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FY2018 Report on Fission Chamber Development at LANL

F2016 Venture Project (NA-22)

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FY2018 Report on Fission Chamber Development at LANL

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Introduction

This report provides a description of a subset of FY18 activities conducted by Los Alamos National Laboratory (LANL) as part of the Nuclear Physics: Cumulative Fission Product Yields (Fission R-values) task of the NA-22 sponsored F2016 Venture Project. Specifically, this report describes the design, fabrication and testing of the Mark II fission chamber that was fielded as part of fission R-value measurements on the Flattop critical assembly located at the National Criticality Experiments Research Center (NCERC).

Historically, fission product R-value measurements were performed by chemical separation and beta counting of samples from carefully controlled fission chamber experiments. These measurements form the basis for reporting data used in debris diagnostics and subsequent modeling. It was also the means to extract absolute cumulative fission product yields (yield per fission or Y_i/f for the i^{th} fission product) that are included in the international nuclear data libraries, and ultimately used in diagnostic evaluations and device modeling. The challenge for this effort was to design a functional fission chamber given the harsh neutron and gamma fields and severe space limitations of the Flattop critical assembly.

Within the LANL R-value framework the absolute number of fissions, N_f, in a macroscopic sample can be determined by the expression

$$N_f = K_i^{j,k} A_i^{j,k}$$

where $A_i^{j,k}$ is the laboratory measured activity of the i^{th} fission product, corrected to zero time, and $K_i^{j,k}$, the so-called K-factor, is a proportionally constant that is determined in a "fission chamber" experiment. The superscripts recognize the fact that the measured activity, and thus the K-factor, will vary with the identity of the fissioning isotope ($j =$ ^{235}U, ^{239}Pu, ...), and the neutron energy spectrum that caused fission ($k =$ thermal, fission, 14 MeV, etc.). Rearranging the above expression, we get

$$K_i^{j,k} = \frac{N_f}{A_i^{j,k}} = \frac{\epsilon_i N_f}{f_i \lambda_i Y_i^{j,k}}$$

and we need but make an accurate measurement of N_f, the number of fissions that occurred in the sample to produce the measured activity of fission product i, to determine the constant of

proportionality[1]. Furthermore, if we expand $A_i^{j,k}$, the time corrected measured activity of the i^{th} fission product, we see that it is a function of the true fission product yield, $Y_i^{j,k}$, the number of atoms of the i^{th} fission product at zero time, times the decay lifetime λ_i, folded with the detector specific efficiency ϵ_i and the branching ratio f_i of the decay radiation being measured. Note, however, that the K-factor is counter specific. It must be experimentally determined for each radiation counter that will be used to determine the number of fissions occurring in a sample based on the measured activity of the i^{th} fission product. Similarly, the yield of the i^{th} fission product can be determined directly from a fission chamber measurement as follows

$$Y_i^{j,k} = \frac{N_i^{j,k}}{N_f} = \frac{A_i^{j,k}}{\lambda_i N_f} = \frac{f_i}{\epsilon_i \lambda_i K_i^{j,k}}$$

Where $N_i^{j,k}$ is the number of atoms of the i^{th} fission product at zero-time.

This last equation also shows how it is possible to use the K-factor to convert the measured activity of the i^{th} fission product to the corresponding absolute fission product yield for the fissioning isotope and neutron energy spectrum used in the measurement. This also demonstrates the need to accurately determine the other independent variables used to make these determinations, as the uncertainties associated with each component will add in quadrature.

Fission chamber measurements rely on two distinct fissile or fissionable samples. The first is generally called a reference or fission foil and is located inside a fission chamber, or more generally a gas proportional counter. The reference foil is intentionally kept as thin as possible to ensure near 100% efficiency to detect at least one of the two fission products. The fission chamber is intended to only count the number of fissions occurring per unit mass of the reference foil, and does not provide any information on yields of the individual fission products. The second sample, usually referred to as the macro foil, is considerably larger in mass and is intended for radiochemical analysis (dissolution, chemical separation and counting) to determine the individual fission product yields per unit mass of the macro foil. In order to determine the yield of a particular fission product per fission occurring in the macro foil, often called the "absolute" yield of the i^{th} fission product, it is necessary to mass scale the number of fissions that occurred in the reference foil to the macro foil. Assuming an isotropic neutron field at the location of the fission chamber and macro foil the number of fissions in the macro foil can be calculated as

$$N_{f,m} = \frac{m_m}{m_r} N_{f,r}$$

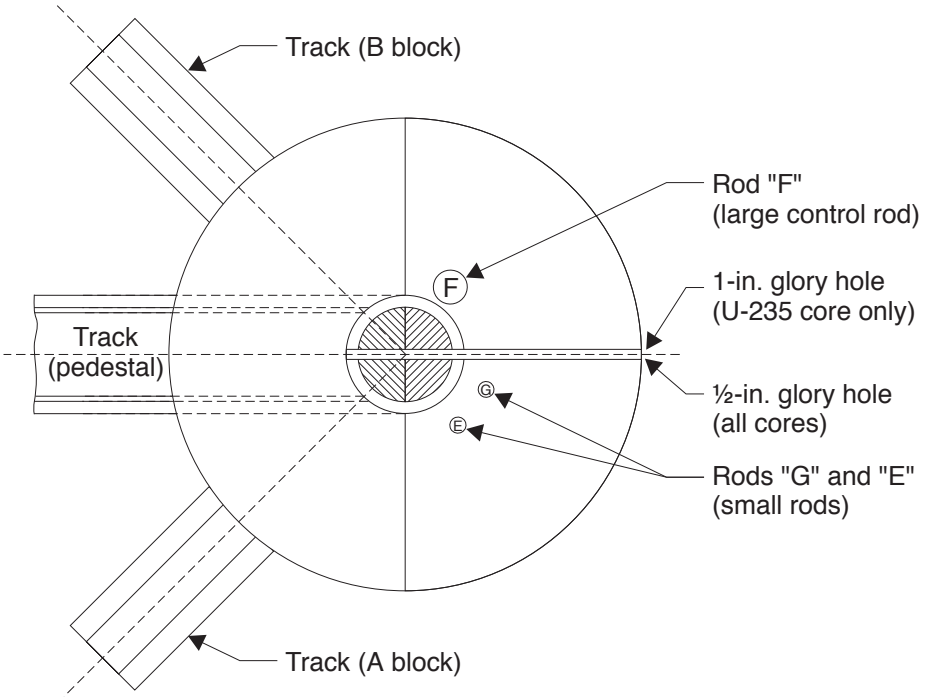
Where $N_{f,m}$ and $N_{f,r}$ are the true number of fissions occurring in the macro and reference foils, respectively, and m_m and m_r are the corresponding masses. The obvious requirement is that the two foils be of the same elemental and isotopic composition. Also, since no detector is ideal it is necessary to carefully characterize the operation of the fission chamber.

[1] Although this sounds simple in theory, in practice it is not. The devil is in the details.

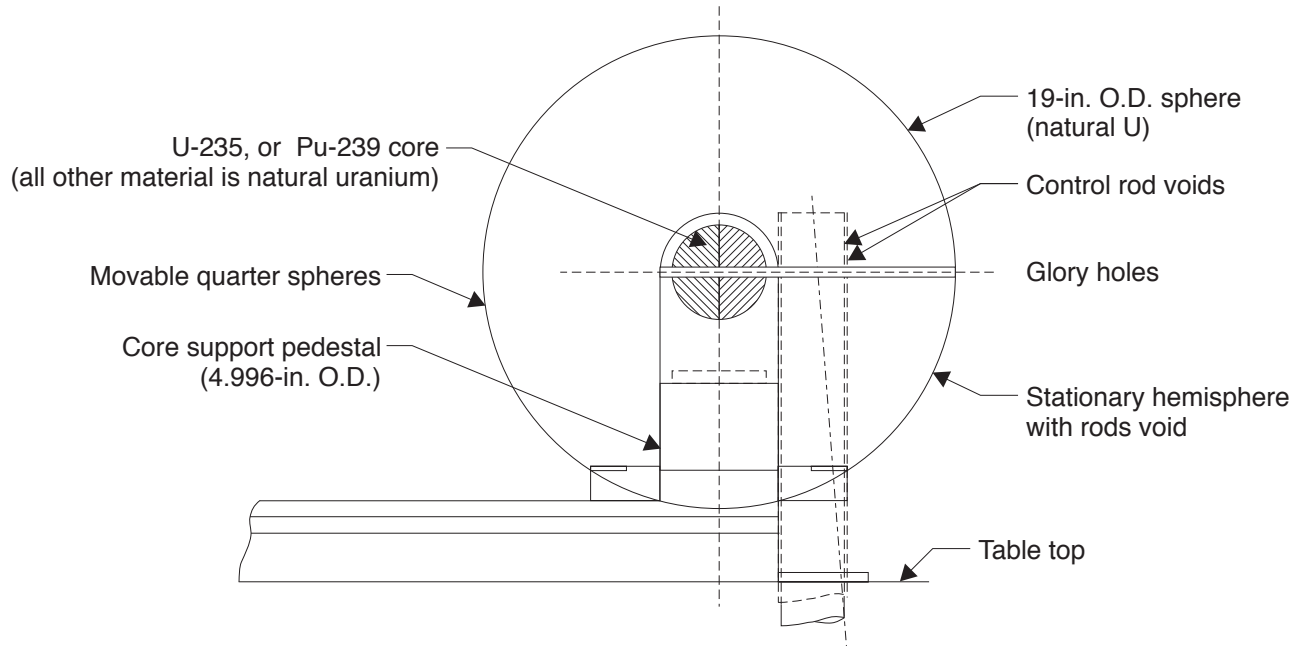
Flattop

The Flattop-HEU assembly, shown in Figure 1: Photograph of the Flattop-HEU assembly at TA-18., consists of a 5-inch diameter spherical core of highly enriched uranium (HEU: enriched to 93% ^{235}U) surrounded by a 7-inch thick natural uranium (^{nat}U) reflector. Plan and Elevation view drawings of the Flattop-HEU assembly are shown in Figure 2. Samples and other measurement equipment can be inserted into the assembly through a ½" diameter "glory hole" that is identified in the Plan View. There is also a set of fuel plugs of several lengths that can be loaded into the glory hole. The flexibility of the Flattop assembly is demonstrated by the fact that the neutron environment (flux and energy spectrum) that is sampled in a particular experiment can be customized, within certain limits, through proper selection of sample and fuel plug loading within the glory hole. Although the drawing indicates a 1" diameter glory hole for the ^{235}U (HEU) core, and also for the ^{nat}U reflector, a reducing sleeve is used to bring the diameter down to ½". Unfortunately, the reducing sleeve for the HEU core has been stuck for some time, so 1" is not an option.

Figure 1: Photograph of the Flattop-HEU assembly at TA-18.



Plan View



Elevation View

Figure 2: Plan and elevation views of the Flattop-HEU critical assembly. Figure was taken from the International Criticality Safety Benchmark Evaluation Program (ICSBEP) Handbook: HEU-MET-FAST-028.

Fission Chamber Design (Mark I)

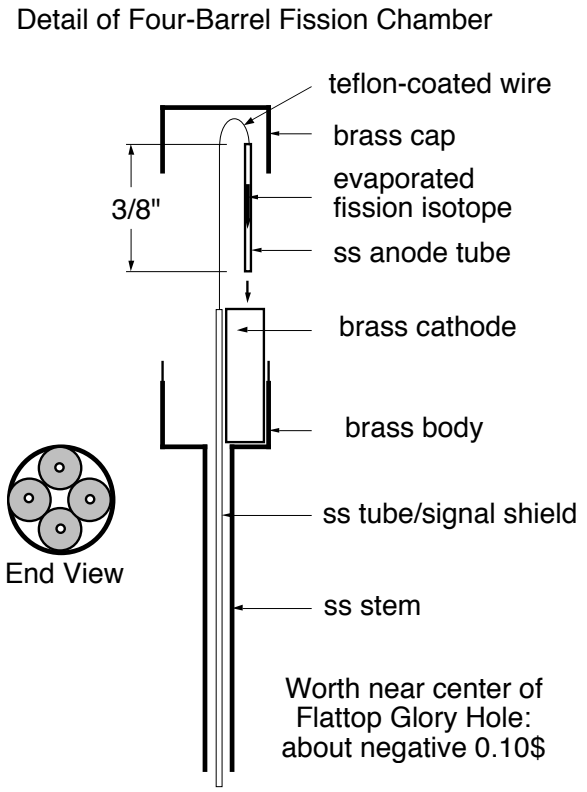
As stated previously, the challenge was to design and build a fission chamber that would fit into the Flattop-HEU glory hole while providing the required data fidelity. To do this we started with the LACEF[2] "four-barrel" design, shown in Figure 3, so named because it consists of four radial ionization chambers, each with a different fissile/fissionable isotope evaporated onto the central anode pin. This design has been used to measure relative reaction rates as a function of position and loading in the Flattop glory hole since the late 1950's, and they are still used at NCERC today. Unfortunately, this geometry presented several issues that would have made the desired measurement significantly more difficult if not impossible. Specifically, the mass scaling between the reference and macro foils is very sensitive to uncertainties in ether. The documentation for the existing four-barrel fission chambers did not include uncertainties on the masses of the actinide deposits. This implies at best 50% uncertainty in each of the recorded masses. Furthermore, after much debate it was determined that an accurate measure of the deposited mass in this geometry would be far from straightforward, and would likely suffer from hard to quantify systematic uncertainties. Finally, the mass scaling also depends on the assumption that the two samples experience the same neutron field. This is easiest to defend if the two have the same geometry, and are collocated to the greatest extent possible. These requirements are best achieved using two parallel disks, one inside and the other outside the active volume of an axial field fission chamber. The former is commonly referred to as a fission or reference foil (micrograms of material) used to count fissions per unit mass during an experiment, while the latter is the macro-foil (10s to 100s of milligrams of material) that would undergo destructive radiochemical analysis following irradiation.

A prototype fission chamber meeting the requirements given above was developed in FY16-17, also shown in Figure 3, which kept the general body shape and gas/signal line, but required a complete redesign of the reference (fission) foils and internal signal generating components to satisfy the requirements for the measurement. In its simplest form, an axial field fission chamber is nothing more than two parallel plates separated by a gas, and between which is applied an electric potential. Charged particles, such as fission fragments, travelling through the gas undergo collisions with the atoms or molecules of the gas, ionizing them into positively charged cations and free electrons. Both begin to move under the influence of the electric field, and the electrons are collected at the positively charged plate, or the anode. This provides the detector signal that is then recorded. When the gas pressure and electric field are properly balanced the collected signal is proportional to the energy deposited in the gas by the passage of the charged particle.

In this design the reference foil and macro foil (labeled ⑥ in Figure 3) were set at ¼" diameter, and positioned adjacent to each other at the front face of the chamber. The anode ⑦ is located at the back of the cylindrical active volume, and the collected signal is readout through semi-rigid coaxial cable that runs through the gas tube ⑨. The detector is pressurized to ~80 psig of

[2] Los Alamos Criticality Experiments Facility (LACEF) was the name applied to TA-18, the site where the critical assemblies were housed and used before they were moved to the Nevada National Security Site ca. 2004.

P-10 gas (a mixture of 90% Ar and 10% methane), which is commonly used in such applications. A schematic of the full fission chamber, less the pressure gauge and fill valve, is provided in Figure 4.



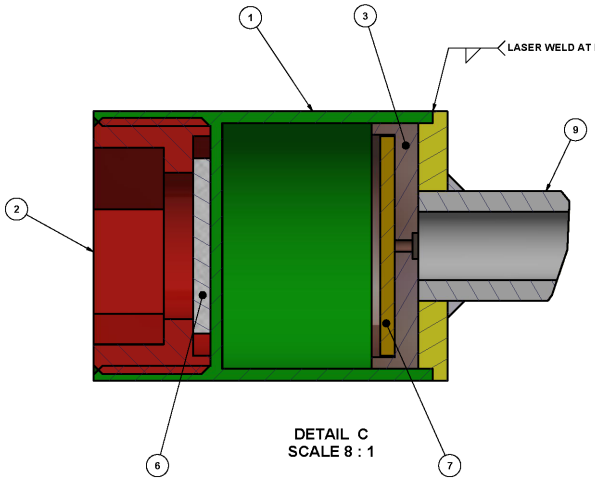


Figure 3: Schematic of the LACEF "four-barrel" fission chamber (left). Schematic of the new fission chamber design (above). The new design consists of a two-piece brass body ① that contains the active volume of the fission chamber and the macro foil ⑥. The reference foil is just inside the active volume opposite the macro foil. The anode ⑦ is opposite the reference foil and rests inside a MACOR® insulator ③. Electrons created by passage of fission products through the gas are collected at the anode, and the resulting signal is carried through a coax cable that runs inside the gas tube ⑨ to an electrical feed-through at the back of the detector.

Figure 4: Schematic of new fission chamber. The detector body shown in Figure 4 is on the far left. The pressure gauge and fill valve would be on the far right.

As mentioned previously, it is important to maintain a well-defined geometrical relationship between the macro and reference foils. To this end we designed a set of electrodeposition cells to properly center a ¼" diameter deposit of uranium for the reference foils that would match the diameter of the macro-foils and their position within the fission chamber. The deposition

cell and an example plate of elemental copper (easier to see than the uranium deposits) is provided in Figure 5. The deposits were made by dissolving small amounts of the same material used for the macro foils, followed by electrodeposition onto a 5-mil titanium substrate. This was deemed to be sufficiently rigid to prevent deformation while loading into the fission chamber.

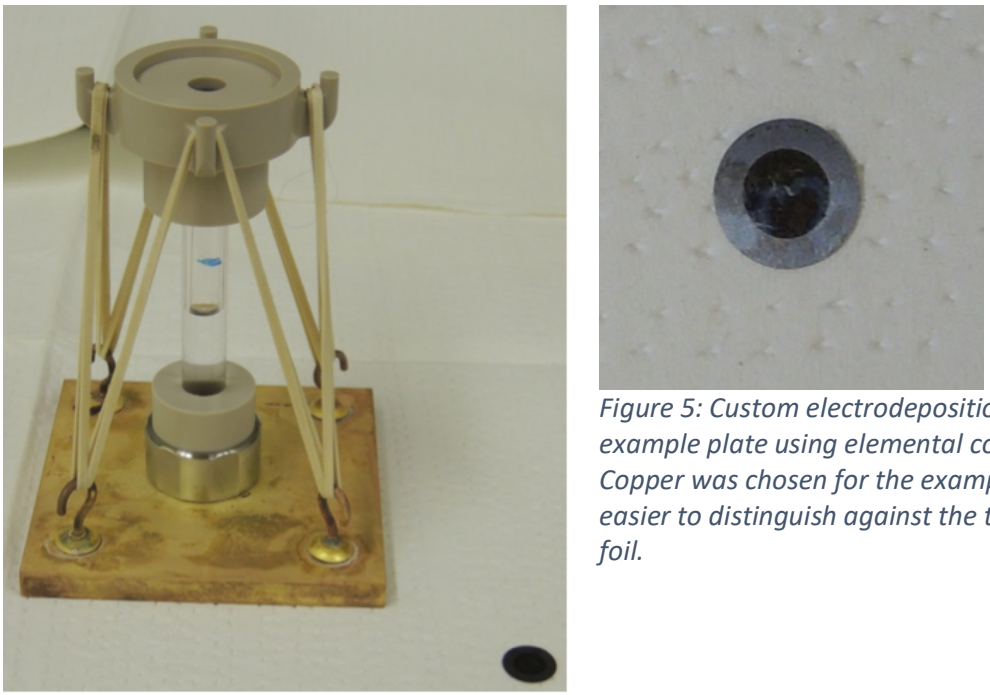


Figure 5: Custom electrodeposition cell (left), and example plate using elemental copper (above). Copper was chosen for the example because it is easier to distinguish against the titanium backing foil.

In order to properly characterize variations in the masses it was decided to prepare a range of deposit masses centered around what was calculated to be the optimal mass based on expected count rate in the fission chamber. For example, given the parameters that have been used on the Flattop assembly in the past we determined that the optimal mass for the HEU reference foil was about 3-5 μg. We therefore prepared ten reference foils ranging in mass from 0.5 to 10 μg. These were then assayed by α counting using several detectors located in the Radiochemistry Counting Facility at TA-48, including commercial Ortec α-spec and two different custom designed counters, a set of two 2π gas proportional counters and a Frisch gridded ionization chamber. The latter two had to be partially refurbished as they had not been used in many years.

Of the ten HEU reference foils we selected one, with 3.19 μg total mass of U(93) as determined by the counting methods mentioned above, to load into the first prototype fission chamber. Seven of the remaining ten reference foils were then subjected to destructive analysis by isotope dilution (^{233}U) inductively-coupled plasma mass spectrometry (Thermo X-series II equipped with an ESI APEX de-solvation sample introduction system). These results were then compared to those obtained by non-destructive α counting, and the results are presented in

Error! Reference source not found.. A linear fit to the data indicates an extremely high degree of correlation. However, the ID-ICPMS data are systematically higher than the results from the non-destructive assay. This is still being investigated, but we believe the bias is the result of the calibrations of the refurbished α-counters. We currently have a plan to clear this up over the next month or so.

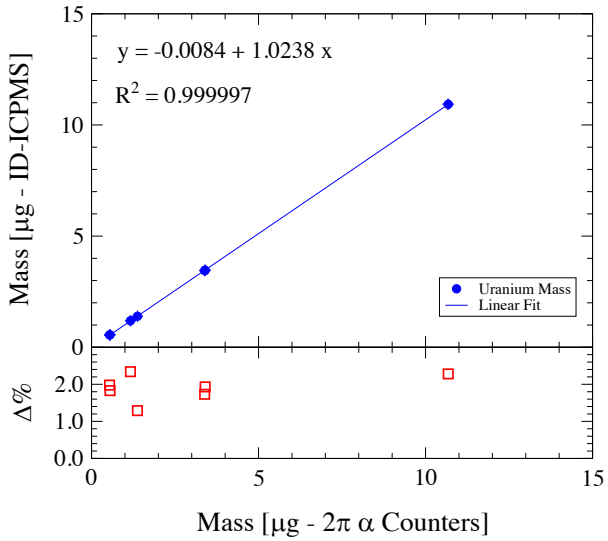


Figure 6: Reference foil mass calibration between non-destructive ⍰ counting and Isotope Dilution (ID) ICPMS (top). Relative difference (%) between the two assay methods (bottom).

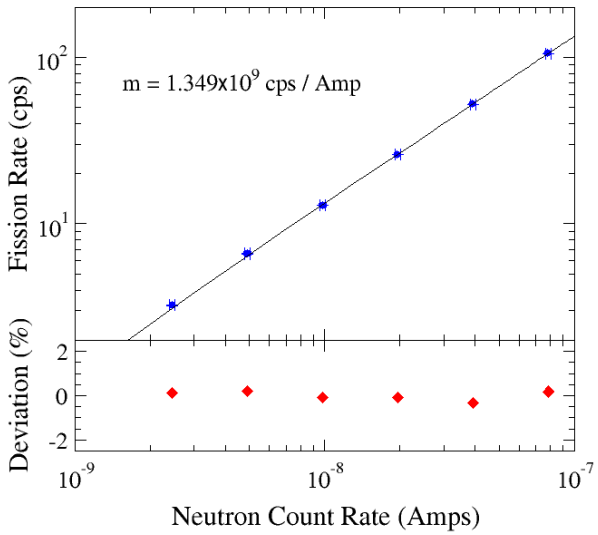


Figure 7: Plot of the fission rate (in counts per second, or cps) observed in the fission chamber as a function of the neutron count rate or power level (in units of Amps) as measured by the linear 1 compensated ion chamber.

While there are still details to work out, the cross calibration between destructive and non-destructive assay of the reference foils provided the necessary confidence that we can make the required mass scaling between the reference foil used in the prototype, assayed only by non-destructive methods, and the macro foils that were subjected only to destructive analysis.

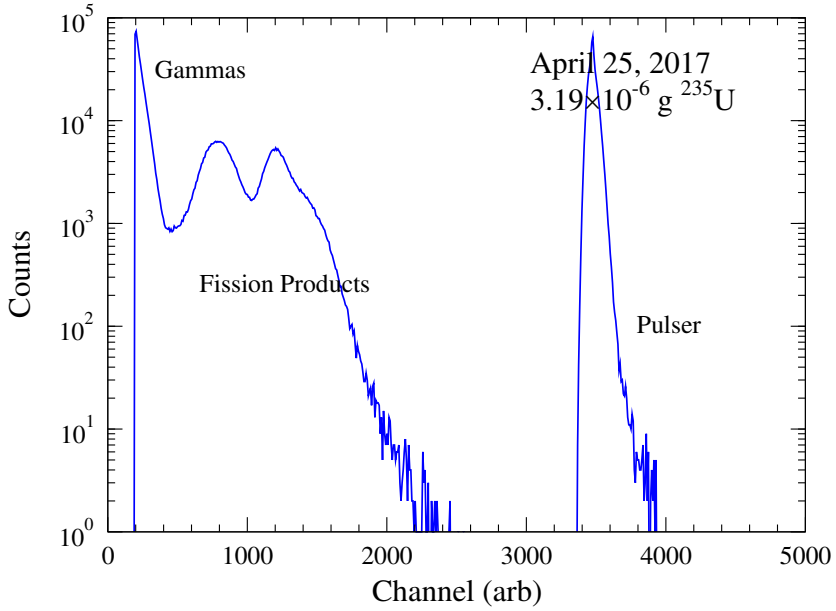


Figure 8: Pulse-height spectrum for fission products measured during the second test on April 25, 2017. The signal sources, gamma rays from the assembly fuel, fission products and the research pulser, are indicated in the plot.

Fabrication and Testing of the Mark II Fission Chamber

In Q2 of FY18 we completed fabrication of parts for the Mark II fission chambers and assembled seven units. Figure 10 and Figure 11 below are examples of the Mark II. The main change from the Mark I was the back-end, shown in Figure 11, which was specifically redesigned to reduce leakage of the P-10 gas and thus allow a longer use time before needing to be recharged. This was necessary because we still do not have the means to charge the chambers at the DAF, and it must be done at LANL before shipping the detectors to the DAF. We worked with a LANL Pressure Safety Officer to certify the P-10 charging stations and the fission chambers. We are currently waiting on guidance to certify a duplicate P-10 charging station for use at the DAF. This would greatly facilitate use of the fission chambers as they would no longer need to be shipped back to LANL for re-filling.

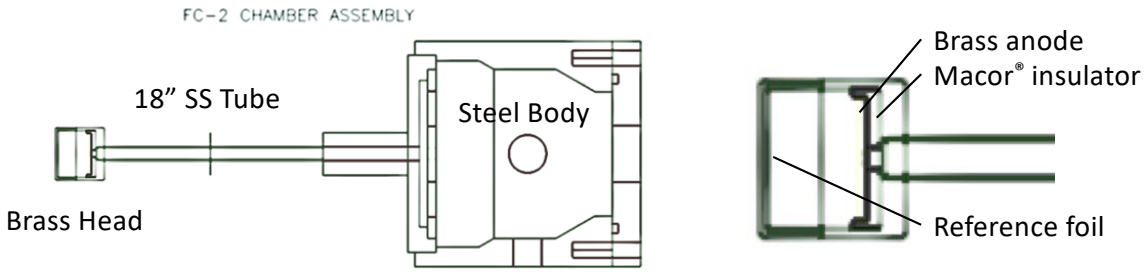


Figure 9: Schematic of the Mark II Fission Chamber (left) showing the three primary components, the Steel (Break-out) Body, the 18-inch stainless steel (SS) Tube, and the Brass Head. The close-up of the Brass Head (right) shows the locations of the reference foil, the Brass anode and the Macor® insulator.

The aluminum break-out of the Mark I was replaced by a more robust steel break-out body in the Mark II as shown on the left in Figure 9. The break-out body provides the volume and surface area to attach the pressure gauge, the gas fill valve and the high voltage/signal feedthrough. All connections were welded except for the pressure gauge, which was threaded in with high pressure epoxy, and the back flange for the SHV electrical feedthrough. The back flange was attached to the main part of the break-out using an industry standard O-ring seal with six equally spaced screws. The only modification to the brass head, shown on the right of Figure 9, was the removal of the eternal cup to capture the macro-foil at the front of the head. It was determined in the 2017 tests that this feature was no more practical than using our standard samples capsules to keep the macro-foil close to the reference foil, but resulted in greater dose to the individual recovering the samples. Figure 10 is a photograph of four assembles fission chambers ready to be loaded with reference foils, and Figure 11 is a photograph of the break-out showing the SHV feedthrough on the left and the gas fill valve below. This particular fission chamber was loaded with a 7.86 μg HEU reference foil and was used in the February 2018 detector tests at NCERC.

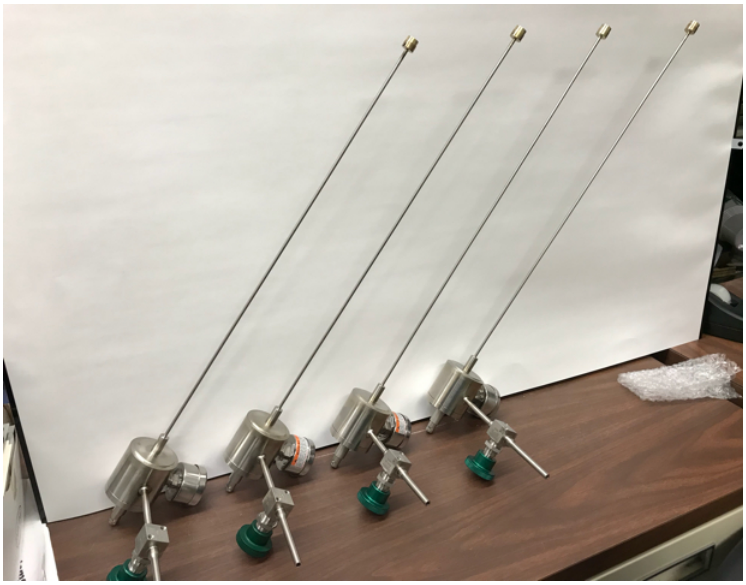


Figure 10: Four Mark II Fission Chambers ready for reference foils.

Figure 11: Mark II Fission Chamber loaded with an HEU reference foil.

In summer of 2017 we sacrificed all of the remaining HEU and DU reference foils to conduct the mass calibration that is shown in Figure 6. For this reason, we had to prepare a new set of HEU reference foils to test the design modifications. The new set of reference foils were prepared by electrodeposition from isopropanol, the same technique used in 2017. Since the modifications should not have affected the detector performance we were fully expecting to observe results similar to what we saw with the Mark I. However, given the time and resources to conduct a fission product yield measurement at NCERC we have always agreed that it is important to test any new detector, even of a previously proven design, on the Flattop assembly prior to a production run.

The week of February 12, 2018 we tested two HEU fission chambers and one blank fission chamber (identical to the other fission chambers except for the lack of actinide material) on the Flattop assembly. As we did the previous year we started at low reactor power and slowly ramped the power level up. Unfortunately, our expectations for the detector performance were not borne out. From the start there was evidence of sever energy degradation in the fission product signals. This was not observed in the test or production runs the previous year. The degradation is clear by comparing the pulse-height spectrum taken from FC HEU-01 (HEU reference foil) in Figure 12 with that from the April 2017 production run in Figure 8. After varying the few parameters available with such a simple detector, checking each of the signal processing modules for failure, and much discussion, we determined that this was most likely due to the reference foil. We loaded the second HEU fission chamber (HEU-02) into the Flattop assembly on February 15. Aside from the reference foil mass the run conditions were nearly identical to the first test. Again, we observed similar degradation of the fission product signals, although less severe, as shown in Figure 13.

Note that we used a Berkeley Nucleonics model PB-5 research pulser during each run of the 2018 tests and the 2017 production run to provide an independent measure of the dead-time of the entire detection system. The broadening of the pulser signal is due to accidental summing with gamma rays from the oralloy fuel of the Flattop assembly. These are independently evident at the lowest energies in the plot. However, the clear separation of the pulser signal from the rest of the spectral features means that it is still a valid measure of the quantity of interest.

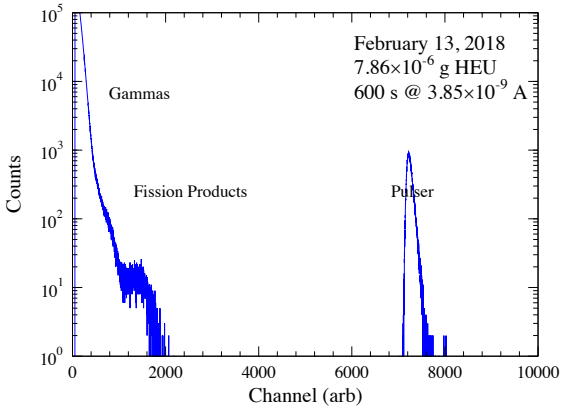


Figure 12: Pulse-height spectrum from fission chamber HEU-01, February 13, 2018. The signal sources, gamma rays from the assembly fuel, fission products and the research pulser, are indicated in the plot.

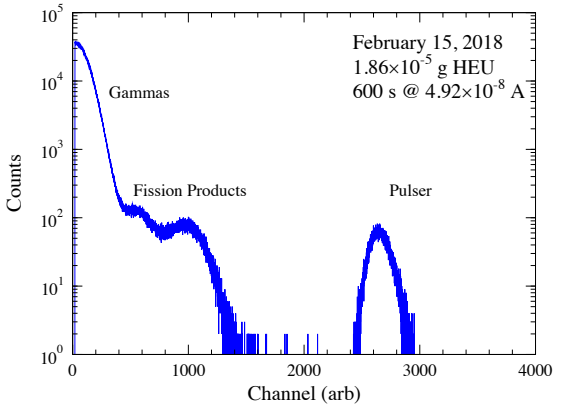


Figure 13: Pulse-height spectrum from fission chamber HEU-02, February 15, 2018. The signal sources, gamma rays from the assembly fuel, fission products and the research pulser, are indicated in the plot.

Upon our return to LANL we used a 2π Frisch-grid ionization chamber that we recently refurbished to take a closer look at the alpha energy spectrum coming off the foils. An example alpha spectrum is provided in Figure 14. Peaks for the ^{234}U and ^{235}U present in the HEU stand

out prominently as expected. However, there are significant alpha counts below the uranium peaks down to the lowest energies. These are indicative of energy degradation of some fraction of the alphas leaving the surface of the uranium deposit. There is a total of 2515 alpha counts between 0.550 and 4.275 MeV, or 5.9% of the total counts in the spectrum. There is also evidence of some low energy tailing on the two uranium peaks. While this does not seem to be significant we expect to see a considerably greater effect with fission products due to the Z dependence of the stopping power. This is an area that will be explored in great detail as part of the next phase of this work. In the meantime, we embarked upon an effort to improve the reference foils by testing alternative electrodeposition methods. The local literature indicated that the preferred methods included ammonium sulfate ($(NH_4)_2SO_4$) and ammonium chloride (NH_4Cl). The literature also indicated a preference for platinum as a substrate. Therefore, we prepared sets of HEU and ^{237}Np reference foils using these two methods on both platinum and our standard titanium backing. We then analyzed each by alpha spectroscopy, Frisch-grid and gross alpha counting. Each looked similar to reference foil 18HEU10.7F, shown in Figure 14: Alpha energy spectrum for HEU reference foil 18HEU10.7F. As expected, spectra for the neptunium foils were further complicated by the β's from decay of the ^{237}Np, as shown in Figure 15.

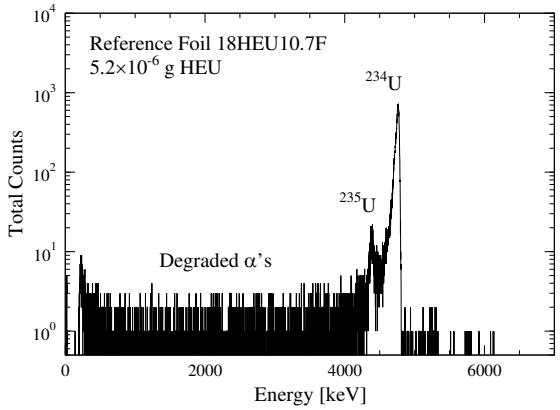


Figure 14: Alpha energy spectrum for HEU reference foil 18HEU10.7F.

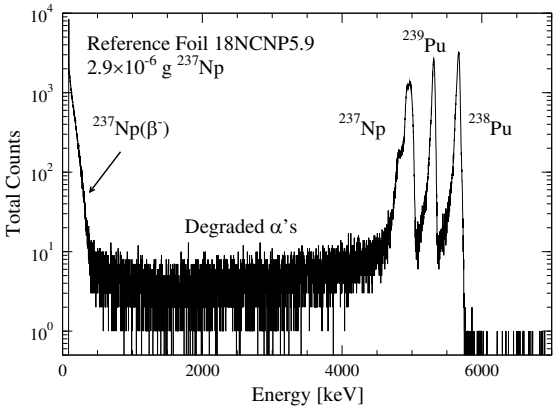


Figure 15: Alpha energy spectrum for ^{237}Np reference foil 18NCNP5.9.

We executed the R-value production run the week of April 23rd. returned to NCERC the week of March 26th.

The FY18 production run was executed the week of April 23rd. The original actinide of interest was ^{239}Pu, but concerns about sample handling and containment prevented us from following that path. Addressing the issues with ^{239}Pu are a priority in the first year of the follow-on effort, and we elected to run ^{237}Np in FY18 as a fall back. We were able to determine suboptimal, but workable settings during our test run the week of March 26th. This allowed us to collect usable

data with the fission chamber, shown in Figure 16, but it will take time to deconvolve the pulse height spectrum, and to correct for the Pu content in the ^{237}Np material. Results of the production run will be covered in a separate report.

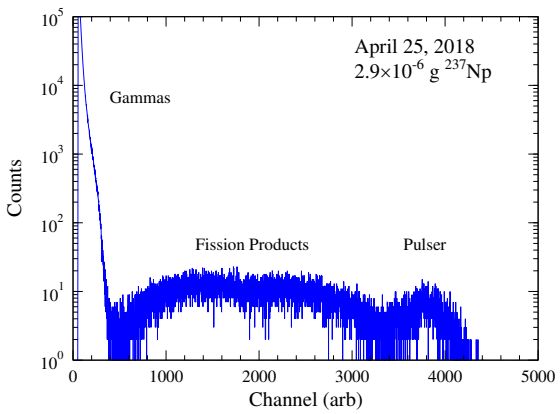


Figure 16: Pulse-height spectrum from fission chamber NP-01, April 23-24, 2018. The signal sources, gamma rays from the assembly fuel, fission products and the research pulser, are indicated in the plot.

Future Activities

A multi-laboratory proposal was submitted to the Nuclear Data Interagency Working Group / Research Program (LAB 18-1903) funding call that includes a continuation of the current efforts. In it we have outlined the following activities to complete development of the fission chamber during the first year of the project.

1. Conduct more rigorous R&D on electro-deposition methods for uranium, plutonium and californium fission reference foils on various substrates. There is very little information in the literature on this. Test deposits will be analyzed by several methods to determine actinide mass and energy loss of α-particles. ^{252}Cf reference foils will be used to accurately determine the energy loss for fission products relative to α-particles. We will select the best method and prepare a set of reference foils for each actinide and analyze for consistency. We will also prepare DU and HEU reference foils by vacuum vapor deposition of UF_4 and compare with the electrodeposited foils. We currently have approval to prepare vapor deposited foils of thorium and depleted uranium. Over the next several months we will be working on approvals for highly enriched uranium (>90% ^{235}U). We are working on a plutonium vapor deposition capability, but that will not be available for some time. Development of the vacuum vapor deposition capability is currently supported by defense programs.
2. Fabricate and assemble two fission chambers using the existing design modified to be able to swap out reference foils. This will allow us to perform bench-top (static) tests of

both fission chambers with ^{252}Cf reference foils to determine baseline performance, then perform tests of both chambers with ^{235}U reference foils using a high activity AmBe neutron source. In year two or three we will start to test ^{239}Pu reference foils using the AmBe source.