The fission gas release is originated by the production rate, and controlled by the diffusion behavior. In addition, the information regarding the level of neutron flux inside the beam tube is also useful to guide our design of the facility shielding. Here, the simulation is based on a simple geometry that has minimal shielding effects. However, the neutron flux needs to be further estimated based on the actual designed geometry and materials, and measured based on the final fabricated device. These two aspects will be discussed in later sections.

**Figure 6.** The thermal neutron flux distributed across the beam tube at different locations along the tube axis.

In order to examine the simulation results, a measurement has been carried out in beam tube #1. An array made of 4 Aluminum plates is inserted into the end of the beam tube, as shown in Fig. 7. On each plate, thin metal wires for activation analysis were placed at the center and on the vertical and horizontal axes close to the edge of the plates. The measurement results of the thermal neutrons along the center of the beam tube are compared to the MCNP simulations, as shown in Table 1.
Figure 7. Left) the measurement array made of 4 Aluminum plates holding the thin metal wires for mapping the neutron flux inside beam tube #1. Right) a picture showing the device inserted in the beam tube.

In the table, it is shown that the measured thermal flux values are roughly consistent with the MCNP calculated numbers. There are still noticeable differences among the two sets of results that cannot be ignored. The calculated drop of the thermal flux along the tube axis (from plate #1 to #4) appears to be larger than that of the measured values, which may be the result of scattering events that were not fully taken into account in the MCNP simulations, or slightly different neutron attenuation in MCNP relative to that of the actual materials.

<table>
<thead>
<tr>
<th>measured position</th>
<th>distance from the end (in)</th>
<th>center thermal flux (measured)</th>
<th>center thermal flux (MCNP)</th>
</tr>
</thead>
<tbody>
<tr>
<td>plate #1</td>
<td>1</td>
<td>$(2.3\pm0.1)\times10^{12}$</td>
<td>$(2.6\pm0.1)\times10^{12}$</td>
</tr>
<tr>
<td>plate #2</td>
<td>7.125</td>
<td>$(1.3\pm0.1)\times10^{12}$</td>
<td>$(1.3\pm0.1)\times10^{12}$</td>
</tr>
<tr>
<td>plate #3</td>
<td>13.125</td>
<td>$(6.4\pm0.3)\times10^{11}$</td>
<td>$(5.9\pm0.2)\times10^{11}$</td>
</tr>
<tr>
<td>plate #4</td>
<td>19.125</td>
<td>$(3.2\pm0.2)\times10^{11}$</td>
<td>$(2.7\pm0.1)\times10^{11}$</td>
</tr>
</tbody>
</table>

Birth rate of certain isotopes and safety limits

The birth rate ($B$) of the fission products is another major factor that determines the release rate, but it is not measured directly. The quantity $B$ is calculated using the cumulative fission yield of certain nuclides and the total fission rate of the sample, which are directly determined by the sample mass, the neutron flux, and the cross sections. For maximum detectability, one can simply increase the sample mass and/or the neutron flux. However, both of these factors have certain limits. The maximum neutron flux that the sample can receive is fixed by the reactor configuration and its operational power level. Analysis using MCNP6 code showed that at the end of beam tube #1 of the PULSTAR reactor, the maximum neutron flux that can be achieved is close to $5\times10^{12}$ n/cm$^2$·s. Meanwhile, safety requirements also impose an upper mass limit on the fuel samples being irradiated. For example, Figure shows the allowed mass of U235 and U238 under assumptions of different neutron flux and irradiation time. It is obvious that U235 is the major limiting factor in the mass limit, at least for short irradiation times. Only for irradiation times beyond $\sim1\times10^6$ s (or $\sim280$ hrs) does the mass of U238 become more important.
Figure 8. Mass limits of certain fuel samples based on different irradiation time and neutron flux

The above figure could be somewhat misleading considering the fact that only low-enrichment samples will be used in our experiments. Under such conditions, the majority of the sample mass is composed of U238. If we assume a certain sample enrichment, the total allowed sample mass can be calculated based on Fig. 8 as a function of irradiation time. Fig. 9 shows the total allowed mass of Uranium Dioxide (UO$_2$) assuming 1% or 10% sample enrichment under different neutron fluxes as a function of irradiation time. Notice that the maximum allowed mass is ~100g with a very short irradiation time (10s), except at the highest flux of $1\times10^{13}$ n/cm$^2$·s.

Figure 9. Mass limits of UO$_2$ assuming 1% or 10% enrichment at various neutron fluxes and irradiation times.
On the one hand, it is preferred that the measurement is taken under steady state conditions when the fission gas isotopes of interest reach or at least approach their saturated concentrations, which requires that the half-life of the particular isotope is short enough. However, the half-life must be long enough so that a reasonable fraction of the nuclide survives transportation from the target chamber to the detector. Table 2 shows a list of select fission gas nuclides that have reasonable half-lives for our measurement. In the table, the time for a specific isotope to approach a certain range of saturated concentration is also given. This time can be used as a guideline for determining the total irradiation time when a steady state measurement is needed. Combining this information with the mass limits given in Fig. 9, the total allowed sample mass can be estimated based on which fission gas needs to be measured under steady state conditions.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life</th>
<th>Time to 97%-99% saturation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xe-135</td>
<td>9.14±0.02 h</td>
<td>46~61 h</td>
</tr>
<tr>
<td>Xe-135m</td>
<td>15.29±0.05 min</td>
<td>1.3~1.7 h</td>
</tr>
<tr>
<td>Xe-137</td>
<td>3.818±0.013 min</td>
<td>19~25 min</td>
</tr>
<tr>
<td>Xe-138</td>
<td>14.08±0.08 min</td>
<td>1.2~1.6 h</td>
</tr>
<tr>
<td>Xe-139</td>
<td>39.68±0.14 s</td>
<td>3.4~4.4 min</td>
</tr>
<tr>
<td>Kr-85m</td>
<td>4.480±0.008 h</td>
<td>23~30 h</td>
</tr>
<tr>
<td>Kr-87</td>
<td>76.3±0.5 min</td>
<td>6.4~8.4 h</td>
</tr>
<tr>
<td>Kr-88</td>
<td>2.84±0.03 h</td>
<td>14~19 h</td>
</tr>
<tr>
<td>Kr-89</td>
<td>3.15±0.04 min</td>
<td>16~21 min</td>
</tr>
<tr>
<td>Kr-90</td>
<td>32.32±0.09 s</td>
<td>2.7~3.6 min</td>
</tr>
</tbody>
</table>

The release rate of certain fission gases from the fuel sample can be estimated assuming steady state conditions. This is true for most of the isotopes listed in Table 2 that only require several hours to saturate, which may be achieved during the typical PULSTAR reactor operations schedule. To be more conservative, we could take ~60 hours to be the minimum run time required before conducting the measurement of Xe-135 under assumed steady state conditions. In this case, the total run time is in the range of ~2×10^5 s in Fig. 9, which allows >1 g of 1% enriched UO2 sample. Given that the neutron flux at the sample is < 5×10^{12} n/cm²·s, the total calculated mass limit for U235 is found to be ~1.65 mg. The actual neutron flux on sample should be somewhat less due to attenuation caused by the sample chamber, and needs to be further calculated and measured. This will be discussed in subsequent sections. Based on the above discussion and the discussion in the previous section, the total release rate of certain nuclides from the sample can be estimated as a function of sample temperature, as shown in Fig. 10.
The detected activity of certain isotopes

The detected fission gas activity at the end of the loop is closely related to the detection efficiency and the transport efficiency of the fission gas isotopes. Assuming steady state conditions with a constant gas flow rate, the detection efficiency is dependent on the tube and can be calibrated experimentally. In order to maximize the detection efficiency, the gas tube that carries the fission products is formed into a helical coil surrounding an HPGe detector end cap so that a significant fraction of the detector solid angle is covered. The absolute detection efficiency can be estimated by comparing the loop geometry to a single point source geometry. At the same time, coincident events that could distort the final energy spectrum also need to be considered. Simulations can also be done to provide further guidance on the experimental characteristics of such a system. Details of this helical loop design will be discussed in later sections of this report.

Based on the estimations discussed in the sections above, the detected count rates of select photo peaks for certain fission products are shown in Figure 11 below. These results assume steady state conditions, and are based on a number of estimated parameters including the total sample mass, particle size, and detection and transport efficiencies, therefore the values can only be taken as rough approximations. The ambient radiation background at the detector is heavily affected by the configuration of the system shielding components which will be investigated in later sections. Nonetheless, the detected rates for many of the peaks are quite good even based on conservative
assumptions. One important feature we observe in the figure is that the expected count rates vary as a function of the gas flow rate, and for certain isotopes, there is an optimum flow rate for their detection. This is understandable because a faster flow would improve the transport efficiency but would also lower the dwell time of the fission products around the detector hence lowering the overall detection efficiency. A tradeoff between these two factors determines the best detection rate, and for different isotopic half-lives, the optimum flow rate is different. This can be used to our benefit by selecting different flow rates for certain isotopes in order to maximize the subject count rate and minimize the background rate from other isotopes at the same time. It is also clear from Figure 11 that for the long half-life gases (e.g. Kr-88, Kr-87, and Xe-135) it is better to use the lowest flow rate so that the other gases have already decayed away upon arriving the detector. This type of measurement may require the utilization of a delay volume in the gas flow loop (see Figure 2 above).

![Figure 11. Estimated count rate of select Xe and Kr decay events in the HPGe detector](image)

1.3 Computational Simulations of Fission Gas Transport

The detectability calculations discussed in the previous sections are based on some simple assumptions. Among these, the steady state condition can only be achieved after tens of hours of continuous operation. The flow of the sweep gas and the fission products it carries has been estimated based on ideal conditions and geometries. The gas diffusion and decay within the fuel particles as well as in the sweep gas need to be further evaluated under more realistic conditions. Meanwhile, other systematic effects need to be considered, such as the non-spherical geometry and the size distribution of the sample particles; temperature variation, temperature shifts, and/or temperature deviation of the samples; and heat transfer within the furnace, the sweep gas, as well as between the furnace and the gas, etc. Some of these aspects require a further understanding of the fluid thermodynamics of the system, and also help us to evaluate the final statistical and systematic uncertainties, which also help to guide the mechanical design and to set the design parameters of the actual system.
Fission gas transport within the fuel particles

As mentioned earlier, the microscopic transportation behavior of the fission products within the fuel is controlled by many factors including - but not limited to - the system temperature, diffusion constant, as well as the fuel morphology. Under ideal conditions, simple models can be set up and analytical solutions can be found straightforwardly based on simple diffusion equations. At the same time, numerical calculations could also be useful in evaluating the diffusion behavior of the fission gases when deviation from ideal conditions occurs, such as the existence of temperature gradients within the fuel, non-ideal fuel particle geometries, time dependent change from the steady state, grain boundaries, etc. COMSOL Multiphysics simulations can be used in combination with MCNP simulations to mimick the fission processes taking place within the fuel, and with MATLAB to consider time dependent diffusion behaviors. More importantly, the numerical simulations are also used as a bridge to link the conceptual design to the mechanical design. The designed system can be virtually tested as a proof of concept when multiple aspects of physical properties of the system interplay – microscopically, thermally, and mechanically.

As an example, the diffusion of Xe-138 in a spherical fuel particle has been examined in COMSOL Multiphysics. The gas concentration across the microsphere can be simulated and the result agrees with the Booth model under steady-state conditions, as shown in Figure. The same release rate can also be computed under such conditions. This is expected since the numerical method used here is based on the same set of diffusion equations.

Investigating the more complex diffusion behaviors that cannot be easily (or possibly) solved analytically would be the main benefit of using such numerical methodology. For example, the Booth model assumes a perfect spherical geometry for the fuel particles, and this needs to be generalized to various shapes to evaluate the geometric effect on the diffusion and release rates. From Figure 12, it is observed that the gas concentration abruptly drops near the surface while staying quite constant deeper within the sphere. This large concentration gradient at the surface is mainly due to the small diffusion constant. In general, only the gas atoms approaching the surface can reach the boundary and be released. Consequently, it is understandable that the dominating factor of the release rate is the total surface area for a certain total amount of fuel. However, unlike the total sample mass, this is not clearly defined under the safety limit mentioned previously.

![Figure 12. COMSOL Multiphysics calculation result of fission gas concentration within a spherical fuel particle.](image-url)
Figure 13 shows three simple cases of particle shapes (cylinder, cube, and sphere), with each case set to have the same surface area. It is found that there are still noticeable differences of their release rates in the range of ~5% due to their volume/mass differences. With the same surface area, the spherical particle has the largest volume, while the cubic particle has the smallest. Samples with the same mass and surface area could still have differences in the release rate purely due to geometric effects, even though it might be only a small percentage. Therefore, it is useful to assess the sample geometric properties including both the mass and the surface area characteristics, as well as the particle size distribution. Consequently, given the total sample mass limit, a higher surface-to-volume ratio particle would produce a higher release rate, and with the same average surface-to-volume ratio, any sample with a narrower particle size distribution is preferred over a broader size distribution to minimize the systematic uncertainty.

**Figure 13.** Release rate of sample particles and its dependence on particle shape. The particles have the same surface area and the diameter of the sphere is 100 μm. The numbers on top of the particles represent their release rates under neutron flux of $5 \times 10^{12}$ /cm²s at 1273K.

**Fission gas transport in the sweep gas**

The transport of the fission gases to the detector after their release is another major aspect that we need to examine using multiphysics modeling. Fig. 14 shows a schematic diagram of the numerical processes in COMSOL Multiphysics that have been considered in simulating the fission gas transportation. Several major physical effects are considered and shown in the chart including the diffusion and the heat transfer in both solid (fuel) and the sweep gas, as well as the fluid/gas flow process. The combination of the various physical properties of the system is dominated by either isothermal or non-isothermal flow equations, with the underlying goal of better understanding and optimizing the transport process and its efficiency, respectively. For example, the birth rate calculated from MCNP is fed into COMSOL as a starting parameter. However, the heat transfer process, including the furnace heating and the sample self-heating, is affected by the flow of the sweep gas and its temperature, which in turn affects the temperature field and the diffusion constant. In addition, the flow of the sweep gas is also affected by the temperature because of the change in its pressure and density fields. The assumption of laminar flow is only valid for small Reynolds numbers, but the existence of the sample particles and the hot zone may perturb the velocity field of the gas and cause the flow to become turbulent.
In order to investigate the temperature field in the furnace, especially in the vicinity of the fuel particles, a simple cylindrical furnace is set up with a Helium gas inlet and outlet at the center of each end surface respectively. A small array of 5×5×5 spherical fuel particles is positioned in the furnace. Helium gas is flowed through the furnace at a constant speed, and the furnace is heated to a constant temperature. The self-heating of the fuel particles is quite minimal comparing to the external heating due to the furnace. The temperature variations can be found among the particles within each layer and also among different layers. The particles facing the inlet have the lowest temperature, which is expected due to the cooling effect of the incoming Helium gas. Overall, the temperature variation is not dramatic given that the Helium flow speed is low and the system temperature is also low. We also note that the temperature variation is much more significant between the different layers than within each layer, and this suggests that it may be beneficial to design the sampling holding device to hold one evenly distributed particle layer instead of a pack of particles in a volume.

As a further step, another model is constructed where a single layer of sample particles are placed in the center of a U-shaped tube, which is acting as a sample holder as well as the gas sweeping tube, as shown in Fig. 15. The diameter of the tube is set to be 3 mm and the diameter of the spheres is 100 μm. The Helium gas flows in from one end of the tube, passes through the sample layer, and flows out of the other end of the tube (indicated by the blue arrows in Figure). A benefit of this design is that the speed of the Helium gas would be roughly consistent throughout the system without any obvious delay (as compared to a sample section with a much bigger diameter), hence shortening the dwell time of the short-lived isotopes.
Figure 15. The temperature field of a simple furnace with Helium gas inlet and outlet attached to the two ends of the cylinder. The temperature variation among the fuel particles arranged in a 5×5×5 array is shown. The temperature variation along the center lines of three select layers is also shown in the lower-right plot.

The major portion of the tube is placed inside the furnace hot zone so that the temperature of the tube boundary is set to a constant value. The heat transfers by conduction from the tube boundary to the Helium gas inside and also by convection to the fuel particles. Radiation heat transfer is also taken into account in this model. The fission heat produced by the fuel is also considered as an additional heat source. The two ends of the tube (the dark grey section in Fig. 16) are not enclosed by the furnace and are exposed to the environment and also radiate heat to the atmosphere (indicated by the red arrows in Figure 16). The boundary conditions of this section are defined by a diffuse surface in COMSOL, following the quadruple law of temperature differences,

\[-\mathbf{n} \cdot \mathbf{q} = \varepsilon \sigma (T_0^4 - T^4)\]

where the \(-\mathbf{n}\) is the unit vector of the outer surface, \(\mathbf{q}\) is the heat flux, and \(T\) is the system temperature field, \(T_0\) is the ambient temperature, \(\varepsilon\) is the surface emissivity, and \(\sigma\) is the Stefan-Boltzmann Constant.
The heating power generated by the fuel particles can be estimated by the product of the total fission rate of the samples and the energy produced per fission (~200 MeV). The total fission rate depends on the estimate of the neutron flux which would need to be calibrated eventually. This heating power is taken as a source term in the calculation of the heat transfer within the fuel particles. In our system, the heat generated by the furnace dominates the temperature field of the Helium gas and the fuel particles. The general heat transfer equations used in the COMSOL simulations is,

$$\rho C_p u \cdot \nabla T = \nabla \cdot (k \nabla T) + Q_p + Q_{vd} + Q$$

where $\rho$ is the density of the heat transmitting media, $C_p$ is the heat capacity of the media, $u$ is the velocity field, $k$ is the thermal conductivity, and $T$ is the temperature field. The source term of the heat is $Q$. The $Q_p$ and $Q_{vd}$ represent the work done by the media (e.g. gas expansion) and by viscous dissipation, respectively.

The flow of the Helium sweep gas inside the U-shaped tube is characterized as the flow of a uniform and compressible media, and can be simplified to a laminar flow at low flow rate. At the same time, Helium can be well defined as an ideal gas. The physical behavior of such flow can be estimated by the Navier-Stokes equations, and combined with the heat transfer equations, a variety
of the gas properties, such as the Helium temperature, pressure, density, velocity, etc., can be computed under a steady-flow state. As an example, Fig. 17 shows the temperature field within the U-shaped tube with the external heater and the fuel particles as the heat sources, and with Helium gas flowing through the system at a constant speed. The simulation result shows the temperature difference among fuel particles, and also shows the residual heat from the gas after exiting the hot zone at the lower-right portion of the tube. The temperature variation among the fuel particles is not dramatic, but will contribute to the systematic uncertainty if not accounted for.

Figure 18, shows the relative velocity field of the Helium sweep gas when the inlet flow speed is set to 0.1 m/s. It is clearly shown that the gas velocity increases significantly once entering the hot zone of the furnace, which is the result of the gas expansion and density decrease. The density profile of the Helium gas is shown in Figure 19. As shown in the figure, the density of the Helium decreased dramatically due to its high temperature and hence the expansion.

![Figure 18. The relative velocity field of the Helium gas with the inlet flow speed of 0.1 m/s.](image)

![Figure 19. The density profile of the Helium gas inside the U-shaped tube](image)

It is worth to note that in Fig. 19, the velocity of the sweep gas is also affected by the inner surface of the tube due to surface friction, as well as the fuel particles. When the gas approaches the fuel particles, the velocity drops significantly at certain locations due to the blocking effect, but also increases in between the fuel particles. This would render the overall flow of the sweep gas, and hence the fission products it carries, as a more complicated vector field that deviates from the simple constant velocity estimates. It may also impose certain limits on the overall flow rate that the system can obtain.
The corrected gas transport efficiencies

The overall fission gas transport efficiency can be calculated simply by assuming a constant time for the gas to arrive at the detector from the fuel particles. As mentioned earlier, this calculation can be further corrected by taking into account the non-isothermal flow of the sweep gas in the vicinity of the hot zone, where the flow velocity field is disturbed by the changes of the gas temperature and density. The surface friction and the geometric blocking effect of the fuel may also play some role in further correcting the transport efficiencies. The sample temperature has a noticeable effect on the transport efficiency since the gas flow speed inside the hot zone is dramatically different from the set flow rate at the inlet/outlet of the system. For example, Fig. 20 shows the transport efficiency of a select nuclide as a function of the furnace temperature. Because of this effect, the length of the tube inside the hot zone is preferred to be as short as possible in our design, so that the temperature-dependence is minimized. Another aspect of the issue is related to the diffusion of the fission products within the sweep gas, and this is also taken into consideration in the COMSOL simulations.

![Graph showing the transport efficiency of the fission gases due to the hot zone temperature.](image)

**Figure 20.** Effect on the transport efficiency of the fission gases due to the hot zone temperature.

The corrected transport efficiencies of the fission gases calculated using COMSOL are listed in Table 3. Here the hot zone temperature is set to 1073K. One simplification made here in the simulations is that the diffusion equation within the fuel was not solved coupled with the rest of the temperature and gas flow field to save computational time. This is reasonable given that the temperature variations among the fuel particles is less than one Kelvin after the fuel geometry optimization. As a result, the fission gas concentration throughout the fuel particle as shown in Figure agrees with the theoretical calculations almost perfectly. Another simplification is made in the region after the gases exit the hot zone. This is a long tube of >100” and the temperature has been assumed to be constant and similar to RT, which is necessary for a reasonable computational time. The isothermal flow in this region can be further assured by implementing a heat exchanger at the hot zone exit, so that the gas is fully cooled to RT and maintains a constant temperature when traveling to the detector.
Table 3. Fission gas transport efficiencies calculated by COMSOL Multiphysics at different flow velocities.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-Life (s)</th>
<th>0.001 (m/s)</th>
<th>0.01 (m/s)</th>
<th>0.1 (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}$Xe</td>
<td>229.08</td>
<td>0.07</td>
<td>44.06</td>
<td>92.41</td>
</tr>
<tr>
<td>$^{138}$Xe</td>
<td>844.80</td>
<td>11.50</td>
<td>81.03</td>
<td>98.36</td>
</tr>
<tr>
<td>$^{139}$Xe</td>
<td>39.68</td>
<td>0.00</td>
<td>0.86</td>
<td>62.23</td>
</tr>
<tr>
<td>$^{87}$Kr</td>
<td>4571.91</td>
<td>66.97</td>
<td>97.42</td>
<td>100.13</td>
</tr>
<tr>
<td>$^{88}$Kr</td>
<td>10224.01</td>
<td>84.40</td>
<td>99.84</td>
<td>100.51</td>
</tr>
<tr>
<td>$^{89}$Kr</td>
<td>189.00</td>
<td>0.02</td>
<td>36.99</td>
<td>90.86</td>
</tr>
</tbody>
</table>

For comparison, the efficiencies assuming a constant flow velocity are listed in Table 4. The results larger than 100% in Table 3 are due to the rounding of numbers in the numerical calculations. The two tables show some noticeable differences, which are believed to be due to the effects discussed above. Note that the simulation has been carried out based on a geometry optimized for having minimal effects due to non-isothermal flow.

Table 4. Fission gas transport efficiencies calculated by assuming simple constant velocities

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-Life (s)</th>
<th>0.001 (m/s)</th>
<th>0.01 (m/s)</th>
<th>0.1 (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}$Xe</td>
<td>229.08</td>
<td>0.04</td>
<td>45.66</td>
<td>92.46</td>
</tr>
<tr>
<td>$^{138}$Xe</td>
<td>844.80</td>
<td>11.93</td>
<td>80.85</td>
<td>97.90</td>
</tr>
<tr>
<td>$^{139}$Xe</td>
<td>39.68</td>
<td>0.00</td>
<td>1.08</td>
<td>63.60</td>
</tr>
<tr>
<td>$^{87}$Kr</td>
<td>4571.91</td>
<td>67.52</td>
<td>96.15</td>
<td>99.61</td>
</tr>
<tr>
<td>$^{88}$Kr</td>
<td>10224.01</td>
<td>83.89</td>
<td>98.26</td>
<td>99.82</td>
</tr>
<tr>
<td>$^{89}$Kr</td>
<td>189.00</td>
<td>0.02</td>
<td>38.67</td>
<td>90.94</td>
</tr>
</tbody>
</table>

1.4 Computations of the detected spectrum

The fission products of interest in our experiments are neutron rich in general and will therefore undergo beta decay and eventually emit photons after de-excitation. From the decay schemes, true coincident and random coincident events will occur and need to be reduced (or corrected) in the final instrumental design. One simple way of reducing such coincident events is to increase the source distance to the detector, however at the expense of significantly lowering the counting rate. A good figure of merit for the best detection of a certain peak can be taken as the peak-to-Compton ratio, which is defined as the ratio of the number of counts in the characteristic photopeak of the highest energy to the average number of counts of the Compton continuum approximately 300 keV to the left of the photopeak. This method can be used to determine the detectability of any isotope of interest. As a simple rule, a specific photopeak has to contain enough counts, $N_D$, to be considered detectable over the background given the Currie equation,

$$ N_D = 4.653\sigma_B + 2.706 $$

where $\sigma_B$ is the uncertainty of the background counts, given by the square root of the counts. The detector is assumed to be operating at a trigger point, $L_C$, which ensures the false rate is no greater than 5% since the corresponding circuitry is not modeled in the MCNP simulations.
In the simulation, the detector is first calibrated for its energy, resolution, and detection efficiency. Ideally, a point source that emits a single photopeak needs to be used for such calibration and be compared to the actual measurement. A low source activity is suitable to prevent pile-up events. However, the final calibration needs to consider the coincident summing, pile-up events, and even the decay during the measurement which will all affect the final calibration and needs to be measured experimentally. In the simulation, the diameter of the coil surrounding the detector end cap is also varied to find the optimum geometry for the experiment.

**Setting up of the HPGe detector geometry in MCNP**

The HPGe detector simulated in the MCNP model is set up according to the ORTEC GEM model series coaxial system. The final configuration can be modified straightforwardly based on any modifications to the system. The dimensions of the HPGe crystal of the simulated detector are listed in the Table 5.

<table>
<thead>
<tr>
<th>Component</th>
<th>dimension (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dead layer height</td>
<td>3.09</td>
</tr>
<tr>
<td>Dead layer diameter</td>
<td>0.86</td>
</tr>
<tr>
<td>HPGe crystal height</td>
<td>4.45</td>
</tr>
<tr>
<td>HPGe crystal diameter</td>
<td>5.90</td>
</tr>
</tbody>
</table>

The simulated spectra of a $^{137}$Cs is shown in Fig. 21 and is compared to a spectrum obtained by experiment. Here, the HPGe detector is not shielded. The point source was placed 6 mm away from the end of the crystal along the z-axis. In the simulated spectrum, the backscattered peak is missing because no material is placed surrounding the crystal. In a more realistic model, the end cap housing is added. The housing material is 1 mm thick Aluminum placed 3 mm away from the crystal. In addition, another 1mm thick stainless steel endcap is placed outside at 3 mm away from the Aluminum casing, as shown in Fig. 22.

![Figure 21. The simulated energy spectrum of 137Cs (left) compared to an experimentally obtained spectrum (right).](image-url)
Figure 22. The top and side view of the detector HPGe crystal with the Aluminum housing and the stainless steel endcap.

Setting up of the background and source distribution

The source is defined using the SP and SI options of the SDEF card in MCNP. The SP card is identified as containing bin probabilities for the SI L distribution by specifying option D. The SI card is the information card and interprets the inputs as discrete source variable values that are energies by specifying option L on the card. The gamma energies for the isotopes of interest are listed on the SI card in MeV, and the corresponding probabilities are listed on the SP card.

Figure 23 shows the background gamma spectrum taken experimentally in the PULSTAR reactor bay, which has been used to generate the source distribution card (SD) in MCNP. The figure also compares the background when the reactor is running at full power versus when the reactor is shutdown. A 4” lead shield was placed around the endcap of the detector, and when the shield lid is closed, it significantly decreases the background noise detected. We need to note that using this background noise is based on the assumption that the shield around the FGR measurement system is sufficient to maintain the background at the same level. Even though this is a plausible assumption because safety rules require that low radiation levels be maintained in the reactor bay, this background measurement needs to be re-confirmed once the FGR facility is running.

Figure 23. The gamma spectra of the reactor bay with the reactor running at full power and shutdown. The spectra were taken for 5 min with the lid of a lead shield open or closed.
Because the experiment is in real time after a steady state is established, the source can be corrected for its decay during the transit time through the helical loop surrounding the detector. Fig. 24 shows the source distribution with the minimal radius when the helical loop is tightly wound around the detector endcap. The decay of the source during the transit time is corrected on average over the whole source volume, and no unevenness of the geometric distribution has been taken into account (meaning the source is becoming weaker along its flow direction).

![Source distribution](image1.png)

Figure 24. The source distribution wrapping around the HPGe detector in MCNP. Top view on the left, and side view on the right.

The MCNP simulated spectra of the fission gases

Figure 25 shows the detected spectra from the MCNP simulation of Krypton and Xenon isotopes when the experiment is conducted at 1073 K at various flow speeds of the sweep gas. It is obvious that at this temperature, the counts in many of the photopeaks are much higher than the typical background obtained experimentally in the previous section, and also much higher than the detectability levels given by the Currie equation. At the lowest flow rate, some of the peaks are missing due to the low transport efficiencies of some short-lived isotopes. This is useful for identifying some of the long-lived isotopes with a much lower total Compton continuum and has also been discussed previously.

![Spectrum](image2.png)

Figure 25. Simulated gamma spectra of Krypton and Xenon at 1037K with sweeping gas velocities of 0.001 m/s (yellow), 0.01 m/s (red), and 0.1 m/s (blue).
Figure 26 shows the gamma spectra of the fission products with an experiment temperature of 1073K and a 0.1 m/s flow speed for a series of source distances. The inner radius of the tube coil around the detector end cap is altered in the simulation from 3.75 cm to 24.75 cm. At large coil radii, the count rates are much lower, but also with low coincidence and signal pile-up.

At the largest source distance, the spectrum is much noisier and some of the Compton edges can no longer be identified. Meanwhile, the background drops to the level of the environmental background and some of the peaks are not high enough to be clearly identified. The count rate dropped more than an order of magnitude when the coil radius increased from 3.75 cm to 12.75 cm, simply due to the decrease of the solid angle of the source that covers the detector.

The peak-to-Compton ratios are calculated using the total counts under a peak divided by the counts in the Compton continuum before the photopeak. Here, in order to compare the different loop radii with different counting rates, in the simulation the spectra are allowed to run for different amounts of time so that the peak heights are the same for consistent comparison. Under such conditions, the accumulation time for the spectrum with the largest loop radius is much longer due to its lower counting rate. Figure 27 shows the peak-to-Compton ratio of a series of photopeaks at various loop radii. It is clear that at the largest source distance, the photopeaks have the highest P/C ratio. This is due to the smaller solid angle of the detector crystal seen by the source at any point, so that the fraction of the true coincident event is lower. However, this cleaner spectrum is at the expense of a much lower count rate and much longer acquisition time.

If we take the background noise into consideration, it has been shown in Figure 26 that the Compton events are partially buried underneath the background, which would definitely deteriorate the P/C ratio at the largest source distance. This background needs to be measured again after the facility is commissioned, and added to the calculation of the final P/C ratios for different

Figure 26. Simulated gamma spectra for a sweep gas velocity of 0.1m/s and varying source distances from the detector.
loop radii. It is anticipated that a radius somewhere in between the smallest and the largest numbers simulated will be the optimal source distance.

![Figure 27. The peak-to-Compton ratio of select photo peaks with different loop radii.](image)

1.5 Simulations of the Furnace Heat Transfer and Cooling

In section 1.3 we discussed the COMSOL Multiphysics simulation results of non-isothermal and isothermal flow of the Helium sweep gas inside the U-shaped tube that partially resides in the furnace hot zone and also holds the fuel sample particles. The boundary condition of the part of the U-shaped tube inside the hot zone have been set to be equal to a certain set hot zone temperature. This is a plausible assumption that assumes the temperature uniformity of the furnace hot zone and its effectiveness of the heat transferred to the sweep gas, however, it needs to be modeled in more detail to not only confirm this assumption, but also to guide the design of the physical furnace structure and its associated accessories. This section focuses on the virtual design of the furnace, with the dimensional limits taken into consideration, and will examine the optimum heater geometry, the effectiveness of the thermal insulation, and possible heat leaks. The effect of an Aluminum heat exchanger will also be investigated, which as mentioned previously is used to 1) pre-heat the incoming Helium gas using the residual heat of the exhaust Helium, and 2) to cool down the exhaust gas before it enters the low temperature part of the furnace device to prevent it from causing any thermal damage.

**COMSOL Multiphysics simulations of a closed heating chamber**

In COMSOL Multiphysics, the model of a heating furnace is composed of mainly four parts: 1) a cylindrical-shaped thermal insulation layer with a hollow cylindrical hot zone in the center; 2) a series of heater rods/elements that are arranged at equal distances from each other and at a close distance from the inner surface; 3) a sample holder in the center of the hot zone; and 4) Helium buffer gas filling the hot zone.
In the heat transfer simulation, the external surface of the thermal insulation layer is set up as a heat emitting surface radiating heat to the environment (as vacuum), and no conductive heating interaction between the furnace and the environmental air is included. The diameter of the thermal insulation cylinder is limited to less than 6.5”, which is consistent with the inner diameter limit imposed by the configuration of beam tube #1. In addition, for safety reasons, an intermediate layer of Aluminum between the beam tube and the heating furnace is added to prevent their direct contact. However, the length of the cylinder is not limited. The cylindrical hot zone is 2” in diameter by 3” long. The configuration of the hot zone is shown in Fig. 28.

![Figure 28. The COMSOL model of the hot zone including the multiple heating rods and a metal block](image)

By applying a certain power to the heating rods, the temperature inside the hot zone and throughout the thermal insulation layer can be calculated using COMSOL, as shown on the left in Fig. 29. The figure on the right hand side of Figure shows the flow velocity of the Helium buffer gas due to the density change when heated, which helps to minimize the temperature gradient inside the hot zone.

![Figure 29. The temperature field (left) and the gas velocity field (right) of a closed furnace chamber simulated by COMSOL.](image)
In our simulations, various configurations of the heating filaments and input power have been tested, in order to achieve the best heating efficiency and temperature uniformity. The simulation also provides guidance for the materials properties that the actual furnace will need in order to achieve similar performance. Fig. 30 shows the temperature variations along the center axis of the cylindrical furnace, and the center part between the arc lengths of 3.5” to 6.5” designates the region of the furnace hot zone. It is clear that a simple U-shaped tube inside the hot zone would not guarantee a uniform temperature along the tube even with the Helium buffer gas. Here, the thermal resistivity has been assumed to be 4 W/m·K. It is noted that at a temperature of ~1400˚C, the external side wall of the furnace is heated up to several hundred degrees at thermal equilibrium, which could cause severe problems if the furnace surface is in direct thermal contact with the surrounding chambers and the reactor beam tube.

Figure 30. The temperature distribution along the center axis of the furnace cylinder. The part in between the arc length of 3.5” to 6.5” is the region of the hot zone.

Figure 31. The temperature distribution along the center axis of the furnace cylinder with a metal thermal block present inside the hot zone.
To reduce the temperature gradient, a block of Molybdenum metal is placed inside the hot zone area, as a sample thermal block to enclose the U-shaped tube. As shown in Fig. 31, the good thermal conductivity of the metal block greatly improves the temperature uniformity in the hot zone region. This minimal temperature gradient is preferable so that the Helium sweep gas can be more easily modeled as has been discussed in section 1.3. A non-uniform temperature field will, on the opposite, introduce more systematic uncertainties and complexities of modeling in this region.

In addition, the thermal insulation layer should have a low thermal conductivity to maintain a high enough temperature gradient, especially when the thickness of the layer is limited to only ~1.5” by geometric constraints. Since the heating power is equal to the integration of the temperature gradient, \( \nabla T \), over the materials total surface \( S \), times the thermal conductivity \( k \), we have

\[
\dot{Q} = \frac{\partial Q}{\partial t} = -k \int \nabla T \cdot dS
\]

Considering that the thermal layers on both flat ends is much thicker than the side wall, the temperature gradient that the chamber can maintain is mostly limited by the cylindrical side wall. If we only solve the above equation for a cylindrical shell with the outer surface at RT, the maximum temperature difference (from the outer surface) in the hot zone can be estimated by

\[
\Delta T = \frac{W}{2\pi kl} \ln(R/r)
\]

Here, \( W \) is the input power, \( l \) is the length of the cylinder which can be taken as 3”, and \( R \) (2.5”) and \( r \) (1.125”) are the outer and inner radii of the cylinder respectively. Given an input power of 300W, \( \Delta T \) could be as high as ~1300°C, for a material with thermal conductivity \( k<0.4 \text{ W/m·˚C} \). As seen in Figure, the highest temperature that can be achieved at an input power of 1000W is ~1100°C at the center of the hot zone, and the side wall of the hot zone can reach >500°C, when the thermal conductivity of the insulation layer is assumed to be 4 W/m-K. This is clearly not acceptable, and a much lower thermal conductivity is required. Given a more realistic number of 0.4 W/m-K that is consistent with the values that some good porous insulation materials can provide, the temperature gradient within the thermal insulation layer should be much higher. Fig. 32 shows the temperature distribution along the center axis with a much lower thermal conductivity. It is noted that to achieve the same temperature at the center, the power consumption is significantly lower. According to the COMSOL simulation results, an input power of ~300W would be sufficient to heat the hot zone to ~1750°C, with the temperature of the external surface at ~500°C (along the red curve and ~1.5” away from the hot zone boundary in Figure). Here the temperature difference, \( \Delta T \), is ~1250°C, which is rather consistent with the simplified theoretical estimates, however, a surface temperature of ~500°C is still not acceptable. This means that a passive heat dissipation method for the furnace (to be operable at >1000°C) is not sufficient to cool the furnace to a safe temperature range, and an active cooling device is necessary.

**Simulation of the water-cooled enclosure**

Given that the chilled water source will be located outside the beam tube at least 10-15 ft away from the water-cooled chamber, the maximum flow rate has to meet the cooling requirement at a reasonable feeding pressure. The pressure drop (in PSI) over the tube length can be estimated by the Hazen Williams equation,
\[ Q = 0.442C \cdot D^{2.63} (\Delta P / L)^{0.54} \]

where \( C = 150 \) is the Hazen-Williams coefficient of Aluminum, \( D \) is the ID of the tube in inches, and \( L \) is the tube length in feet. If we assume a pressure drop of 10 PSI over the whole length (back and forth) of the water route, the maximum flow rate a 0.18”ID tube can provide is \( \sim 1.5 \text{L/min.} \) This would allow a cooling power of

\[ W = C_p m \Delta T \]

where \( C_p = 4.186 \text{ J/g·°C} \), which is the specific heat capacity of water, \( m \) is the mass flow rate, and \( \Delta T \) is the temperature rise. If a cooling power of 500 Watts is assumed, a 5°C temperature rise at the cooling chamber will be observed according to the simple heat capacity calculation above, which seems to be very promising.

**Figure 32.** The temperature distribution along the center axis of the furnace cylinder with a metal thermal block present inside the hot zone and a thermal conductivity of 0.4 W/m·K for the thermal insulation layer.

In order to confirm this rough estimate, the effectiveness of a water-cooled jacket enclosing the furnace cylinder is simulated using COMSOL Multiphysics. Due to space limits, the thickness of the water layer needs to be minimized so that the thickness of the thermal insulation layer is not severely reduced. In the COMSOL model, a water layer 0.25” thick is set up surrounding the furnace, which is connected to a 1/4” inlet tube (ID of 0.18”) and a 1/4” outlet tube for water circulation. The water enclosure is made of thin Aluminum, so that an excellent thermal conductivity can be achieved. The chamber has multiple internal baffles to guide the flow of the water, as shown in Fig. 33. Here the water inflow speed rate is set to 1.5 m/s through a 0.18”ID tube, which corresponds to a flow rate of approximately 1.5L/min. The temperature of the water at the inlet is set to be 20°C. The simulation result shows that the chamber can be cooled fairly well with the local cold area close to the location of the water inlet where the water is injected into the chamber. The water is warmed up during its travel around the chamber and the returning half of the chamber is noticeably warmer than the other half. The center of the thermal insulation that is not covered by water is also warmer than the water temperature. The temperature of the water only slightly increased to 23°C at the exit, which is better than the simple theoretical estimate. This
is probably due to the fact that the heat is also radiated to the environment which is not considered in the rough calculation.

We also need to note that the gamma radiation emanating from the reactor also generates significant heating in the furnace chamber. From a rough estimate from the gamma spectrum of the reactor core, the thermal block itself would be heated by the gamma field at a rate of ~50W, which will add to our temperature baseline significantly. This can be corrected to some extent if we flow the Helium buffer gas through the chamber and actively cool the hot zone area if needed.

The gamma heating on the water cooled chamber would also increase the required water flow rate. From the heat capacity and water flow rate calculations, if we assume the highest flow rate of a water chiller is 1 gallon/min, and a temperature rise of 40°C in the returning loop, the total cooling power the water can provide is ~1×10⁴ J/s. This is more than enough to overcome the gamma heating of the chamber. We need to note that at such flow rate, the pressure of water needs to be approximately 60 PSI through a 1/4” tube (0.18”ID) over the length of 30 ft, which is quite high. Considering this, two sets of 1/4” tube water inlets and outlets are preferred. With two sets of water circulation routes, the water pressure only needs to be 15 PSI to obtain the same total flow rate.

Figure 33. Velocity of the water flow within the cooling jacket color coded by the flow streamlines (here color code has been rescaled to better differentiate the differences in water velocity). The water inlet and outlet are at the upper right of the diagram.
The stability and uniformity of the temperature is critical to the experiment, and it is important to minimize any disturbance to the temperature field inside the hot zone area. From the simulation result of the U-shaped tube in section 0, it is useful to implement a heat exchanger at the exit of the hot zone so that the outgoing Helium sweep gas can be quickly cooled down and the incoming Helium can be pre-heated. An efficient heat exchanger would also minimize the net heat that is carried away from the hot zone by the sweep gas so that the heating efficiency is increased and the temperature field is less perturbed. This will also prevent any damage that might be caused by the hot outgoing gas to non-heated areas.

A simple cylindrical heat exchanger that encloses the incoming and outgoing gas lines made of Aluminum is modeled in COMSOL. As shown in Fig. 35, the hot Helium gas traveling through the upper tube to the right can be quickly cooled down even at the highest flow rate we expect. The temperature drops to 200-300˚C from 1500˚C within a distance of 3”. Meanwhile, the cold
Helium is heated to 100-200°C from RT. This result suggests that a simple Aluminum heat exchanger should be sufficient for our purposes. We need to be cautious here though the actual heat exchange rate would probably be lower than in this ideal simulation because of imperfect thermal contact between the gas tube and the Aluminum block. However, to our benefit the heat capacity of Helium is small given the low density of Helium and its flow rate. The total amount of heating power that the Helium gas withdraws from the furnace is only <1% of the total input power. Therefore, the heat exchanger would act more as a safety precaution for the system. Similarly, the heat of the hot zone can also leak through the metal gas tubes that carry the sweep gas through the insulating layer in terms of thermal conductivity. Figure 36 shows the COMSOL simulation result for the furnace when the metal gas tubes are included and conducting heat away from the hot zone. It is found that no significant temperature rise is taking place outside the furnace when the gas tubes and the heat exchanger are in direct contact with the external flanges. The heat dissipation from the water cooling system as well as radiation to the environment is sufficient to keep the all the external surfaces close to RT.

Figure 36. Heat leak test of the furnace through the gas flow tubes

1.6 The Design of the FGR Measurement Facility Subsystems

Overview of the Design Principles and the Main System

The mechanical design of the system is restrained by a number of factors, including the material properties and availability, and the feasibility of fabrication and associated costs. We also need to evaluate the impact to measurement uncertainties due to any deviation from the ideal simulation conditions. The major limiting factors and design considerations are summarized in Fig.37. The diagram on the right lists the components, of which the physical design and geometric optimization were aided by using the simulation methods discussed in detail in the previous sections.
It is worth noting that the mechanical design and its geometric optimization may be an iterative process. The conceptual geometry is first tested with numerical methods, which then subsequently results in an initial mechanical design. In the later step, material property requirements, availability, and feasibility of fabrication need to be investigated. In a second round, the models for numerical simulations are accordingly built based on the initial physical design to confirm the expected performance. In some cases, the process takes multiple rounds before the final design is reached.

Figure 37. A diagram of system design principles and limiting factors that need to be taken into consideration.

Figure 38. A schematic of the design of the main FGR instrument.

A drawing of the main FGR measurement system is shown in Fig. 38. Roughly speaking, the main system contains two layers – the enclosure (the outer layer) and the high temperature furnace and the connection structure (the inner layer). Note that the high temperature furnace has its own enclosure that is independent from the outer layer. The sample holder is in the middle of the hot zone of the furnace, and the Helium gas can be swept through the sample and subsequently fed into a helical loop that circles around the end cap of an HPGe detector. The whole furnace that carries the sample can slide along the axis inside the outer layer so that the neutron flux incident on the sample can be manipulated. All the gas and water tubes are fed through the outer layer so
that the entire system is encapsulated to further protect the beam port. The following sections will discuss the design of each system component in detail.

The Design of the Sample Holder

One of the most critical components of the system is the sample holding device, which needs to securely hold the fuel particles which are on the order of several milligrams in mass and ~100 μm in particle size, while simultaneously allowing the sweep gas to flow through the particles at ~1000°C and even higher temperatures.

The double mesh design of the fuel particle holder

The geometry of such a sample holding device is shown in Figure 39. The fuel particles are held between two commercially available electroformed Molybdenum grids, which are then enclosed in two cylindrical Molybdenum alloy (TZM) pieces with center holes. The meshes meet several requirements in this situation:

1. A small size (~3 mm in diameter) with a variety of opening sizes (100-300 μm) available that can be used for different types of fuel particles of small masses.
2. The open area is 50% and can be as high as 65%, which allows sweep gas flow.
3. Molybdenum is able to meet the high system temperature requirement.
4. Commercially available and cost effective (<$4 per piece).

Figure 39. The double-mesh sample insert that holds a layer of sample particles and allows the sweep gas to flow through.

Given the small sample amount that is allowed by the safety limit and the small size of the sample insert, the operation of sample loading has to be monitored under an optical microscope. Figure 40 shows a micrograph of some dummy stainless steel particles loaded onto a Molybdenum mesh taken by an optical microscope. The operation has to be performed accurately and a single-hair precision brush has been used.
This cylindrical sample insert is designed to work together with a sealed sample holder block that allows the switching out of samples and needs to maintain the Helium gas seal. The sample insert and holder block are designed to survive high temperatures, and incorporate an Alumina or graphite based sealing gasket. Both the cylindrical inserts and the sample holder block are made using TZM due to its higher recrystallization temperature. The mechanical strength of TZM is much better than pure Molybdenum after cooling down from high temperature, which is especially useful for maintaining the compression of the Helium gas seal. The internal structure of the sample block replicates the simplified U-shaped tube design that has been discussed in the previous sections. Figure 41 shows the machined sample inserts and the sample block.

![Sample Inserts and Block](image)

**Figure 40.** Pictures taken under an optical microscope of dummy particles loaded on a Molybdenum mesh.

![Sample Inserts and Block](image)

**Figure 41.** Left) the two-piece Molybdenum sample insert. Right) the sample holder block with cap. The two halves of the U-shaped gas channel and the position for holding the sample insert are shown. Gasket grooves surrounding the gas channel are also shown.

**The design of the sample stick**

The sample stick has two functions:

a) It mechanically holds the sample block inside the furnace chamber.

b) It provides the gas and power connections between the external gas lines to the sample block.
In addition, a clamp-on heat exchanger between the sweep gas lines on the cold side of the stick would help to pre-heat the incoming Helium sweep gas using the residual heat of the outgoing sweep gas, which is concomitantly cooled. The pre-heating of the sweep gas is useful in maintaining the designated temperature of the fuel particles. Figure 42 shows a drawing of the sample stick. The bottom plate has two miniature gasket grooves so that it can be sealed to the internal side of the chamber flange. Soft Aluminum gaskets may be used because the cold side temperature should not be significantly higher than RT. The Aluminum heat exchange also supports power wires that connect the power feedthroughs and the heating element (see Figure). The two connection tubes (1/4” OD) and the base plate are also made of TZM alloy and are welded to the sample holder block and the base plate.

![Figure 42. The design of the sample stick that makes the sweep gas connection between the external lines and the sample block. The two pieces of the clamp-on Aluminum heat exchange are also shown.](image)

**The Design of the Sample Furnace**

Due to dimensional limitations, and based on the COMSOL simulation results mentioned previously, the sample furnace was designed to be an air-tight high temperature (>1000˚C) furnace, with an outer diameter of <6” and with an Aluminum enclosure which also acts as a water-cooling jacket.

The design of the heating element

The heating element is designed with custom made TZM coils. This is necessary to accommodate the size limit for the hot zone since typical commercial heating rods that have specifications to above 1000˚C are bulky in size and are difficult to be implemented with such
small heating chambers. The electrical resistance of TZM is decent and with a 0.5 mm diameter wire, the total resistivity of each pair is ~0.9Ω, which makes the total resistance of the element ~7.2Ω. The heating filaments are configured into 8 pairs with one end connected so that it fits into the 16 grooves of the 99.6% pure Alumina candle, as shown in Fig. 43. The multiple pairs of 0.5mm heating filaments should provide enough redundancy, heating uniformity, and sufficient lifetime at high temperature.

Figure 43. Left) one pair of the TZM filament coil. Middle) the supporting structure of the heating filaments made of pure Alumina. Right) the 8-pairs of TZM heating filaments fitted into the supporting candle and connected to the connector pins.

The design of the furnace body

The COMSOL simulations have shown that due to the limitation on the furnace size, a water cooling jacket needs to be provided in order to maintain a safe external temperature for the furnace chamber. This unfortunately requires further reduction of the thickness of the thermal insulation layer, and as a tradeoff, the water layer has to be as thin as possible but still thick enough to provide enough water flow and hence cooling power.

The furnace chamber is also designed to be air tight, so that it can be filled with a Helium buffer gas. This is to prevent the oxidation of the TZM filaments at high temperature since Molybdenum has a strong affinity for oxygen at elevated temperatures. The chamber needs to be made of Aluminum for minimum neutron activation. Figure 44 is a schematic drawing of the water-cooled furnace chamber, and the thickness of the water jacket layer is ~0.3”. The limitations on the outer and inner diameter of the water-cooled chamber also impose challenges on the geometric configuration of the water inlet and outlet. A typical water-cooled chamber has the water inlet/outlet attached to the side, so that the gasket groove (or the knife edge) can be machined on the flat end of the chamber. However, this is not compatible with our tight dimensional limits. As shown in Fig. 44, the water lines have to be attached to one end of the chamber, which requires that this end be fabricated as a single piece and not detachable. The loading of the heating element and the thermal insulation layer must take place on the other end of the chamber. The power and gas feedthroughs have to be connected to the same side as the water lines, since the loading side is facing the reactor core, requiring the chamber to have openings on both ends.
The thermal insulation layer and the sample chamber

As mentioned previously, the thermal insulation layer around the furnace should have a very low thermal conductivity to maintain a high enough temperature gradient at a thickness limited to only \( \sim 1.5" \). The result from COMSOL multiphysics simulation suggests that a thermal conductivity of \( \sim 0.4 \) W/mˑK is adequate to maintain an approximate 1250°C temperature increase (relative to the environment) at an input power of 300W. The requirement for such a thermal conductivity can be achieved by using highly porous Alumina/Silica, which can also operate at such high temperatures. The final configuration of the thermal insulation layer is composed of two layers of high temperature porous material that can be custom machined. Table 6 lists the two select materials from Zircar Ceramics that have a high maximum temperature and low thermal conductivity. The SALI-2, which is composed of 80% Alumina and 20% Silica, and can withstand 1800°C and also has a 1340°C softening temperature, will be used as the inner layer of the furnace. The AL-30AAH, which has even lower thermal conductivity, will be used as the outer layer. Given such low thermal conductivities, the furnace should be able to provide \( \sim 1400°C \) with an input power of 250W, based on the above equations.

### Table 6. The materials properties of the chosen thermal insulation layers

<table>
<thead>
<tr>
<th>Material</th>
<th>Layer</th>
<th>Composition</th>
<th>Max temp</th>
<th>Thermal conductivity 250°C</th>
<th>Thermal conductivity 1350°C</th>
<th>Softening temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>SALI-2</td>
<td>Inner</td>
<td>( \text{Al}_2\text{O}_3 ) 80%, ( \text{SiO}_2 ) 20%</td>
<td>1800°C</td>
<td>0.15 W/mˑK</td>
<td>0.34 W/mˑK</td>
<td>1340°C</td>
</tr>
<tr>
<td>AL-30AAH</td>
<td>Outer</td>
<td>( \text{Al}_2\text{O}_3 ) 98.4%, ( \text{SiO}_2 ) 1.6%</td>
<td>1700°C</td>
<td>0.12 W/mˑK</td>
<td>0.23 W/mˑK</td>
<td>---</td>
</tr>
</tbody>
</table>

Figure 45 shows the exploded geometry of the two-layer design of the thermal insulation structure. All the end caps have offset corners to minimize heat loss. As mentioned earlier, the thermal insulation layers need to be loaded from the front end (the end facing the reactor core) of the furnace chamber due to dimensional limitations. The heating element resides inside the inner can of the insulation, and its power connections go through multiple holes in the layer and connect to the power feedthrough of the chamber flange. Since the whole chamber is closed and air/vacuum tight, the power feedthroughs are sealed to the flange adjacent to where the sample stick base is mounted. On the end of the hot zone, heating filaments are attached to a series of TZM rods that
act as power conducting wires. In order to make the connection between the two ends, a detachable supporting structure has to be made for the assembly of the whole furnace chamber.

![Figure 45. An exploded view of the two-layer thermal insulation structure.](image)

The final design of the sample furnace

Figure 46 shows how the power feed through rods are connected. On the furnace side, the power wires from the heating filaments are extended to the outside of the furnace. On the capping side, the sample stick is attached to the endcap flange that also carries the power feedthroughs as well as gas feedthroughs. An adaptor neck is attached to the endcap flange to make the chamber connection to the furnace. At the same time, the heat exchanger clamped to the sample stick (also see Fig. 42) also acts as a supporting structure for a series of wire-guiding rings with multiple holes that allow the power wires to slide in. A series of connecting pins are used to make the mechanical connection between the two halves. The sample stick also has a clamp-on thermal insulation structure that seals the opening of the hot zone. The assembled structure of the complete sample chamber is shown in Fig. 47.

![Figure 46. The connecting mechanism of the power feeding rods](image)
The temperature controlling and monitoring system

As shown in Figure 48, the heating element of the furnace is driven by an external DC power supply connected to a temperature controller. The maximum power of the DC power supply is 1kW, however, according to the COMSOL simulation results, the furnace may not need this full power to drive the hot zone to the 1000°C level. The feedback of the control loop is composed of two sets of thermocouples for redundancy in case one of them fails. These pairs of thermocouples are fed into the hot zone through a straight center tube of 1/4” diameter. The temperature reading will be the average of the two thermocouples to reduce uncertainties, and each thermocouple will act as the backup of the other.

Because the thermal insulation layer has very low thermal conductivity, the heating of the sample would be very responsive. However, the cooling of the system will be slow, and a separate control loop for cooling is needed. The cooling is performed by flushing the Helium buffer gas (not the sweep gas) through the hot zone, the flow rate of which can be controlled via a mass flow controller. This cooling loop can also be used as a safety mechanism.
The High Temperature Irradiation Resistant (HTIR) thermocouples, which are fabricated at Idaho National Lab, are also immune to high irradiation given their superb stability and performance under such conditions. The HTIR thermocouples use non-standard materials, and they have to be calibrated and compared to other standard types of thermocouples, which is done at INL. Figure 49 shows the output voltages of several standard types of thermocouple as a function of temperature. The output voltages of HTIR thermocouple is shown in black points and the curve is fitted as a 5th order polynomial.
The Design of the Inner Layer

The sample heating chamber will be positioned at the end of beam tube #1 adjacent to the reactor core. The length of the beam tube is approximately 110” long, and all the gas feed lines, power lines, and the water cooling lines need to be extended to the outside of the beam tube (see Figure 50). This inner layer structure slides into the outer Aluminum tube, which makes the whole instrument standalone and fully enclosed by an Aluminum layer so that the reactor beam tube is protected. The space in between the outer layer and the beam tube is filled with water (see Figure 51), so that the radiation field in this region is almost fully blocked. The inner beam is also connected to the end of a manipulator, so that the furnace can be retracted if needed to vary the neutron flux incident on the sample. There is a bushing made of pure Alumina close to the furnace end to support its weight and also facilitate the sliding of the entire inner layer inside the outer tube.

Figure 50. The connecting structure from the furnace/sample chamber to the external pumping station.

Figure 51. A schematic drawing that shows how the outer tube and the pumping station fit into beam tube #1.

This structure also acts as a supporting beam that holds multiple borated polyethylene and lead radiation shielding discs. In order to minimize the radiation streaming from the beam port during operation, multiple borated polyethylene and lead discs are needed to shield the neutrons and gammas from the reactor core. Based on the calculations, a thickness of ~6” of borated polyethylene discs will attenuate the neutron flux an order of magnitude. Here, 24×1” borated polyethylene discs are divided into two sections, each of which should attenuate the neutron flux by two orders of magnitude. The water and gas lines in between these two sections are bent slightly so that the two borated polyethylene sections are offset without any axially aligned through holes. The gamma rays will be blocked by 5×1” lead discs positioned close to the beam tube exit.

Because the shield poly discs and lead discs add significant weight to the supporting Aluminum inner beam, stress analysis has been done to assure the gravitational deformation is within an acceptable range. It was discovered that if the inner beam is only supported from the two ends, the
deformation would be large enough to allow the discs to sag and scrape the inner surface of the outer chamber. Considering this, two more Alumina bushings are added to the design in between the two shield sections to better distribute their weight along the inner beam. After this modification, the maximum deformation is within 0.002”, which is well within the allowed limit (see Figure 52). In addition, extra shield discs may be installed to further block the streaming radiation field out of the beam port.

![Figure 52. Stress analysis of the inner beam with 12 poly shield discs and 5 lead discs installed. The deformation has been scaled up, and the actual maximum deformation is on the order of 0.002” with extra supporting bushing structures.](image)

Another important aspect that must be considered is the insertion and extraction processes of the entire system in the reactor beam tube. The insertion procedure is more straightforward. First, the outer Aluminum tube with the sealing plate is installed, and the water seal is tested. Then the inner beam attached to the pumping station sitting on a cart is rolled into position, and a connection is made between the inner and the outer layer close to the beam port position. The extraction procedure is more complicated. The fuel samples and the chamber will be activated following irradiation, and in order to safely remove the furnace, the instrument has to be stored inside a shield room (discussed in following sections) to decay for a period of time. This requires that the shield room both accommodates the extracted part while it decays and does not interfere with reactor operations.

Figure 53 shows the two-step extraction procedure that will fulfill the above requirements. At first, the manipulator is detached from the pumping station due to the dimensional limitations imposed by the back wall of the shield room, so that the entire system can be pulled back at least 24”. The space between the end of the beam tube and the outer chamber will be filled with water, so that the furnace inside the outer chamber is not further activated during reactor operation. After most of the short-lived isotopes have decayed away in the instrument component (mainly in the furnace), the furnace chamber will be extracted. This will require that the pumping station is detached from the outer chamber tube. The inner beam is also sectioned so that the back section can be moved away. In this step, the outer tube is pushed forward again to make space for the furnace. It will also be filled with shielding blocks and closed off. The furnace and the front section of the inner beam will continue to stay inside the shield room for further decay until they are safe for handling and servicing.

43
Figure 53. A two-step retraction process that pulls the inner layer out for service

The above extraction procedure added the requirement that the inner connection/supporting beam needs to be sectional. The front section that is connected to the furnace chamber is relatively short, so that it can be pulled out of the beam port without touching the back wall of the shield room. The water lines, gas lines, and the power lines need to be disconnected while the two sections are separated. A push-pin structure is needed at the disconnecting point for the power wires, as shown in Figure 54.

Figure 54. The disconnection interface of the inner beam

The Design of the Outer Layer and the External Shield

The outer layer of the system is connected to a pumping station so that it is vacuum tight. The main tube that is inserted into beam tube #1 is made of Aluminum for minimal activation. This tube bears the weight of a series of borated polyethylene and lead rings for extra shielding, and also seals the water inside the beam tube. The water needs to be sealed in two locations, where the Aluminum plate on the outer tube bolts to the wall, and where this plate contacts the outer tube. Two sets of rubber gaskets are used to accomplish this, as shown in Figure 55. The wall plate is bolted directly to the external wall of the reactor, and the end plate is tightened to the wall plate to compress the rubber O-ring against the outer tube.
An external shield room around the beam port #1 area will be built to maintain the normal background radiation level in the reactor bay area. This is necessary even though the FGR main system already includes multiple layers of borated polyethylene and lead shielding. The amount of external shielding needed can be estimated by examining the neutron and photon source strength and the amount that exits the beam port. More specifically, the photon source term can be estimated by doubling the short-lived fission product inventory attenuated by a certain beam aperture. This aperture defines a volume of the reactor core that is aligned with the beam tube, which gives a volume fraction that is applied to the source term,

\[ f = \frac{\pi r^2}{ld} \]

where \( r \) is the radius of the aperture, \( l \) and \( d \) is the length and depth of the reactor fuel, respectively, which can be taken as 24” and 15” for the PULSTAR reactor. The inner radius of BT #1 is 3”, and the photon source term of an operating reactor at 1MW is estimated to be

<table>
<thead>
<tr>
<th>MeV Group</th>
<th>Average MeV</th>
<th>Reactor photons/s</th>
<th>BT#1 photon/s</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-1</td>
<td>0.5</td>
<td>3.40×10^{17}</td>
<td>2.67×10^{16}</td>
</tr>
<tr>
<td>1-2</td>
<td>1.5</td>
<td>8.42×10^{16}</td>
<td>6.61×10^{15}</td>
</tr>
<tr>
<td>2-3</td>
<td>2.5</td>
<td>2.06×10^{16}</td>
<td>1.62×10^{15}</td>
</tr>
<tr>
<td>3-4</td>
<td>3.5</td>
<td>5.42×10^{15}</td>
<td>4.26×10^{14}</td>
</tr>
<tr>
<td>4-5</td>
<td>4.5</td>
<td>2.29×10^{15}</td>
<td>1.80×10^{14}</td>
</tr>
<tr>
<td>5-7.5</td>
<td>6</td>
<td>1.11×10^{15}</td>
<td>8.74×10^{13}</td>
</tr>
<tr>
<td>total =</td>
<td></td>
<td>4.53×10^{17}</td>
<td>3.56×10^{16}</td>
</tr>
<tr>
<td>fraction=</td>
<td></td>
<td></td>
<td>7.85×10^{2}</td>
</tr>
</tbody>
</table>

The volume fraction can be calculated, and is equal to 7.88×10^{-2}. Of course, the FGR instrument occupying BT #1 would only leave a straight through aperture of ~0.19” in diameter.
which contains the thermocouples. The rest of the cross-sectional area of the beam tube is filled with multiple layers of shields, which result in a very small spot of the leaked gamma field.

The neutron flux has been calculated using MCNP and examined experimentally in section 0. The thermal fluence rate can be roughly estimated to be $1.3 \times 10^{12} \text{n/cm}^2 \cdot \text{s}$ at 38 cm from the center of the reactor core. However, this rate is estimated and measured based on an array of thin Aluminum plates, instead of the actual furnace, which would attenuate the neutron flux more significantly. The recalculated thermal neutron flux is shown in Fig. 56, and it is estimated that at the 38 cm position, the fluence rate is dropped to $\sim 4 \times 10^{11} \text{n/cm}^2 \cdot \text{s}$.

![Figure 56. Thermal neutron flux calculated from MCNP based on the designed furnace chamber with appropriate materials](image)

Given the dimensional limitations in the area external to the beam port, the total distance from the center of the reactor core to the inner surface of the external shield room is $\sim 14.5$ ft. If we use the inverse square law, and assuming a straight beam opening is present in the main FGR measurement system, the incident neutron flux on this rear wall is calculated to be $\sim 1.36 \times 10^9 \text{n/cm}^2 \cdot \text{s}$. It is noted that the fraction of fast to thermal neutrons decreases with distance from the core initially due to interactions with the fuel loop test apparatus. At distances beyond 60 cm, BT1 is filled with shielding and lesser volumes of test equipment. The non-thermal neutron fluence rates can be estimated based on the MCNP calculations. The calculated fluence rates at 60 cm were adjusted using the inverse square of the distances to give the fluence rates at 442 cm (14.5 feet). The fast neutron fluence rate of $1.26 \times 10^8 \text{n/cm}^2 \cdot \text{s}$ can be obtained at the inner shield wall, assuming the beam tube is not shielded.

For neutron shielding, approximately 1 cm of Borated polyethylene is equivalent to 2 cm of concrete or 4 cm of lead, and the tenth value layer (TVL) of concrete is approximately 40 cm. When calculating the neutron albedo dose, a value of 0.7 is assumed for the neutron albedo dose for the worst case at 90 degrees, i.e. where the beam scatters off of the inner wall of the shield. Materials and total shield thickness required to achieve external personnel dose-equivalent rates of less than 2 mrem/h are listed below.
Table 7. The estimated shielding thicknesses required

<table>
<thead>
<tr>
<th>Material</th>
<th>East (inch)</th>
<th>South (inch)</th>
<th>West (inch)</th>
<th>Top (inch)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Concrete</td>
<td>24</td>
<td>24</td>
<td>36</td>
<td>36</td>
</tr>
<tr>
<td>Lead</td>
<td>4</td>
<td>10</td>
<td>4</td>
<td>6</td>
</tr>
<tr>
<td>BPE</td>
<td>36</td>
<td>36</td>
<td>27</td>
<td>28</td>
</tr>
</tbody>
</table>

Microshield was used for the exposure rate calculation for the direct beam. The exposure rate at the sides and top of the shielding adjusted the dose rate at the inside wall (beam stop) with an exposure albedo factor, attenuation, buildup, and distance. The Microshield results showed that the shielded exposure rates vary from ~0.05 mR/h to ~0.1 mR/h at different locations outside the shields. In addition, the HPGe detector for the FGR measurement is shielded at least by another 4” of lead, which further drops the total exposure rate to <4 μR/h of gamma energy from 0 to 6 MeV.

Non-thermal neutrons are more of a concern for neutron dose-equivalent rates from scattering and penetrating a shield. Thermal neutrons are more likely to interact by absorption producing activated materials and gamma photons, i.e. capture photons from the (n,γ) reaction and photons emitted from the decay of activated materials. From the above discussion, fast neutrons are 1.5 percent of the thermal neutron fluence rate and have an albedo dose factor of 0.3 to 0.7. NCRP Publication 51 provides dose-equivalent per unit fluence (rem cm²) from neutron scattering and capture interactions vs the thickness of concrete (g cm⁻²). Shielding estimates are made using the trend line equation for thermal and fast neutrons for equivalent concrete mass density thicknesses.

Figure 58. The design of the shield room outside the beam port #1.
Figure 58 shows the design of the shield room directly outside beam port #1. The east and west side walls of the room have sufficient shielding to meet the requirement given in Table 7. However, due to dimensional limitations, the south side of the shield needs to be addressed mainly inside the BT1. The external shield thickness is primarily 24 inches of concrete. At the center area of the shield wall, additional layers of lead (8”) and BPE (8”) will need to be supplemented to catch the radiation from the beam port.

The total dose-equivalent rate is given by the sum of the neutron and radiative capture dose-equivalent rate and reactor photon exposure rate outside the BT 1 shield room, and is estimated to be approximately 1.7-2.1 mR/h, as shown in Table 8. We need to note that the dose rates here are relatively conservative (high) estimates since in the design mentioned in previous sections, the BT1 is filled with multiple layers of Borated polyethylene plates that accumulate to a total thickness of 36”。 In addition, multiple lead discs would be needed to stop a significant fraction of the gammas. The only unshielded aperture is 0.18” inner diameter associated with the center tube penetration for the thermocouple wires. However, this tube is filled with two 1/16” diameter Molybdenum sheathed thermocouple wires, which roughly occupies a quarter of the opening volume and cross sectional area. This can be treated as a quarter of the beam tube length is fully filled with such material, roughly equivalent to 30 inches of lead. The remaining neutron and gamma leakage out of the BT1 is fairly focused and can be caught by a beam end pocket made of BPE and lead, which are at least 12” and 8” thick respectively. The size of the pocket depends on the divergence of the leaked radiation field.

### Table 8. The total dose-equivalent rate at different locations outside the shield.

<table>
<thead>
<tr>
<th>Location</th>
<th>Neutron Rate</th>
<th>Gamma Rate</th>
<th>Total Rate 1MeW</th>
<th>Total Rate 1.8 MeW</th>
</tr>
</thead>
<tbody>
<tr>
<td>South Side (beam stop)</td>
<td>1.00 mR/hr</td>
<td>4.83×10^{-2} mR/hr</td>
<td>1.05 mR/hr</td>
<td>1.89 mR/hr</td>
</tr>
<tr>
<td>East Side</td>
<td>0.88 mR/hr</td>
<td>2.27×10^{-1} mR/hr</td>
<td>1.11 mR/hr</td>
<td>2.00 mR/hr</td>
</tr>
<tr>
<td>West Side</td>
<td>0.88 mR/hr</td>
<td>7.33×10^{-2} mR/hr</td>
<td>0.95 mR/hr</td>
<td>1.71 mR/hr</td>
</tr>
<tr>
<td>Top</td>
<td>0.86 mR/hr</td>
<td>1.00×10^{-1} mR/hr</td>
<td>0.96 mR/hr</td>
<td>1.73 mR/hr</td>
</tr>
</tbody>
</table>

**Other safety measures of the FGR system**

The safety measures taken to protect the reactor and the test instrument are a critical aspect of the design considerations. Part of this has been discussed in previous sections relating to the mass limit of the fuel samples, and to the design of the internal and external shielding of the radiation fields. There are several additional safety features that are implemented into the system design.

One safety feature is the design of the furnace heating control and monitoring system. As mentioned before, multiple heating coils and thermocouple wires are installed in parallel inside the furnace to provide enough redundancy to run the experiments. A separate system monitors the temperature outside the furnace. These monitoring systems verify the correct operation of both the furnace heating and cooling systems. A feedback loop is implemented from the water chiller to the temperature controller, so that if the water chiller fails, the furnace heating elements are instantly turned off. In addition, the buffer gas needs to be flushed through the hot zone to assure its temperature quickly drops from high temperature if required. The integrity of the cooling water...
lines are monitored, both by the chiller itself (which is automatic) and by water sensors and coolant flow monitors.

Another safety feature is related to the monitoring of the fission gases. Any leak from the fission gas sweep line would result in contamination of the other parts of the instrument and could cause safety concerns in the reactor bay area. Therefore, in addition to the sealed furnace and gas tubes, the outer layer of the system is completely sealed so that any gas leakage can be detected since the space between the inner layer and the outer layer is maintained under vacuum. The outer layer also prevents the beam tube from coming into contact with the external surface of the furnace and cooling assembly.

Finally, in addition to the automated redundant safety shutdown systems, manual override and shutdown buttons are provided for the furnace heater and other critical systems.

1.8 The Implementation and Testing of the FGR Measurement Facility

The Assembling and Testing of the Sample Heating Furnace

Figure 59 shows the assembled sample stick. The flange that holds the power and gas feedthroughs including the gas tubes are made of commercially pure Titanium. Calculations show that at this position, the Titanium and TZM will not be activated significantly, or the activated isotopes are all short lived. The two TZM tubes are laser welded to the base plate and the sample holder block, and the Alumina insulation plugs are clamped onto these two parallel tubes to close off the furnace. The options for gasket material that can perform under such high temperatures is limited. Two types of gasket materials, Alumina-based and graphite based, have been tested. The Alumina-based gasket is stable and will not oxidize at high temperature. Unfortunately, this mechanically flexible material is too porous to effectively seal the Helium sweep gas, and the graphite-based gasket is finally chosen. The graphite-based gasket is also bendable and easy to machine, but will oxidize at high temperature. Considering this, in the experiment, the hot zone must be flushed with Helium before operating at high temperatures. A precision cut fitting the gasket materials to the designed gasket groove was performed using laser cutting.

Figure 59. The fully assembled sample stick attached to the end flange of the furnace chamber.
It has also been discovered that the laser welded joints of the TZM sample holder were extremely brittle and delicate. For extra safety precaution, it will be run at a slight negative pressure inside. Considering this potential problem, a Titanium alternative has also been fabricated to run at slightly lower temperature ranges (below 1000˚C) than TZM (see Fig. 60). The welding of Titanium is much easier and robust, and the vacuum test showed no sign of leaking. At the same time, this sample holder block has a simpler geometry and a flat gasket groove.

![Figure 60. A Titanium version of the sample holder stick (Left) side view. (Right) Top view.](image)

**The Assembling and Testing of the Sample Furnace**

The supporting candle of the TZM filaments are made of pure Alumina, as shown in Fig. 61. Two end plates are also made to protect and confine the heating filaments. The top plate also has feedthrough holes for the filament wires allowing them to be connected to custom made TZM pins.

![Figure 61. The heating filament supporting candle made of Alumina.](image)

The winding of the TZM filament is performed on a center tube that is mounted to a slowly rotating platform. Caution has been taken to assure that all the filaments have the same number of turns and diameter, so that the voltage and current during heating is consistent. The 8 pairs of filaments are first connected in series and combined into 4 pairs to provide optimal resistivity, and then connected in parallel to provide enough redundancy in case any filament fails.

The TZM filament wires are connected to a series of TZM rods fed through specially made TZM connectors. Once assembled, the heating element is positioned inside the two layers of
Alumina/Silica foam, as shown in Fig. 62. Then the insulation cap is positioned while the power connector rods are guided through a series of holes in the cap. Finally, the entire assembly is slid into the Aluminum water cooled chamber.

Figure 62. The assembling of the main furnace structure including the heating element.

The water-cooled chamber is sealed by a soft 1100 Aluminum ring gasket. The gasket has to be made thin so that the inner diameter is flush with the inner diameter of the chamber in order to accommodate the insulation layer. The rings are cut by EDM, with a width of 0.1” and a diameter of 5”. The knife edge at the end surface of the chamber and the gasket are shown in Fig. 63. The weld bead on the internal surface of the chamber have to be removed to accommodate the outer diameter of insulation layers. The Aluminum furnace chamber has been leak tested by connecting to a high vacuum chamber. The vacuum level can reach \( \sim 1 \times 10^{-7} \) Torr when the water cooled chamber is not loaded with the porous insulation and the heating filaments. The setup of the vacuum test is shown in Fig. 64. When the chamber is fully loaded, a rough vacuum of \( \sim 1 \times 10^{-3} \) Torr can be achieved due to the presence of the highly porous insulation layer. Under running conditions, this chamber will be pumped down and flushed with Helium several times to assure that the buffer gas contains minimal Oxygen content and will prevent the oxidation of the heating filaments.

Figure 63. Left) the knife edge at the end surface of the water cooled chamber. The weld beads on the internal surface have to be removed to allow the insulation layers to slide in. Right) the thin Aluminum gasket sitting on the knife edge after testing.
The temperature controlling and monitoring system

The heating element was first tested in open air to low temperatures below 100°C to minimize any possible oxidation, and to test the response of the chamber. The 8 sets of heating filaments are connected in series to an Ametek 1000W DC power supply. The output of the power supply is remotely controlled by the temperature controller via a 0-10V analog DC output signal. Fig. 65 shows the experimental setup of the low temperature test in air. The wire leads of the filaments are temporarily bridged by copper connectors in series, and the thermocouple is fed into the center hole of the chamber. The temperature controller shows the trend chart of the set point, power output, and the measured temperature of the system as a function of time.
With the success of the initial tests, the temperature control system was then tested at higher temperatures with the chamber actively pumped under vacuum and water cooled. The vacuum at high temperature was maintained at ~0.3 Torr using a rotary mechanical pump. The highest furnace temperature tested was 1200°C, while the exterior surface of the furnace chamber was maintained at a temperature of 20°C by the circulating cooling water jacket. At 1200°C, the steady state temperature inside the furnace was maintained with a fluctuation of less than 1°C with a total heating power of ~250W. These test results indicate that the TZM heater filaments were responsive and the insulation layer was effective (see Fig. 66). The total power consumption also roughly agrees with the COMSOL simulation results. The assembled chamber is shown in Fig. 67.

**Figure 66.** The temperature controller data of a test where the set temperature (blue curve), the output power (purple curve), and the actual temperature (red curve) are shown.

**Figure 67.** The final assembling procedure of the furnace chamber.
The inner connection beam

The inner connection beam is an Aluminum tube with lateral access openings milled along its axis. The tube carries multiple power lines and the gas/water lines, as well as supporting BPE discs for neutron shielding. The power lines are protected by Alumina tubes and run through the interior shielding blocks, as shown in Fig. 68.

Figure 68. The inner connection tube that carries the power lines and multiple shielding discs.

The space between the inner connection beam and the outer tube is also shielded with polyethylene and lead discs (shown in Fig. 69). As mentioned previously, the inner beam is sectioned into two parts to facilitate a better operational process when extracting the system from beam tube #1. For this purpose, the power lines and the gas/water lines can be connected and disconnected at the joint of the two sections. The power lines are connected using a push-pin structure as shown in Fig. 70.

Figure 69. The extra polyethylene and lead shielding blocks (Left and middle) and the quick power connection joint (right).

The back section (closer to the beam port exit) of the inner beam is significantly longer than the front section (closer to the reactor core), and is also made of Aluminum alloy 6061. This section is equipped with multiple layers of polyethylene discs. In addition to the polyethylene blocks for neutron shielding, multiple lead blocks are also fabricated to minimize the gamma leak from the beam port.

Figure 70. The back section of the inner connection tube and the lead discs.
The remote control of the manipulator

The position control of the furnace is realized using a motorized manipulator with 24” travel distance. As shown in the previous chapter, this manipulator is located inside the shield room where the radiation level is high during reactor operation and has to be controlled remotely from outside. The absolute and relative positions can be of the furnace structure can be controlled accurately either by using the remote control software (see Fig. 71) or by a physical front control panel (see Fig. 72).

![Remote control software of the manipulator](image1)

**Figure 71.** The remote control software of the manipulator.

![Front control panel of the manipulator](image2)

**Figure 72.** The front control panel of the manipulator.
The Outer chamber with the Pumping Station

The outer chamber of the main system is composed of the pumping station and a 6.45” OD Aluminum tube, which fits into beam tube #1 with an ID of 6.5”. The fitting test was conducted with success (Fig. 73), and left only a minimal amount of contamination on the tube which was easily removed. Following the fit test, a standard 8” Aluminum flange was welded to the end of the tube, and an Aluminum plate was welded to the other end to seal it.

Figure 73. The test fit of the outer Aluminum tube into beam tube #1.

The flanged-tube was then vacuum tested with the pumping station chamber attached. The associated chamber, ports, and the pumping station were also tested (see Fig. 74). The tests resulted in a vacuum of \( \sim 10^{-8} \) Torr. At this vacuum level, any gas and water leak from the connection lines can be detected and can be identified utilizing a residual gas analyzer. The pumping station sits on a cart so that the device can be moved easily as needed.

Figure 74. The assembled outer chamber and the associated pumping station. The vacuum test was successful.
The Fabrication of the Lead Shields

The lead shields installed inside the inner beam with a diameter of ~2” are machined directly from standard lead bricks. The lead discs that are positioned in between the inner layer and the outer layer beams (OD~6”) are ordered from Nuclear Lead Co. Inc. Some of the larger lead rings (OD~15”) that are positioned in between the beam tube and the FGR outer layer external lead rings were made in house by melting multiple standard lead bricks and casting using an Aluminum mold (Fig. 75).

Figure 75. Casting the 15” OD lead rings.

The HPGe detector setup and calibration

A CAEN digital amplifier with on board MCA has been setup and tuned. The system energy resolution is similar to a well-tuned analog system, but it is expected that the digital system will have much better throughput with higher performance and less dead time. The low dead time would reduce the number of pulses that are treated as pile-up events in conventional systems and help to improve the counting accuracy, given that the absolute count rates are important. The top figure in Fig. 76 shows the interface of the digital data acquisition system, where the preamplifier pulses are digitized and the pulses generated by a real time trapezoidal filter based on FPGA. A number of shaping parameters can be adjusted to produce the best energy resolution. The bottom figure shows a representative energy spectrum of a Co-60 source taken at an input count rate of 1 kcps. The current energy resolution is found to be 1.85 keV at 1333 keV peak.

It needs to be noted that in order to count the amount of released fission products, the radioactive gases are designed to flow through a helical coiled tube surrounding the detector end cap, as discussed in the previous chapters. Here, acquiring absolute detection efficiencies with good accuracy are an important aspect of quantifying the total rate/amount of released gases. A standard liquid source is needed to calibrate the detection efficiency using the same helical geometry. A certified calibration multinuclide solution is acquired for such purpose.

In order to calibrate the efficiency of the detector, several standard point sources were also used, including the Eu-154, Cs-137, and Co-60. The activities of these sources were all approximately 5 μCi. The sources were positioned at various distances from the detector end cap. The efficiencies at different gamma
peaks of point sources are shown in Fig. 77. The calibration of these point sources is also compared to the multinuclide liquid source, as shown in Fig. 78. It is observed that the detection efficiencies of the EZIP source are consistently lower than that from the SRM source, which might be related to the slightly different geometry of the two sources and the self-attenuation of the liquid EZIP source. Further investigation will be conducted on the cause of this slight systematic shift.

Figure 76. (Upper) Preamplifier pulses and trapezoid filter pulses. Lower) Energy spectrum for calibration.

Figure 77. Efficiency calibration with point sources at various source-detector distances.
The Insertion Test of the Main Chamber

The test insertion of the main chamber into beam tube #1 took place after the reactor core was off loaded, and was divided into two steps. First, the outer tube was disconnected from the main pumping station, and was fit into beam tube #1 as shown in Fig. 79. The wall plate was attached and fastened to the exterior wall of the reactor. The outer tube can slide along inside the beam tube, and was inserted all the way to the inner end directly adjacent to the reactor core. The fixer plate was tightened with a rubber O-ring in between the two plates and the beam tube was then flooded with water to check for any leaks. The result showed no leaks of water.

Figure 79. The insertion of the system main chamber.

Figure 78. Efficiency calibration with standard liquid sources.
After the water seal test, the test chamber sitting on the pumping station was rolled into position and connected to the outer tube (Fig. 80). Vacuum test of the entire system and instrumentation was performed and found to be satisfactory. At this stage, full operation of the FGR and sample testing awaits licensing approval by the US NRC.

Figure 80. The main chamber inserted in beam tube #1 with connected instrumentation for initial testing.
2. UO$_2$ Fuel Samples and Characterization

Two types of UO$_2$ samples with different average particle sizes were examined using the on-site positron annihilation lifetime spectroscopy (PALS) instrument to study their defect properties. PALS measures the decay lifetime of positrons upon their first injection into the materials of interest, which is directly related to the electron density and is sensitive to the concentration and type of vacancy defect and defect clusters. The sample particles are loaded into a small sealed chamber. The sample loading process was done inside a glove bag filled with Helium gas to prevent any possible contamination of the environment. A $^{22}$Na source is placed in the middle of the particles to make sure most of the positrons emitted from the source are stopped inside the fuel sample. After sample loading is completed, the chamber is sealed and checked for any contamination before removing from the glove bag. The process is shown in Fig. 81.

![Figure 81. Left) the sample holder for PALS measurement of the UO$_2$ sample particles. Middle) a glove bag is used to load the UO$_2$ particles for PALS measurement. Right) pictures of the UO$_2$ samples of two different particle sizes.](image)

The PALS measurement is conducted using the bulk PALS system, as illustrated in the block diagram shown in Fig. 82. This system has complete digital signal processing and data acquisition enabled by a LeCroy Wavepro 7300A digital oscilloscope. The pulses from the PMTs are digitized and fitted with a customized algorithm in real time to extract timing information, which results in a sub-200 ps timing resolution. Meanwhile, an analog system runs in parallel with the digital system for higher count rate experiments which have lower requirements for timing resolution.

![Figure 82. A block diagram of the PALS instrument](image)
The PALS spectra of the two samples are shown in Fig. 82, and the spectrum fitting results are shown in Table 9. The two samples, labeled UO$_2$-50 and UO$_2$-170, showed very similar spectra and fitted lifetimes. The first lifetime, $\tau_1$, at ~215 ps is significantly larger than the typical lifetime of 160 ps in standard UO$_2$, but rather close to that in the ADOPT UO$_2$. In addition, the second lifetime component, which is in generally related to defect clusters and/or grain boundaries, has a very low intensity, indicating little positron trapping in such type of voids. The PALS results show that the two UO$_2$ samples are very similar in their morphology, even though their particle sizes are quite different. This may also suggest that these particles have a lack of grain boundaries, which should trap positrons and produce different positron intensities for the lifetime components when the particle sizes are different. This is considered desirable for meeting the final objectives of this project.

![Figure 83. The PALS spectra of UO$_2$ fuel samples](image)

**Table 9. PALS analysis results of two UO$_2$ samples with different particle sizes.**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Particle size (μm)</th>
<th>$\tau_1$ (ps)</th>
<th>$I_1$ (%)</th>
<th>$\tau_2$ (ps)</th>
<th>$I_2$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>UO$_2$-50</td>
<td>297-500</td>
<td>218±1</td>
<td>95.2±0.6</td>
<td>480±20</td>
<td>4.8±0.6</td>
</tr>
<tr>
<td>UO$_2$-170</td>
<td>88-105</td>
<td>215±1</td>
<td>94.2±0.6</td>
<td>470±20</td>
<td>5.8±0.6</td>
</tr>
</tbody>
</table>
3. Investigation of fission gas mobility mechanisms in fuel

During the course of this project studies were performed to explore the potential of using ab initio molecular dynamics to understand fission gas localization within the vacancy structure of UO₂ fuel. The transition frequency, barrier energy, and incorporation energy were extracted from the model, which demonstrated the ability for using such an approach for estimating key parameters needed for understanding fission gas mobility behavior.

Furthermore, as fission gas mobility is considered driven by vacancy migration mechanisms, a novel technique was explored for observing the motion of vacancies in a fuel matrix. In this case, the investigation was performed using classical molecular dynamics models and demonstrated the ability of establishing the dynamic structure factor of vacancies. This clearly enables direct extraction of diffusivity of vacancies and vacancy clusters.

The above techniques will be used to support interpretation of the data measured using the FGR loop.

Appendix A gives two summary papers that describe this work.
Project Publications


Appendix A
Ab Initio Molecular Dynamics Analysis of Xenon Incorporation in Uranium Dioxide

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INTRODUCTION

The fission process in nuclear fuel produces volatile fission product gases, such as xenon, that migrate out of the fuel and into surrounding structures. Consequently, these fission product gases may affect thermal performance and integrity of the fuel. Fission gas release models, used in modern fuel performance codes such as BISON, are currently under investigation to improve their accuracy and ability to simulate the fission gas release phenomenon and its consequences. For uranium dioxide (UO₂), the most commonly used fuel in commercial power reactors, fission gas release models are known to have significant uncertainties [1]. In the interest of improving such models, several complex diffusional mechanisms have been examined using a variety of techniques such as Nudge Elastic Band (NEB), utilizing both Density Functional Theory (DFT) and semi-empirical inter-atomic potential functions [2]. One proposed diffusion mechanism in UO₂ is through tetravacancy (i.e., 2UV + 2OV) structures, which is expected to provide a suitable means for the incorporation and diffusion of xenon in the fuel crystal.

In this work, a model was developed to study the incorporation of xenon in UO₂ via a tetravacancy using ab-initio molecular dynamics (AIMD). This is a first principles technique that abandons the use empirical force fields as usually is the case of classical MD analysis. Furthermore, in contrast with static transition analysis tools such as NEB, AIMD is able to capture the dynamics of a given process.

COMPUTATIONAL DETAILS

In AIMD the interatomic forces are not based on empirical fits, as with traditional molecular dynamics (MD) methods, but are instead predicted directly from the electronic structure. Equilibrium dynamics of a system may be simulated by AIMD using the Car Parrinello method [3], in which the Born-Oppenheimer approximation is assumed such that the dynamics of the simulation are defined on the time scale of the dynamics of the atomic system. In this work, density functional theory (DFT) is used to solve for a self-consistent solution to the electronic structure (i.e. electron density) at each MD time step. Subsequently, the system is evolved using forces that are derived from the electronic structure using Hellman-Feynman force theorem [4].

The AIMD simulations were performed using the VASP [5-6] DFT code and its associated PAW pseudopotentials with the GGA-PBE exchange correlation functions [7-9]. Additionally, a plane wave cutoff of 400 eV was used along with a 1x1x1 k-mesh. The simulation consisted of a 3x3x4 triclinic UO₂ super cell with the addition of a xenon tetravacancy (105 atoms) illustrated in Fig. 1. Using the canonical ensemble (NVT), simulated with the Nose thermostat, the system was equilibrated for 2 ps at 1500 K (taken as a representative PWR average fuel temperature [10]) with 4 fs timesteps [11]. Following equilibration, the system was run for an additional 4 ps with 4 fs timesteps to study the localization of xenon within the tetravacancy.

RESULTS AND ANALYSIS

Over the course of 1000 time steps, the ensemble was allowed to progress at equilibrium. As a measure of equilibration and verification that the AIMD model satisfies the ergodic hypothesis, the velocity distribution over the last 500 steps of equilibration was compared to the Maxwell-Boltzmann distribution. As required by the ergodic hypothesis and a well-equilibrated system, Fig. 2 shows that the velocity distribution approaches the Maxwell-Boltzmann distribution. The 350 m/s and 1250 m/s maxima in the velocity distribution correspond to the uranium and oxygen contributions to the velocity distribution respectively.

Following equilibration, the equilibrium dynamics of the xenon position data as well as the resulting energetics of the system were analyzed. Using the visualization capabilities in VMD, xenon motion within the tetravacancy was examined (Fig. 3).

In addition to the dynamic evolutions of the surrounding system, the xenon motion process is defined by the atom’s transition from the proximity of one uranium vacancy to the other. Motion of the xenon within the tetravacancy varied significantly in each of the three planes of motion. It is
important to note that the displacement illustrated in Fig. 3 represents only one of the three components comprising the total displacement shown in Fig. 4. In order to quantify the energetics of a xenon transition, the barrier energy was correlated with the displacement of the xenon atom from its starting position in the uranium vacancy (Fig. 4).

As it relates to Figures 3 and 4, the transition frequency ($\nu$ transitions/picosecond) for xenon is provided below in Table 1, along with xenon incorporation energies into trivacancy and tetravacancy defects. The values obtained from this work were found to agree reasonably well with past research. This shows that the AIMD model is describing the system with a reasonable degree of accuracy. The incorporation energy was calculated using the relation

$$E_{\text{incorporation}} = E_{\text{filled, defect}} - E_{\text{empty, defect}}$$

In each case (i.e., with Xe or without Xe), $E$ is the total energy of the equilibrated atomic system. Using the xenon density of states (DOS) along with a sinusoidal fit function, the transition frequency for the xenon motion sequence was determined and compared to simulation data (Fig. 5). Specifically, the 8.71 THz maximum from the xenon DOS was converted from angular frequency to transition frequency and checked for accuracy using a fit function of the same angular frequency and a spread similar to the simulation data.

**CONCLUSIONS**

The translation of xenon within a tetravacancy has been investigated using ab-initio molecular dynamics. The transition frequency, barrier energy, and incorporation energy were extracted from the model, demonstrating its ability to capture a variety of parameters and its potential for use in studying fission gas diffusive behavior. Future refinements to the model will seek to develop additional insight into Xe and Kr mobility and will likely involve executing more elaborate AIMD simulations to investigate this phenomenon.
Table I. Xenon transition frequency and incorporation energies.

<table>
<thead>
<tr>
<th>Incorporation Energy (eV)</th>
<th>This work</th>
<th>Previous work</th>
</tr>
</thead>
<tbody>
<tr>
<td>trivacancy</td>
<td>2.57</td>
<td>1.16 – 3.65</td>
</tr>
<tr>
<td>tetravacancy</td>
<td>3.48</td>
<td>2.00 – 3.93</td>
</tr>
</tbody>
</table>

**REFERENCES**

DIRECT VACANCY TRACKING IN URANIUM DIOXIDE USING MOLECULAR DYNAMICS

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INTRODUCTION

The production, migration, and release of fission products in nuclear fuel has posed a significant design challenge for many years [1]. Those receiving the most attention (i.e., xenon and krypton) exist in gaseous form and are thought to diffuse by the aid of vacancies on the host (UO$_2$) lattice [2]. Developing a better understanding of these intragranular phenomena requires a more comprehensive picture of vacancy mobility. However, with no experimental analogue, the ability to obtain such knowledge from simulation becomes necessary.

In this work, a classical molecular dynamics (MD) model for UO$_2$ was implemented for simulating oxygen vacancies at elevated temperatures. Using a new methodology, the resulting trajectories were used to track the defect population. This provided a valuable tool for directly quantifying the dynamic nature of vacancies in nuclear fuel. Additionally, these defect trajectories allowed the determination of various diffusive parameters of interest (e.g., vacancy diffusion coefficient, jump frequency, jump duration, correlation coefficient).

VACANCY TRACKING AND DIFFUSION

In crystalline UO$_2$, uranium and oxygen generally diffuse across several distinct sublattices (i.e., fcc uranium, sc oxygen, interstitial) via a process known as jump diffusion. Furthermore, this process also brings vacancies into the vicinity of fission gas atoms, potentially mobilizing them from their previously trapped states. While a variety of mechanisms may produce a set of equivalent outcomes, the concept of jump diffusion is quite simple and generally involves an atom (U or O in this case) hopping into an empty adjacent lattice site (vacancy). Equation 1, where \( \langle R^2(t) \rangle \) is the mean square displacement (MSD), applies irrespective of the specific diffusion mechanism and has been used extensively to yield diffusion coefficients of atoms and pseudo-particles (in this work) alike [3].

\[
D = \frac{1}{6t} \langle R^2(t) \rangle
\]

Typical investigations of diffusive species via atomistic simulation involve the inherent capability to track interacting atoms. Consequently, any dynamic property is readily determined from the atomic trajectories. However, the ability to directly track an arbitrary number of single vacancies, or any other pseudo-particle arrangement (e.g., vacancy clusters), in 3-D space over the course of an MD simulation is largely unavailable, if not non-existent. The algorithm developed in this work tracks these defects retroactively by taking advantage of a predefined pristine lattice with thermal fluctuation zones containing each lattice site. As an atom hops into an adjacent vacancy, the atom’s path is searched in a way that allows the concurrent reassignment of that vacancy’s position. This process is repeated as many times as necessary, and adjustments for more anomalous jump mechanisms are included.

To test and evaluate the methodology, oxygen vacancies were selected as a suitable species due to their high mobility at the temperature of interest (1700K). The expected relationship between the diffusion of oxygen atoms and that of oxygen vacancies (Eq. 2), where \( D_V^0 \) is the diffusion coefficient for the atom via vacancies, \( D_V \) is the vacancy diffusion coefficient, and \( f_V \) and \( C_V \) are the correlation coefficient and vacancy concentration respectively, holds under the equilibrium conditions studied in this work [4].

\[
D_V^0 = f_V D_V C_V
\]

MD MODEL AND SIMULATION

After removing a random set of 50 oxygen atoms from a pristine 6x6x6 crystalline UO$_2$ supercell (Fig. 1) containing 2592 atoms, the system was simulated using the LAMMPS code [5].

Fig. 1. 6x6x6 pristine crystalline UO$_2$ supercell (larger, black U atoms and smaller, red O atoms).
Measuring roughly 33x33x33 cubic angstroms, the system was equilibrated at 1700K under a Nose-Hoover thermostat in an NVT ensemble over 100 ps (1 fs timestep). Subsequently, the ensemble was run an additional 5 ns for the mobile oxygen vacancies to reach the asymptotic diffusion region. This process was repeated to achieve better statistics in the parameters of interest. The rigid ion potential of the Buckingham-Coulomb form (Eq. 3-4) used herein (Tables I-II) was selected from previous work by Harp [6] and was determined to be a suitable representation for the purposes of evaluating the vacancy tracking methodology. Here, $A_{ab}$, $p_{ab}$, and $C_{ab}$ are fitted parameters for interactions between species $a$ and $b$, $q_a$ and $q_b$ are the charges on species $a$ and $b$ respectively, and $r_{ij}$ is the distance between atoms $i$ and $j$.

$$E_{Buck}(r_{ij}) = A_{ab} \exp \left( \frac{-p_{ab}}{r_{ij}} \right) - \frac{q_a q_b}{r_{ij}}$$

$$E_{Coul}(r_{ij}) = \frac{q_a q_b}{4 \pi \epsilon_0 r_{ij}}$$

Table I. Mass and charge assignments for U and O atoms

<table>
<thead>
<tr>
<th>Species</th>
<th>Mass [amu]</th>
<th>Charge [elem. unit]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium</td>
<td>238.02891</td>
<td>3.826355</td>
</tr>
<tr>
<td>Oxygen</td>
<td>15.9994</td>
<td>-1.9131775</td>
</tr>
</tbody>
</table>

Table II. Buckingham potential parameters

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>U-U</td>
<td>0.00</td>
<td>1.00</td>
<td>0.00</td>
</tr>
<tr>
<td>U-O</td>
<td>873.9018</td>
<td>0.40392739</td>
<td>0.00</td>
</tr>
<tr>
<td>O-O</td>
<td>50259.34</td>
<td>0.15285</td>
<td>72.653</td>
</tr>
</tbody>
</table>

RESULTS

Once the atom trajectories were generated in LAMMPS, the vacancy tracking algorithm was used to output a variety of jump statistics (Table III) in addition to the vacancy trajectories. The oxygen atom diffusion coefficient was also computed for judging the calculated vacancy diffusion coefficient, $D_v$, in Eq. 2, along with the vacancy correlation coefficient, $f_v$.

Work by Govers et al. (using Morelon and Basak potentials) [7] and Kukla and Smirnova (using Morelon, Yakub, and Potashnikov potentials) [8] yielded diffusion coefficients for oxygen atoms and oxygen vacancies respectively in hypo-stoichiometric UO$_2$ that are consistent with the values obtained in this work.

Furthermore, the vacancy correlation coefficient determined from Eq. 2 was found to be less than one, as is expected for vacancy-assisted diffusion, but greater than the geometric correlation coefficients for sc lattices (on which O atoms and O vacancies generally hop).

Table III. Vacancy jump statistics and diffusion parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Avg Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jump Frequency (Single Vac.) [ps$^{-1}$]</td>
<td>1.347</td>
</tr>
<tr>
<td>Jump Duration [fs]</td>
<td>318.233</td>
</tr>
<tr>
<td>O Vac. Diff. Coeff. [m$^2$/$s$]</td>
<td>2.003x10$^4$</td>
</tr>
<tr>
<td>O Atom Diff. Coeff. [m$^2$/$s$]</td>
<td>3.419x10$^{-10}$</td>
</tr>
<tr>
<td>Imposed Vac. Conc. [%]</td>
<td>1.96</td>
</tr>
<tr>
<td>$^{3}$O Diff. [%]</td>
<td>1.707</td>
</tr>
<tr>
<td>Imposed Stoichiometry [O/U]</td>
<td>1.942</td>
</tr>
<tr>
<td>Vac. Correlation Coeff. (f$V$) [(%)]</td>
<td>0.868</td>
</tr>
</tbody>
</table>

By unwrapping the vacancy coordinates during the postprocessing stage, the MSD was computed, averaged (Fig. 2-3), and subsequently used with Eq. 1 to provide the oxygen vacancy diffusion coefficient. A similar method was used for determining the oxygen atom diffusion coefficient. This procedure also allowed the visualization of the vacancy trajectories both collectively (Fig. 4) and individually (Fig. 5).
CONCLUSIONS

In this work, a new methodology for tracking vacancies in nuclear fuel was devised for better understanding the intragranular processes by which fission products migrate. Numerous simulations were carried out at high temperature and imposed vacancy concentrations. Post analysis using the vacancy tracking algorithm yielded diffusion parameters that agree well with theory and previous work. Furthermore, the method provides a variety of visualization possibilities. Future work will likely increase the breadth and complexity of the target system as well as add additional capabilities to the algorithm.

ACKNOWLEDGMENTS

This work was partially supported by the US Department of Energy (DOE) Nuclear Energy University Program (NEUP).