

Development and optimization of the micro-pocket fission detector

by

Daniel Marc Nichols

B.S., Kansas State University, 2016

AN ABSTRACT OF A DISSERTATION

submitted in partial fulfillment of the requirements for the degree

DOCTOR OF PHILOSOPHY

Alan Levin Department of Mechanical and Nuclear Engineering  
Carl R. Ice College of Engineering

KANSAS STATE UNIVERSITY  
Manhattan, Kansas

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# ABSTRACT

Recent developments in advanced reactor technologies such as small modular reactors and the growing interest in microreactor technology has emphasized the need for increasing the capability of in-core neutron monitoring sensors. Next generation reactor concepts have decreased cost and complexity, while also decreasing the reactor core form factor. With these small-sized reactor cores, there is a need for smaller form-factor, and reliable sensors. Micro-Pocket Fission Detectors (MPFD) have been under development for the past decade, with the capability to detect local neutron flux within a spatially discrete pocket. Recent advancements to the technology have focused on improving the manufacturability and design of the device and to demonstrate unique use-cases for the technology. Extensive simulations were conducted to optimize detector chamber characteristics and to determine construction materials, operating characteristics and expected response for various reactor environments. New manufacturing techniques were developed to improve the repeatability of the design and allow for mass-production. During the research presented herein, MPFDs were successfully operated at a new maximum power level of 13 GW<sub>th</sub> at TREAT, tracking neutron flux from  $10^{14} - 10^{17}$  n cm<sup>-2</sup> s<sup>-1</sup>. A set of 24 MPFD nodes were dispersed throughout the UWNR core providing real-time spatially-discrete three-dimensional neutron flux monitoring. The average detection efficiency for array 3 was determined to be  $(22.1 \pm 1.64) \times 10^{-12}$  cps/nv for pulse-mode operations, and  $(6.89 \pm 0.25) \times 10^{-21}$  A/nv for current-mode operations, constituting a 0.129% and 19.30% detection efficiency for pulse and current mode respectively. During tests at UWNR, MPFDs were utilized to produce a rich dataset which will be used to validate code as part of the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program.

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# ABSTRACT

Recent developments in advanced reactor technologies such as small modular reactors and the growing interest in microreactor technology has emphasized the need for increasing the capability of in-core neutron monitoring sensors. Next generation reactor concepts have decreased cost and complexity, while also decreasing the reactor core form factor. With these small-sized reactor cores, there is a need for smaller form-factor, and reliable sensors. Micro-Pocket Fission Detectors (MPFD) have been under development for the past decade, with the capability to detect local neutron flux within a spatially discrete pocket. Recent advancements to the technology have focused on improving the manufacturability and design of the device and to demonstrate unique use-cases for the technology. Extensive simulations were conducted to optimize detector chamber characteristics and to determine construction materials, operating characteristics and expected response for various reactor environments. New manufacturing techniques were developed to improve the repeatability of the design and allow for mass-production. During the research presented herein, MPFDs were successfully operated at a new maximum power level of 13 GW<sub>th</sub> at TREAT, tracking neutron flux from  $10^{14} - 10^{17}$  n cm<sup>-2</sup> s<sup>-1</sup>. A set of 24 MPFD nodes were dispersed throughout the UWNR core providing real-time spatially-discrete three-dimensional neutron flux monitoring. The average detection efficiency for array 3 was determined to be  $(22.1 \pm 1.64) \times 10^{-12}$  cps/nv for pulse-mode operations, and  $(6.89 \pm 0.25) \times 10^{-21}$  A/nv for current-mode operations, constituting a 0.129% and 19.30% detection efficiency for pulse and current mode respectively. During tests at UWNR, MPFDs were utilized to produce a rich dataset which will be used to validate code as part of the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program.

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## LIST OF ACRONYMS

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|      |                                                                |
|------|----------------------------------------------------------------|
| ASA  | Acrylonitrile Styrene Acrylate                                 |
| ATF  | Accident Tolerant Fuel                                         |
| ATR  | Advanced Test Reactor Facility                                 |
| ATRC | Advanced Test Reactor Critical Facility                        |
| BOE  | Buffered Oxide Etch                                            |
| BTU  | British Thermal Unit                                           |
| BWR  | Boiling Water Reactor                                          |
| CAD  | Computer-aided Design                                          |
| CEA  | French Alternative Energies and Atomic Energy Commission       |
| CNC  | Computer Numerical Control                                     |
| CTE  | Coefficient of Thermal Expansion                               |
| DI   | Deionized Water                                                |
| DOE  | U.S. Department of Energy                                      |
| DRT  | Dynamic Range Test                                             |
| DU   | Depleted Uranium                                               |
| ED   | Electrodeposition                                              |
| FWHM | Full-Width Half Maximum                                        |
| HEU  | Highly Enriched Uranium                                        |
| HTGR | High Temperature Gas-cooled Reactor                            |
| HTTL | High Temperature Test Laboratory                               |
| INL  | Idaho National Laboratory                                      |
| ISA  | International Standard Atmosphere                              |
| KSU  | Kansas State University                                        |
| LEU  | Low-Enrichment Uranium                                         |
| LDRD | Laboratory-Directed Research & Development                     |
| LFTR | Lithium Fluoride Thorium Reactor                               |
| LLD  | Lower-Level Discriminator                                      |
| MCNP | Monte Carlo N-Particle                                         |
| MFC  | Miniature-Fission Chamber                                      |
| MFP  | Mean Free Path                                                 |
| MITR | Massachusetts Institute of Technology Nuclear Research Reactor |

|          |                                                                  |
|----------|------------------------------------------------------------------|
| MPFD     | Micro-Pocket Fission Detector                                    |
| MSV      | Mean Square Voltage                                              |
| NAA      | Neutron Activation Analysis                                      |
| NASA     | National Aeronautics and Space Administration                    |
| NE       | Office of Nuclear Energy                                         |
| NEAMS    | Nuclear Energy Advanced Modeling and Simulation                  |
| NEUP     | Nuclear Energy University Program                                |
| NIM      | Nuclear Instrument Module                                        |
| OSCC     | Outer Shim Control Cylinder                                      |
| PEEK     | Polyether Ether Ketone                                           |
| PHS      | Pulse Height Spectrum                                            |
| PMMA     | Polymethyl Methacrylate                                          |
| PTFE     | Polytetrafluoroethylene                                          |
| QA       | Quality Assurance                                                |
| RTD      | Resistance Temperature Detector                                  |
| SEM      | Scanning Electron Microscope                                     |
| SFR      | Sodium-cooled Fast Reactor                                       |
| SMARTlab | Semiconductor Materials and Radiological Technologies Laboratory |
| SMFC     | Sub-Miniature Fission Chamber                                    |
| SMR      | Small Modular Reactor                                            |
| SPND     | Self-Powered Neutron Detector                                    |
| SPGD     | Self-Powered Gamma-ray Detector                                  |
| SRIM     | Stopping Range in Matter                                         |
| STP      | Standard Temperature and Pressure                                |
| TLD      | Thermoluminescent Dosimeter                                      |
| TREAT    | Transient Reactor Test Facility                                  |
| TRIGA    | Training, Research, Isotopes, General Atomics                    |
| TRL      | Technology Readiness Level                                       |
| UV       | Ultraviolet                                                      |
| UWNR     | University of Wisconsin-Madison Nuclear Reactor                  |
| VLSI     | Very Large-Scale Integration                                     |
| XRF      | X-Ray Fluorescence                                               |

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---

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## DEDICATION

---

I dedicate the work presented herein to GOD from which I have been given the strength to persevere through the tasks laid before me. I dedicate this work to my wife Karis, who has been a firm advocate for my success and has seen me through the trials of life. I dedicate this to my parents, Lanson and Celia, who encouraged me to continue to strive toward excellence and have sacrificed so much for my success. Finally, I dedicate this work to my brother Benjamin, who has always supported me and continues to be my strongest champion to great ambitions in life.

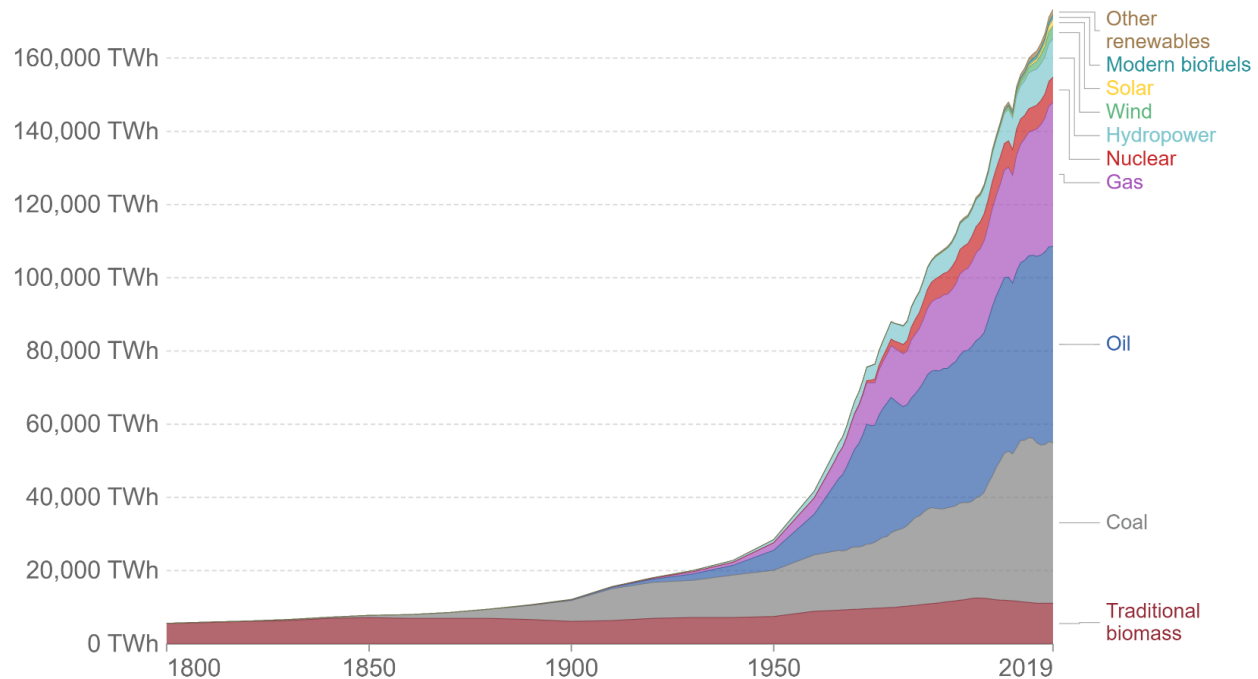
This chapter focuses on presenting the motivations and context to pursue this research. Presented herein this dissertation includes a description of the contributions to the science of reactor instrumentation and the importance of these developments. Additionally, an organization of the topics included in this dissertation is provided.

## **1.1 Motivation for Research**

Nuclear power has played a vital role in power production across the global, spanning from the 1950's until present day [1]. As of 2019, approximately 5% of the total global power consumption (approximately 610 quadrillion BTUs) is supplied through the use of nuclear reactors, and an additional 300 quadrillion BTUs (+50%) is estimated to be needed within the next three decades [2] as shown in Fig. 1-1. Increasing global power supply will require the adoption of Gen IV reactors, Small Modular Reactors (SMR), and microreactors. To support these up-and-coming technologies, reactor instrumentation must be advanced to keep pace, both to support material experimentation and power production. Additionally, with increased interest and attention to developing commercial space travel and extraterrestrial exploration and colonization, nuclear reactors have been determined to be the only viable long-term solution to fulfill power production and versatility requirements [3].

## Global primary energy consumption by source

Primary energy is calculated based on the 'substitution method' which takes account of the inefficiencies in fossil fuel production by converting non-fossil energy into the energy inputs required if they had the same conversion losses as fossil fuels.



Source: Vaclav Smil (2017) & BP Statistical Review of World Energy

OurWorldInData.org/energy • CC BY

*Fig. 1-1. Plot of historical global energy consumption by production method [4].*

To facilitate many of these needs, the nuclear power industry has shifted its design focus to concentrate on compact designs with different temperature, pressure, and radiation environments. These operational constraints require a fundamental shift in reactor instrumentation design to comply with the new requires. A prominent new objective of the nuclear power community is to provide a method of autonomous reactor operation without the need of human intervention [5]. Radiation detector technologies that are compatible with these requirements will need to be able to tolerate higher temperature, pressures, and corrosive materials while providing accurate real-time monitoring capabilities and packaged in a small form factor. Several competing technologies already exist which would satisfy the requirements of the next generation: Miniature and Sub-Miniature Fission Chambers (MFC & SMFC) under development in France, Self-Powered Neutron Detectors (SPND) widely developed around the world, and Micro-Pocket Fission Detectors (MPFD) developed in the United States.

Each of these active neutron monitoring sensors provide valuable in-core detection capabilities. What makes MPFDs unique are their spatially-discrete sensitivity and manufacturing simplicity. The neutron reactive material used in MPFDs are on the order of 1  $\mu\text{g}$  or less of fissile material and with a detection volume less than  $4.9 \times 10^{-5} \text{ mm}^3$ . This small volume allows the MPFD to be accurately approximated as a point detector. MPFDs also have the unique capability of multi-node packaging, which allows for a string of sensitive nodes dispersed along a length and housed in a single detection envelope. Current electronics allow for simultaneous monitoring and processing of real-time data for up to 8 nodes. These characteristics make the MPFD an attractive and viable option for use in current reactors, and future SMR and microreactor designs.

## 1.2 Contributions to the Science

Prior MPFD research focused on taking the MPFD platform from benchtop testing to in-core deployments. Customized electronics were designed and implemented into the MPFD measurement system, and testing was performed to characterize preliminary response to various neutron flux conditions. Additionally, previous MPFD research successfully developed a method of coating uranium and thorium fissile deposits for use in the manufacturing of detectors [6]–[9]. The research presented herein built off the prior accomplishments by wholistically analyzing the design of the detector (fissile coating, chamber shape, electric field, components for manufacturing, etc.) to develop and optimize the design of the MPFD. These developments were inter-woven with periodic experimental testing that informed researchers about potential drawbacks of the design. During this research, the MPFD platform has now been experimentally tested in a larger variety of in-core conditions compare to prior research. Testing such as reactor pulsing demonstrated the fast response of the detectors, whereas steady state flux tilting performed at UWNR demonstrated the ability of the platform to measure localized perturbations in neutron flux. Moreover, high-power pulse testing at TREAT, demonstrated the ability for the detector platform to survive and successfully track extreme neutron flux environments ( $10^{17} \text{ n cm}^{-2} \text{ s}^{-1}$ ). The development of the technology is not complete and further research must be performed to commercialize the technology, but this research has provided several contributions to the science of the MPFD technology that will be mentioned.

### **1.2.1 Maximum Power Level Testing**

Two MPFDs were tested, one with natural enrichment uranium and one with natural enrichment thorium, at a new maximum power level of approximately 13.7 GW<sub>th</sub>, which occurred at the Transient Reactor Testing (TREAT) facility during summer 2018. MPFD nodes tracked the 13.7 GW<sub>th</sub> reactor pulse which had a FWHM of 115.8 ms. The uranium and thorium nodes respectively measured the reactor pulse to have a FWHM of 128.5 ms and 130.2 ms [10]. During this reactor transient, MPFDs experienced a maximum neutron flux estimated to be approximately  $10^{17}$  n cm<sup>-2</sup> s<sup>-1</sup>. These reactor pulse transients were part of a larger in-core testing campaign at TREAT. As testing continued, the MPFD nodes showed signs of detector performance degradation, but continued operating for several other high-power pulses ranging between 1 - 13 GW<sub>th</sub>. During each of these high-power tests, the MPFDs were able to track reactor power level across approximately 3.5 decades of power.

### **1.2.2 Three-Dimensional Core Mapping**

A set of 24 MPFD nodes were dispersed throughout the University of Wisconsin-Madison Nuclear Reactor (UWNR) core providing real-time spatially-discrete three-dimensional neutron flux monitoring. During tests at UWNR, MPFDs were utilized to produce a rich dataset which will be used to validate code as part of the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program. Experimental tests included steady state operations with flux tilting, and transients such as square wave, ramp, and pulsed reactivity insertions.

### **1.2.3 Fast Transient Tracking**

Reactor pulsing experiments were conducted at 3 separate research reactors (UWNR, KSU and TREAT). For each deployment, the measured FWHM of the MPFD closely tracked reactor pulse transients response within  $\pm 15\%$ . During these tests, MPFD achieved successful tracking resolution of 0.25 ms. Each test provided unique outcomes for the research. Tests at KSU demonstrated the first successful tracking of two pulse transients (\$2.50 and \$2.77 reactivity) with 11.46% and 8.61% deviation from Fuchs-Nordheim predicted FWHM and gave experimental insight into necessary operating characteristics for future tests. The UWNR tests signified the first experiment during which MPFD nodes were placed in a three-dimensional set to measure a reactor pulse. Previously, simultaneously deployed MPFD nodes had only successfully measured axial core locations. Finally, testing at TREAT allowed for the study of node response degradation and

facilitated the opportunity to push the detectors to the highest flux recording to date. Over the course of the seven high power TREAT tests, ranging from 1 – 13 GW<sub>th</sub>, it was observed that MPFD response reduced by 26.6% for a MPFD node with a uranium fissile layer and 29.9% for a MPFD node with a thorium fissile layer.

#### **1.2.4 Electric Field Optimization**

During detector development for the UWNR experiments, electric field simulations were conducted, and the diamond chamber geometry was confirmed to have a detrimental electric field orientation that precludes its use in multi-node MPFD arrays. This discovery led to the adoption of a new chamber geometry (clover design) that allows for more successful multi-node operation in a common-cathode geometry. This new design also optimizes charge carrier trajectories inside the detector chamber while migrating in the electric field.

#### **1.2.5 Device Calibrations**

Extensive calibration efforts were conducted as part of the experiment campaign at UWNR. Calibration measurements were conducted in a single reference location using a variety of activation foils. This calibration procedure represents the first attempt to experimentally correlate MPFD response to the local neutron energy spectrum. Detection efficiencies were determined for MPFD array 3 to approximately be  $(22.1 \pm 1.64) \times 10^{-12}$  cps/nv for pulse-mode operations, and  $(6.89 \pm 0.25) \times 10^{-21}$  A/nv for current-mode operations, constituting a 0.129% and 22.98% detection efficiency for pulse and current mode respectively.

#### **1.2.6 Instrument Deployment Design**

For the upcoming Laboratory-Directed Research & Development (LDRD) experiment at ATRC, extensive MCNP simulations were conducted to determine the fission rate and fission fragment deposition spectra in the MPFD for the given deployment location (B-8 Position). These simulations were used as the basis when selecting the fissile mass for use in the MPFD. Additionally, simulations were used to estimate non-neutron signal impact from gamma rays, alpha particles, and beta particles. These values were compared against fission fragment energy deposition estimates and were used to build an estimated energy deposition spectrum.

### **1.2.7 Improved Detector Fabrication Techniques**

A very-large scale integration (VLSI) technique known as photolithography was used to produce high precision Ti/Pt electrode contacts for use in the electrodeposition process. This technique has also improved the manufacturing repeatability of the detectors, and will significantly reduce the cost per piece, opening the door for mass-production of the devices. Previous methods had limited production to between 48 and 256 disks per evaporation, in comparison to the new photolithography method which can yield an estimated 600 disks per evaporation with low feature variance; 0.005 mm standard deviation for a feature diameter of 0.756 mm.

## **1.3 Organization of the Dissertation**

In Chapter 2, the discussion focuses on the history of nuclear discoveries and radiation detectors, fundamental radiation particles, nuclear reactor in-core conditions, neutron interactions with matter and alternative neutron flux monitoring technologies. Chapter 3 focuses on device design characteristics including detector operating principles, electric field optimization, and estimates of radiation sensitivity. Additional details are given regarding construction material selection, geometric considerations, and finally, fissile coating selection. Chapter 4 focuses on device manufacturing and assembly, detailing the various designs that have been produced during the progression of this research and modifications to certain construction processes. The chapter concludes with remarks about post-assembly reliability testing, and commentary regarding practical approaches for detector deployment. Chapter 5 describes MFPD auxiliary systems and testing including calibration efforts and physical hardware. In-core testing conducted at various reactors is presented in Chapter 6 including steady state, transient and calibration experiments. These experiments are broken up into different sections dedicated to the four different reactor deployments mentioned: the Kansas State University TRIGA Mark II Nuclear Research Reactor (KSU), the University of Wisconsin-Madison Nuclear Reactor (UWNR), the Transient Reactor Testing (TREAT) facility, and the Advanced Test Reactor Critical Facility (ATRC). Finally, Chapter 7 summarizes the overall body of work present herein. Included in Chapter 7 is additional commentary on the overall progression of the research, a formalization of the contributions to the science, and a proposal of future work on the MPFD technology.

This chapter focuses on the background information such as the history of radiation, characteristics of fundamental particles, reactor environment conditions and considerations, neutron reactive materials and applications, and finally, neutron flux monitoring technologies.

### 2.1 Introduction to Radiation

While the existence of atoms had been theorized by Greek philosophers around 400 BC, the discovery and observation of atoms would not be made until the 20<sup>th</sup> century. These discoveries ultimately led to many key advancements in technology, medicine, and humanity's understanding of how the universe works. In this chapter an abbreviated history of the key discoveries in nuclear physics and engineering are discussed. Additionally, background information about different radioactive particles, reactor characteristics, and neutron detection methods are presented. The chapter concludes with a brief description of alternative neutron-flux monitoring technologies presently available.

#### 2.1.1 History of Nuclear Discoveries

The study of radioactivity and the particles emitted by radioactive material began in 1895 with the observation of cathode rays by a German engineer, Wilhelm Conrad Röntgen. He observed penetrating rays during the operation of a Crookes tube; with these rays causing fluorescence of a nearby plate coated with barium-platinocyanide. During the years that followed, the public referred to these rays as “Röntgen rays”, but Röntgen preferred and eventually named them “x rays”[11]. This was the first discovery of radiation and its interaction with materials [12].

In 1896, a French engineer Antoine Henri Becquerel, was experimenting with photographic plates and various rocks from his father's geology collection. He serendipitously discovered that some of the samples were composed of uranium and potassium caused an image to form on the photographic plates after development without being exposed to direct sunlight. After subsequent experimentation he deduced that the photographic plates were being exposed by some unknown emission coming from the material inside the rock samples. He noted that rocks with higher uranium content caused a denser image on the developed photographic plates. This emission was first termed as “Becquerel Rays” but is now understood to be a conglomeration of alpha particles,

beta particles, x rays and gamma rays. Later in 1903, Becquerel, his doctoral student Marie Skłodowska Curie, and her French physicist husband Pierre Curie were awarded the third Nobel Prize in Physics for their work in the discovery of spontaneous radioactivity [12].

While these discoveries started the radioactive era, the mechanisms for why these emissions occur and what comprises the fundamental building blocks of the atom had not yet been determined. In 1897 a British physicist, Joseph John Thomson, experimented with evacuated cathode ray tubes and deduced that particles with a negative charge were emitted from the cathode surface during operation. These particles were eventually named “electrons” as suggested by an Irish physicist George Johnston Stoney due to the particle being responsible for the fundamental unit quantity of electricity. It is important to note that Stoney actually suggested the name to be “electrines” rather than the presently used form “electrons” [13].

Later in 1899, a former student of Thomson, Ernest Rutherford, used a gold foil electrometer to understand the particles being emitted by a uranium ore sample. Through his experiment he was able to determine that two types of radiation, which he named “alpha rays” and “beta rays”, were emitted from the sample and required different thicknesses of aluminum foil to attenuate (see Section 2.2.4 for additional information). During subsequent experimentation, Rutherford also demonstrated that alpha rays and beta rays could be deflected in a magnetic field, although the alpha rays were deflected to a lesser degree in comparison to beta rays. These discoveries earned Rutherford the 1908 Nobel Prize in Chemistry. Further, his experiments showed alpha rays and beta rays had opposite electrical charge. We now know these emissions to be alpha particles ( $^4\text{He}$  nuclei) and beta particles (electrons emitted from the atomic nucleus). Upon examination of the experimental results, Rutherford noted that there appeared to be particles that did not seem to deflect in the magnetic field, but he assumed that these must be highly energetic beta particles. It wasn’t until 1900, that a French physicist and chemist, Paul Ulrich Villard, repeated Rutherford’s experiment using radium samples and discovered that indeed there was a third ray being emitted by the sample. Although Villard discovered this emission, in 1903 Rutherford named this radiation “gamma rays”. Later in 1918, an American physicist, Arthur Holly Compton, demonstrated that gamma rays lose energy after scattering off an electron. This phenomenon earned Compton the 1927 Nobel Prize in Physics and is referred to as Compton Scattering (see Section 2.2.2 for further discussion).

In 1932, one of Rutherford's students, James Chadwick, was conducting experiments with alpha-particle emitting materials such as polonium and radium. He coupled the alpha-particle sources with various low mass elements such as boron and beryllium and identified that a neutral particle was being emitted when an alpha particle, emitted by the polonium or radium, struck boron and beryllium targets [14]. A similar experiment had previously been performed by Marie and Pierre Curie's daughter, Irene Curie-Joliot, and her husband Jean Frédéric Joliot-Curie, but the conclusion was limited to only identifying the amount of energy these particles possessed. Chadwick modified the experiment by using a gas-filled detector with nitrogen and hydrogen gas near the low mass materials to measure the energy of the neutral particle via recoil protons. Through this process, he was able to determine the mass of the neutral particle to be like the mass of a proton. Given that the particle has mass like a proton, but having a neutral relative charge, Chadwick named this particle the neutron. He received the 1935 Nobel Prize in Physics for his discovery.

After the discovery of the neutron, scientists began exploring neutron interactions with different elements. In 1934, Italian physicist Enrico Fermi, observed neutron-induced transmutation of a variety of elements such as phosphorous, iron, silicon, aluminum, and chlorine [15]. From these experiments, Fermi concluded that elements exposed to neutron radiation converted (as known as transmuted) into radioactive isotopes which often decayed via beta-particle emission but would sometimes change to a different element via proton emission. In 1936, German chemists Otto Hahn and Fritz Straßmann, along with Austrian-Swedish physicist Lise Meitner discovered that uranium bombarded by neutrons produced lighter elements. This discovery was unexpected, but the results indicated that sufficiently heavy elements like uranium can be fractured into much lighter elements such as barium. Meitner observed that the sum of the product atomic masses was not equivalent to the starting mass and therefore concluded that energy was released because of the uranium nuclear fracturing (see Section 2.3 for further discussion of energy release). As part of this discovery, Lise Meitner is credited with coining the term nuclear fission [16].

Between 1932 and 1939, scientists had speculated that energy could be harvested from neutron induced transmutation of elements, but these methods would require a sustained neutron source. Enrico Fermi, Leo Szilard (a Hungarian-American physicist and one of Albert Einstein's students), and Jean Frédéric Joliot-Curie became aware that the splitting of uranium would yield both energy and additional neutrons via experimentation. During this time, Nazi Germany was

beginning to grow in military might, and Leo Szilard feared that the knowledge of sustained fission could be used as a weapon by the Nazis. He urged both Fermi and Joliot-Curie to forego mention of this discovery, thus hiding it from the world, to avoid Nazi interest in potential fission-based weapons. Fermi reluctantly complied with Szilard's request, but Joliot-Curie ignored, and in 1939, he published along with Russian physicist Lew Kowarski, and German physicist Hans von Halban discoveries pertaining to the release of energy and neutrons resulting from fission. Their discoveries included observations that neutron-induced fission of uranium yielded  $3.5 \pm 0.7$  neutrons, and they theorized that this amount could be utilized to produce exo-energetic transmutation chains [17]–[19], also known as a chain reaction. These observations eventually led to the development of nuclear weapons via the Manhattan Project in the United States, as well as the exploration of fission-based power generation. In 1941, Fermi in conjunction with Szilard, proposed a design for a fission reactor. On December 2<sup>nd</sup>, 1942, as part of the Manhattan Project, this design came to fruition when a Chicago team of scientists led by Fermi created a sustained chain reaction using a reactor pile built under the football stadium, Stagg Hill, on the University of Chicago campus [1], [20].

### **2.1.2 Brief History of Radiation Detectors**

In the early 1900s, while studying under the tutelage of Ernest Rutherford, Hans Geiger was set to the task of counting alpha-particle decay of a radiation source using a microscope and ZnS scintillation screen. A flash of light would be produced when alpha particles struck the scintillation screen. To accurately count individual particles, Geiger was required to stare into the microscope for long periods of time, counting the flashes; a task that proved to be quite tedious and exhausting. In an effort to develop a more efficient and less exerting method, Ernest Rutherford and Hans Geiger designed and built a gas-filled radiation detector in 1908, which counted radiation events by drifting the ionization gas in an electric field to produce a signal [21]. Alpha particles entering the chamber would ionize the gas, resulting in charge carriers that drifted toward the respective electrodes. The device consisted of a metallic cylinder with a thin metal wire in the center insulated by ebonite corks at both ends of the chamber. This device constituted the first gas-filled radiation detector. In 1928 Geiger and his student Walther Müller improved the device design which became commonly known as the Geiger-Müller counter.

Later in the 1930s various new forms of gas detectors utilizing neutron reactive materials such as boron-lined and  $\text{BF}_3$  were developed that resulted in higher detection efficiency for

neutrons [22], [23]. In 1940 Otto Robert Frisch developed a new type of gas detector, the gridded ionization chamber which provided a high accuracy method to measure the energy of incident radiation [24]. In the late 1940s, ion chambers were lined with fissile material, often in the form of  $U_3O_8$ , as a neutron reactive coating [25]. These new detectors are known as fission chambers and are commonly used in reactors as neutron flux monitors and are deployed both exterior to, and directly inside the reactor core.

## 2.2 Fundamental Radiation Particles

A common misconception about radiation often leads to confusion amongst the public. For the discussion of radiation and emission phenomena, the following sections will only focus on ionizing radiation or radioactively-emitted particles which are common in nuclear science and will exclude emission phenomena such as non-ionizing electromagnetic radiation or exotic forms of radiation, such as muons, bosons, various forms of neutrinos, etc.

### 2.2.1 Common Charged Particles (Alpha & Beta Particles)

As discussed in Section 2.1.1, alpha particles and beta particles were some the earliest discovered forms of radiation. Both forms of radiation are common decay products of unstable isotopes which are common in the natural environment. Alpha particles and beta particles are also found in abundance inside nuclear reactors as by-products of the fission process and subsequent decay of daughter isotopes.

#### Basic Characteristics

Alpha particles are one of the most stable subdivided configurations of a nucleus. As such, it is commonly emitted by high-Z radioactive isotopes to become stable. An alpha particle comprises two protons, two neutrons and no electrons, giving it a +2 relative charge and a rest mass of 4.001506 amu ( $3727.4 \text{ MeV c}^{-2}$ ). Due to the mass and charge of the alpha particle, it has short ranges in air and can be terminated by small thicknesses of low-Z materials (approximately 2-4 cm in air). The range of alpha particles can be determined as,

$$R(T) = 0.693T^2 + 3.544T + 0.58, \quad (1)$$

where the range,  $R$ , of the alpha particle in mm of air (STP), is a function of its kinetic energy ( $1 < T < 15$ ) measured in MeV. This equation is based on previous work with defined semi-empirical

formulae as well as through charged-particle simulations [26], [27]. The equation assumes a density of  $1.225 \text{ kg m}^{-3}$  for air at sea level ( $15^\circ\text{C}$ ) as determined by the International Standard Atmosphere (ISA) model from ISO 2533:1975.

Beta particles are first-generation leptons which have a relative charge of -1 or +1. The most common first-generation lepton is the negatively charged electron which constitutes matter. The electron antimatter counterpart is the positron which has a +1 relative charge. Both electrons and positrons have a rest mass of  $0.000549 \text{ amu}$  ( $0.511 \text{ MeV c}^{-2}$ ). Beta particles can penetrate further into matter than alpha particles due to a lower magnitude relative charge and mass. As an electron travels through matter, due to the extremely low mass in comparison to the surrounding medium, it will scatter at large angles off the electron clouds of the surrounding atoms, which results in an erratic path deviating by a large angle from the initial incident vector. The mean range of electrons within a material can be approximated with,

$$\rho R_P \left[ \frac{\text{g}}{\text{cm}^2} \right] = \begin{cases} 0.412E^{1.265-0.0954 \ln E} & 0.1 \text{ MeV} \leq E \leq 3 \text{ MeV} \\ .530E - 0.106 & 2.5 \text{ MeV} \leq E \leq 20 \text{ MeV} \end{cases} \quad (2)$$

where,  $E$ , is the energy of the incident electron, and  $\rho$ , is the density of the target material [28]. Note that this empirical approximation can be versatile as it can use modified to fit most materials given the material density. There are other charge particles such as deuterons, tritons, etc. which will not be discussed but can be found elsewhere.

### 2.2.2 Common Electromagnetic Particles (x rays and Gamma rays)

X rays and gamma rays are common by-products of nuclear decay and constitute a large portion of the radiation released during the operation of a nuclear reactor. The gamma-ray flux in a nuclear reactor is directly proportional to the fission rate inside the reactor core, and thus can be used as a method to monitor reactor power. Note that this proportionally only holds inside core and begins to deviate beyond the core boundary.

#### Basic Characteristics

X rays and gamma rays are forms of ionizing electromagnetic radiation, which are emitted by radioactive isotopes and are distinguished based upon the emission origin in the atom. Both x rays and gamma rays are massless and have neutral relative charge. X rays are short wavelength (between approximately  $10 \text{ pm}$  and  $10 \text{ nm}$ ), high energy photons that are emitted from the de-excitation of an electron. X rays are emitted natural when electrons transition orbital shells or

synthetically using x-ray generators and can span energy ranges between approximately 50 eV and 150 keV. Characteristic x-ray energies are unique to the electron orbital transition and element, while x-ray generator energies are a function of acceleration voltage.

Gamma rays typically have shorter wavelengths and higher energies than most x rays and are emitted during de-excitation of the nucleus. Gamma rays commonly span energies between approximately 10 keV to 20 MeV, with some uncommon gamma rays exceeding energies of GeV [29], [30]. Gamma-ray emission energy is based on excitation states of the atomic nucleus and are therefore characteristically unique to each isotope. Because of this isotope unique emission trait, gamma rays can be used to trace and identify quantities of specific isotopes. Known as gamma-ray spectroscopy, this technique is part of a forensic tracing method known as neutron activation analysis (NAA) and will be discussed further in Section 2.5.2. Because both x rays and gamma rays are neutral particles, they can only interact with matter via non-coulombic methods.

### **Interactions**

There are three primary methods through which photons interact with matter. Photoelectric effect is the process of a photon interacting with the entire atom, resulting in the ejection of a photoelectron from an orbital shell, typically the K shell. The vacancy left by the ejected photoelectron is then filled by an electron from an outer orbital resulting in the emission of a characteristic x ray or an Auger electron [1]. In another process known as Compton scattering, a photon can scatter off a free electron or an orbital electron, transferring some of its momentum to the electron. Due to the relative mass of the electron, and despite the zero mass of a photon, the photon will only transfer a fraction of its total energy, in the form of momentum, and the photon will be deflected off the electron. The final primary interaction is known as pair production, during which a photon with kinetic energy greater than two times the rest mass of an electron can interact in the strong nuclear field near the atomic nucleus causing the production of an electron-positron pair. The electron and positron are each imparted with half the total photon energy minus two times the rest mass of a first-generation lepton (electron). The pair will travel through the medium independently, in opposing directions, with the positron eventually annihilating with an electron to produce annihilation photons each with 511 keV of energy and directly opposing trajectories [1]. For comparison, all three interaction energy ranges are shown in Fig. 2-1 [31]. Note that photoelectric absorption is the primary method of energy transfer for photons below 100 keV for most elements. The Compton scattering interaction dominates energy transfer for photons between

100 keV and 1.5 MeV, above which pair production is the primary method of energy transfer [12], [26], [32]. These regions of primary interaction fluctuate depending on the target material and is graph, for comparison, between  $\text{Al}_2\text{O}_3$  and uranium in Fig. 2-1. Other interactions exist, such as Rayleigh scattering and triplet production, but will not be discussed in this work. Additional information about these interactions can be found in [12].

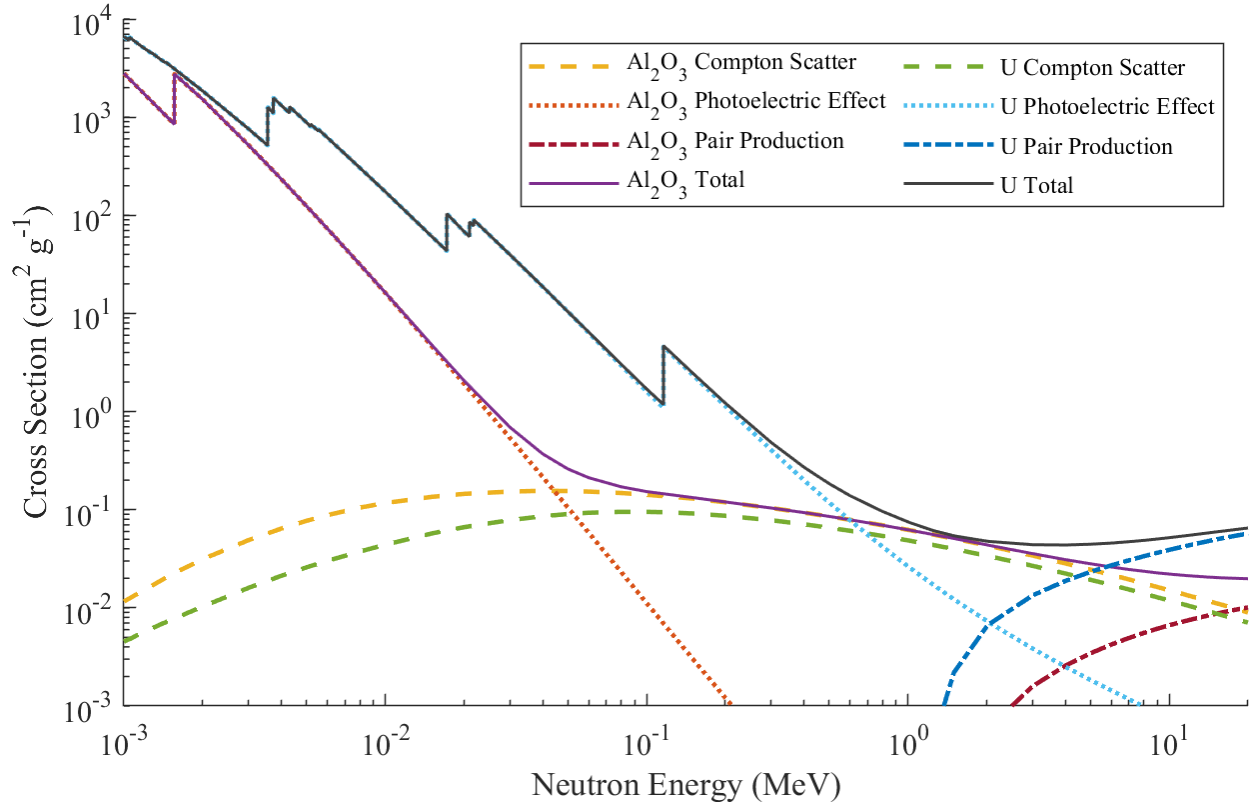


Fig. 2-1 Attenuation coefficients for gamma-ray interactions with alumina ( $\text{Al}_2\text{O}_3$ ) and uranium (U). Note that for aluminum, only the K shell peak is viewable, whereas for uranium, the M, L, and K shell are shown [31].

### 2.2.3 Neutrons

A neutron is one of several composite subatomic particles that exist and is a fundamental building block of positive matter. Neutrons are primarily found in the nucleus of an atom but can exist outside the atom, because of nuclear decay, for a short time duration. Nuclear reactors are one of a few examples of environments where neutrons exist in large populations.

### Basic Characteristics

Neutrons were first discovered in 1932 by James Chadwick to have a neutral relative charge and the approximate mass of a proton (see Section 2.1.1 for additional historical information). It was later determined that a neutron has a rest mass of 1.008665 amu (939.565 MeV  $c^{-2}$ ), which is 1.0014 times the rest mass of a proton [1]. A free neutron, not bound to an atomic nucleus, has a half-life of 10.2 minutes [1], [20], [33], [34]. Because of the weak force interaction in one of the down quarks, a  $W^-$  boson, with a rest mass of 86.312946 amu (80.379 GeV  $c^{-2}$ ), is emitted which promptly decays after  $3 \times 10^{-25}$  seconds into an electron ( $e^-$ ) and electron antineutrino ( $\bar{\nu}_e$ ), as shown in Fig. 2-2 [35]. These products have a continuum of possible emitted energies, with the electron average energy being 301.37 keV and an end-point energy of 782.347 keV (the Q value of the decay) [34].

### Interactions

Much like gamma rays, neutrons have neutral charge and do not ionize media as they traverse through matter. Therefore, neutrons do not lose energy to a medium unless it is scattered or absorbed by the atomic nuclei in the material. While neutrons and electrons can interact, the primary mechanism of neutron energy loss is a result of interactions with nuclei [36]. Neutron interactions, both scatter and absorption, are split into subcategories of interactions which identify the resulting products of an interaction event.

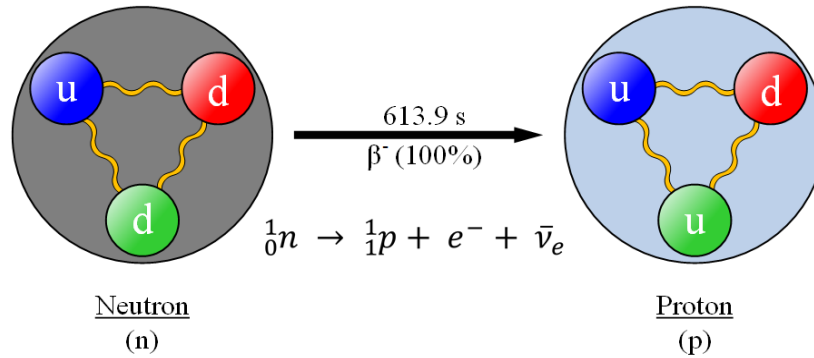


Fig. 2-2. Illustration showing internal quark conversion process. Note that either of the starting down quarks can convert into an up quark, and this interaction is not limited to a specific quark “color”.

The two main subcategories of scattering interaction; elastic scattering and inelastic scattering. For elastic scattering a neutron can potentially scatter, during which the neutron does not enter the nuclear envelope and form a compound nucleus, and instead reflects off the nucleus, and transfers a fraction of its kinetic energy in the form of momentum. The other form of elastic

scattering is known as elastic resonant scattering, during which the neutron is absorbed by the target nucleus, forming a compound nucleus, with subsequent emission of the incident neutron and the target nucleus is left at ground state. Much like in potential scattering, elastic resonant scattering transfers a fraction of its kinetic energy to the target nucleus in the form of momentum. Inelastic scattering occurs much like elastic resonant scattering, but after the neutron is emitted, a portion of the energy is converted and leaves the target nucleus in an excited state [12], [20]. There are also exotic scattering interactions such as nuclear spallation, which only occur at high neutron energies. Absorption interactions have four main subcategories including electromagnetic (when the nucleus absorbs a neutron and produces either a prompt or delayed gamma-ray emission), light-ion (absorption resulting in the emission of protons, deuterons, alpha particles, etc.), neutral absorption (absorption resulting in the emission of multiple neutrons), or fission (which results in the fracturing of the nucleus and accompanied by emissions of gamma rays, beta particles, and additional neutrons). These interactions are categorized and depicted in Fig. 2-3 and Fig. 2-4.

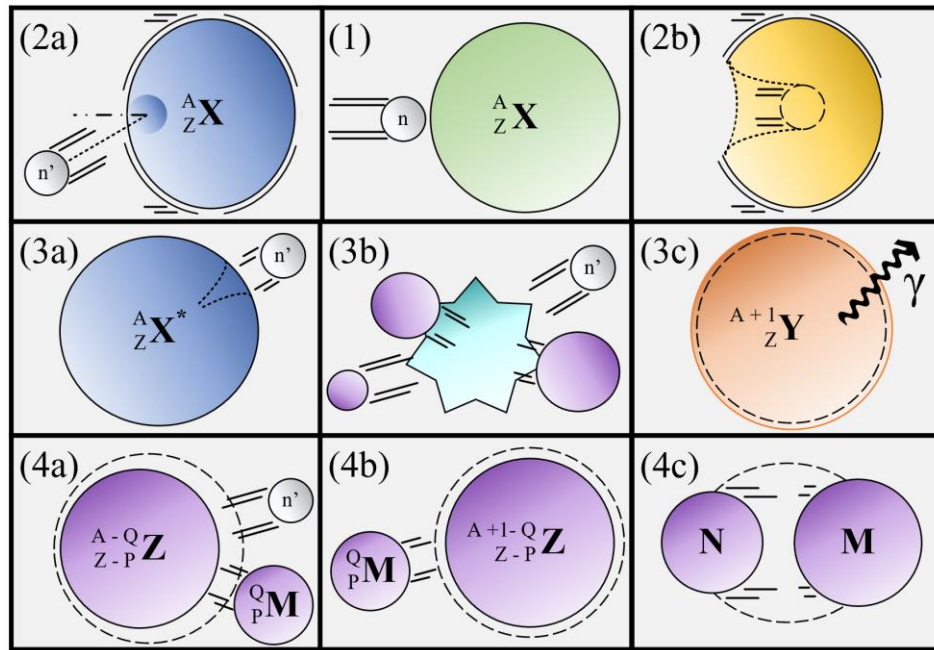


Fig. 2-3. Diagram depicting neutron interactions and consequences. (1) Neutron incident onto target nucleus leading to (2a) potential elastic scatter, or (2b) compound nucleus formation. (2b) compound nucleus formation can then result in (3a) neutron passing-through and losing energy either Resonant elastic or inelastic scatter, (3b) nucleus disintegration/spallation, (3c) excited compound nucleus with gamma-ray emission. If the neutron has sufficient energy, it may cleave nucleons from the atom (4a). An excited nucleus (3c) may emit a cluster of nucleons (4b) or undergo fission (4c).

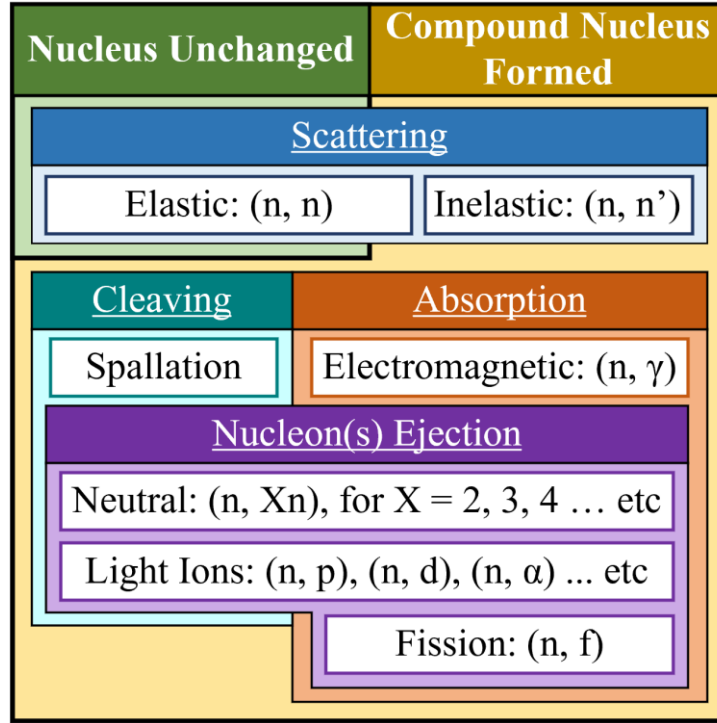


Fig. 2-4. Diagram depicting how various neutron interactions are categorized.

## Sources of Neutrons

Due to the inherent instability of unbound neutrons, they can only be found outside an atomic nucleus because of various decay schemes, cosmic or manmade production. As will be discussed in further detail in Section 2.3, neutrons produced by fission reactors or spontaneous fission, can span from 0 eV to 20 MeV. Fusion reactions such as D-D, and D-T, produce Gaussian neutron energies centered around 2.54 MeV and 14.2 MeV respectively. The fusion of T-T produces a continuum of energies between 0 eV and 9.3 MeV [37]. Binary sources including  $(\alpha, n)$  reactions such as AmBe, PuBe, and AmAl, and photoneutron  $(\gamma, n)$  reactions can produce neutron energies ranging from 0 eV to 12 MeV, and 0 eV to 1.5 MeV respectively [32]. For the work described herein, the primary source of neutrons used in experiments were from nuclear fission at various experimental reactors throughout the United States.

### 2.2.4 Attenuation

For neutrally charged radiation, such as gamma rays, x rays and neutrons, the primary method of energy loss occurs from collisions with the medium through which the radiation travels. If a neutral particle loses enough energy to the medium, it has a high probability of being absorbed.

The combination of scattering and absorption determine the number of particles lost within a given thickness of material. Observing the difference in the number of particles and their respective energy impinging upon a material and the subsequent reduction after traversing the material is known as attenuation. Attenuation is an exponential function dependent upon particle interaction probabilities in the material and the thickness of the material. Fig. 2-5 illustrates an ideal attenuation scenario and depicts an equation which is used for uncollided flux determinations.

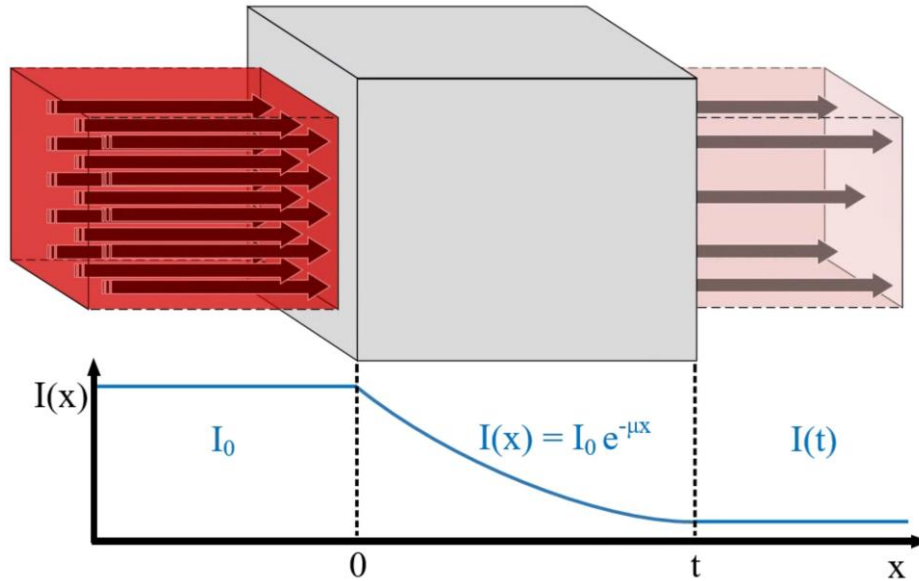


Fig. 2-5. Diagram and conceptual graph depicting uncollided flux attenuation of neutral particles through matter.

$I_0$ , also known as incident intensity, describes the incident flux impinging upon the material and  $I(x)$ , describes the uncollided flux as a function of depth into the material.  $x$  describes the depth into the material (typically in units of cm), and  $\mu$ , describes the linear attenuation coefficient for the material in reference to the incident particle type. For both gamma rays and neutrons,  $\mu$ , is given in  $\text{cm}^{-1}$ , which is the reciprocal of mean free path (MFP). The MFP represents the average distance traveled by a particle before an interaction has occurred. Tabulated values for both gamma rays and neutrons are widely available in previous works [31], [38].

### 2.2.5 Additional Particles

While other particles which contribute to various quantum mechanical phenomena exist, discussion will be left out of the work herein, but can be found in various other locations. A cursory glance at a few of these particles, such as W bosons and neutrinos, have been or will be mentioned

in chapters throughout this work, but due to the minor impact on the research contained herein, a more in-depth discussion of these auxiliary particles will not be given.

## 2.3 Nuclear Reactor In-Core Environment

A nuclear reactor produces electrical power by utilizing heat extracted from the fission of fuel. Fuel in nuclear reactors is primarily composed of uranium ceramic compounds and the primary isotope for energy production is  $^{235}\text{U}$  (although alternative fuel mixtures include plutonium, neptunium, and thorium isotopes). During the fission process fissile material absorbs a neutron causing excitation and fracturing. This process results in the release of 200 MeV of energy which can be deposited in the local medium in the form of heat. Most of the energy released is transferred to the environment via kinetic energy of the fission fragments, but a more comprehensive balance sheet is shown in Table 2-1. Additionally, the fission process releases 2-3 neutrons at high energies (see Section 2.2.3 for further discussion) which can then go on to induce fission in the surrounding fissile material. A critical reactor state (where reactor power remains steady), is achieved if at least one fission neutron causes another fission event, thus constituting a self-sustaining chain reaction.

Table 2-1. Energy released, and energy recovered because of  $^{235}\text{U}$  fission.

|                                           | Energy from<br>Fission (MeV) | Recoverable in<br>Core (MeV) |
|-------------------------------------------|------------------------------|------------------------------|
| <i>Prompt:</i>                            |                              |                              |
| kinetic energy of the fission fragments   | 168                          | 168                          |
| kinetic energy of prompt fission neutrons | 5                            | 5                            |
| fission $\gamma$ -rays                    | 7                            | 7                            |
| $\gamma$ -rays from neutron capture       | -                            | 3-9                          |
| <i>Delayed:</i>                           |                              |                              |
| Fission product $\beta$ -decay energy     | 8                            | 8                            |
| Fission product $\gamma$ -decay energy    | 7                            | 7                            |
| Neutrino kinetic energy                   | 12                           | 0                            |
| Total energy (MeV)                        | 207                          | 198-204                      |

Source: Data from Shultis & Faw 2008 [1]

The neutron population in the core is therefore coupled to the number of fissions that have occurred within a span of time. Because the neutron population represents the local fission rate of the reactor, it is highly advantageous to utilize local and spatially-discrete sensors to monitor neutron flux and therefore power.

Inside a nuclear reactor, radiation levels far exceed most natural radiation levels by several orders of magnitude. As part of the fission process, radiation is emitted in the form of neutrons, heavy ions, beta particles, gamma rays, and some exotic particles such as neutrinos. Of these radiation types, only neutrons, beta particles and gamma rays are of concern. Heavy ions have short travel lengths in solids (10 micrometers) and neutrinos have low interaction cross sections with matter (1 pico-barns), thus both particles are of little consequence. Neutron flux magnitudes in research reactors such as the Kansas State University TRIGA Mark II Nuclear Research Reactor have a maximum steady state flux magnitude of  $10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$  and can achieve  $10^{17} \text{ n cm}^{-2} \text{ s}^{-1}$  during reactor pulsing [39]. Gamma-ray fluxes in the reactor are near the same order of magnitude as neutron flux.

### 2.3.1 In-Core Neutron Energies

In a nuclear reactor core a wide spectrum of neutron energies exist. Neutrons are first “born” at high energies after the fissioning of an atomic nucleus. Depending on the target isotope which undergoes fission, a varying number of neutrons are released, which boil off the fission fragments directly following the splitting of the nucleus. These neutrons are released into the reactor core environment and begin to scatter off surrounding atoms, losing portions of their kinetic energy as they transfer momentum. This energy transfer is dependent on the relative mass of the neutron in comparison to the target atoms is given as

$$E_F = \frac{(A^2 + 2A \cos(\theta) + 1)}{(A+1)^2} E_i, \quad (3)$$

where  $A$  defines the mass number of the target atom,  $E_i$  is the initial neutron energy,  $E_F$  is the final neutron energy, and  $\theta$  is the angle at which the neutron scatters off the target atom [12]. As neutrons reduce in energy and therefore speed, they begin to reach thermal equilibrium with the surrounding environment; at this point they are considered thermal or “slow” neutrons, reducing to speeds of  $2200 \text{ m s}^{-1}$  for room temperature environments ( $20^\circ\text{C}$  or  $293 \text{ K}$ ). As neutrons decrease in speed, the probability of absorption and subsequent fission in the fuel increases dramatically. The neutron energy spectrum inside a nuclear reactor is commonly characterized by three energy ranges: fast, epithermal, and thermal neutrons. Additionally, each energy range plays a key role in the neutron economy of the reactor and can be observed to determine reactor dynamics.

#### Fast Neutrons

The initial energy of neutrons released during fission, range from 0 – 10 MeV, with some born at energies as high as 20 MeV, but with low probabilities. Commonly referred to as the fission neutron energy spectra, (shown in Fig. 2-6 for various isotopes), can be modeled by the Watt distribution described in Eq. (4), where  $E$  is the neutron energy, and  $E_W$  and  $T_W$  are variables provided in Table 2-2.

$$W(E) = \frac{e^{-(E+E_W)/T_W}}{\sqrt{\pi E_W T_W}} \sinh \sqrt{\frac{4E_W E}{T_W^2}} \quad (4)$$

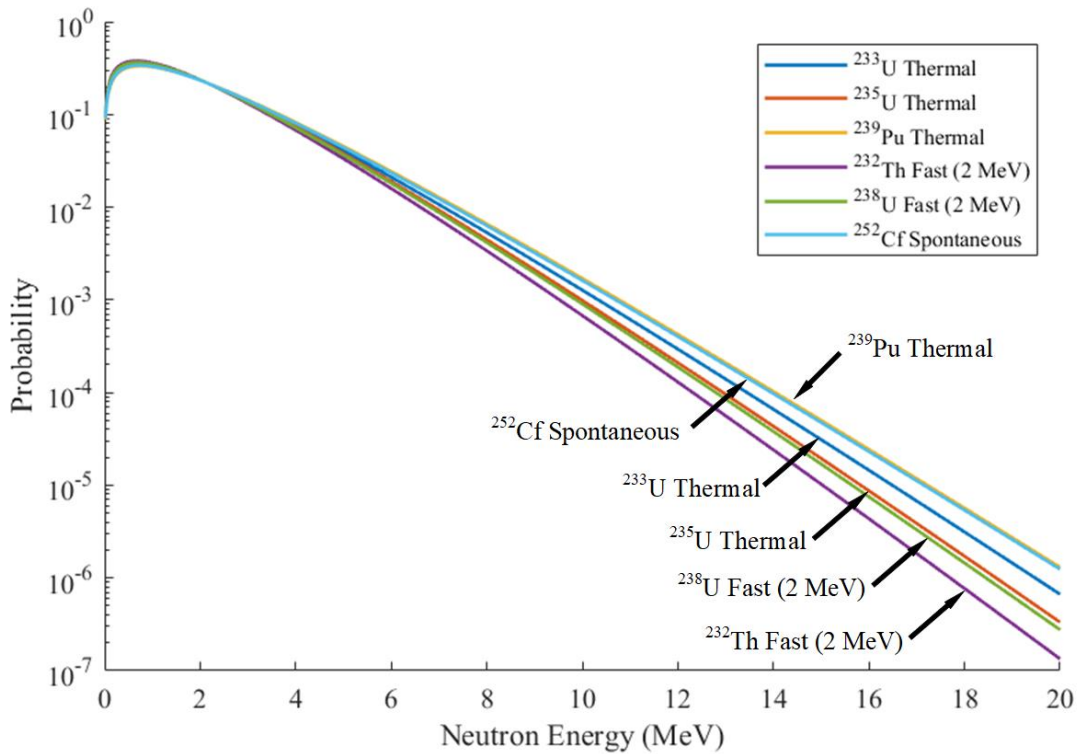


Fig. 2-6. Fission neutron energy spectra for various isotopes.

As will be discussed more in depth in Section 2.4, most isotopes have low interaction cross sections with fast neutrons. For this reason, fast neutrons are difficult to observe using neutron detection systems. Additionally, the fast neutron flux is primarily monitored for material testing purposes such as material degradation studies and is only of secondary interest for reactor power monitoring purposes.

Table 2-2. Variables for use in Eq. (4) to determine fission neutron energy spectrum.

| Nuclide           | Type of Fission | $E_w$  | $T_w$ |
|-------------------|-----------------|--------|-------|
| $^{233}\text{U}$  | thermal         | 0.3870 | 1.108 |
| $^{235}\text{U}$  | thermal         | 0.4340 | 1.035 |
| $^{239}\text{Pu}$ | thermal         | 0.4130 | 1.159 |
| $^{232}\text{Th}$ | fast (2 MeV)    | 0.4305 | 0.971 |
| $^{238}\text{U}$  | fast (2 MeV)    | 0.4159 | 1.027 |
| $^{252}\text{Cf}$ | spontaneous     | 0.3590 | 1.175 |

Source: Data from Shultis & Faw 1999, 2008 [1], [38]

## Epithermal Neutrons

As neutrons lose sufficient energy from scattering, they cross over from the fast region into the epithermal region. Although epithermal neutrons still have considerable energy and speed (between 10 eV to 1 MeV), enough energy has been lost to drop below the threshold energy required to cause fissionable isotopes to undergo fission, (see Section 2.4.3 for further discussion). For nuclear fission reactors, this region exhibits a reciprocal of energy ( $1/E$ ) shape predominately due to energy loss from scattering interactions. The reason for this  $1/E$  behavior is primarily due to the nature of fractional neutron energy loss via scattering as a function of its kinetic energy. As most reactors use materials such as beryllium, graphite, or water as moderating/reflecting materials, the loss of kinetic energy throughout the epithermal region is dominated by these low- $Z$  materials. Fig. 2-7 illustrated the cross sections of the most common moderating materials used in nuclear reactors.

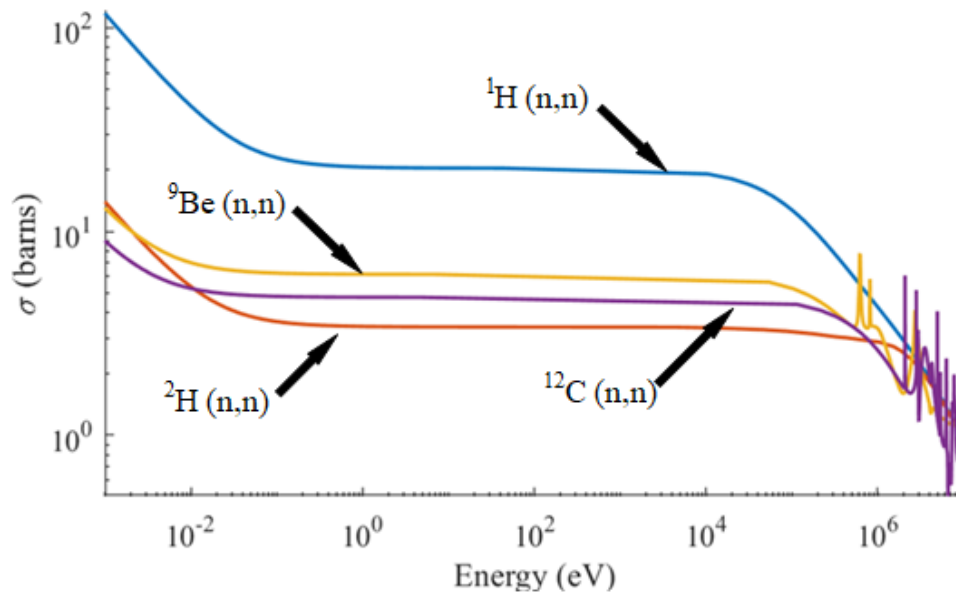


Fig. 2-7. Elastic scatter cross sections for the most common neutron reflector materials.

Because the elastic scattering cross section of most isotopes remain constant in this energy region, a constant fraction of energy loss occurs due to neutrons scattering off the reactor materials. In practicality, the resonance region deviates from an absolute  $1/E$  shape with various dips in neutron population due to resonance absorption peaks of the reactor materials [20]. If a neutron loses enough energy, it begins to reach thermal equilibrium with its surroundings, and is considered a thermal neutron (energies below 10 eV).

### **Thermal Neutrons**

Thermal neutrons are commonly considered the lowest neutron energy region, is described as such due to neutrons possessing energy levels which are close to thermal equilibrium with surrounding material. The Maxwell-Boltzmann equation is commonly used to describe the neutron energy population in the thermal region of the reactor neutron energy spectrum. This equation closely resembles the Maxwell–Boltzmann distribution which describe probability of particle velocities in gas kinetics. However, in the gas kinetics equation, temperature  $T$  describes the temperature at which the gas exists, whereas temperature  $T$  in the neutron equation, typically describes the temperature of the media in which neutrons scatter and lose their energy. In gas kinetics, particles lose their energy as they interact with the volume boundary and other gas molecules. While neutrons can scatter off each other, due to their small size, it is improbable, and therefore, the main mode of energy transfer occurs when neutrons scatter off the atoms of the medium. The Maxwell–Boltzmann neutron equation is defined as follows,

$$\varphi_{maxwell}(E) = \frac{2\pi n \sqrt{E} e^{-\frac{E}{kT}}}{(\pi kT)^{\frac{3}{2}}} = 2 e^{-\frac{E}{kT}} \sqrt{\frac{E}{\pi kT}}, \quad (5)$$

where,  $E$  is energy of the neutron (eV) typically spanning between 0 and 2 eV,  $n$  is  $kT$ ,  $k$  is Boltzmann's constant ( $8.62 \times 10^{-5}$  eV K<sup>-1</sup>), and  $T$  is temperature (K) of the surrounding medium [1], [20]. Typically, the temperature of the majority moderator in a reactor is considered for the representative neutron energy spectrum. Various Maxwellian neutron spectra are shown in Fig. 2-8 as a function of temperature of the medium.

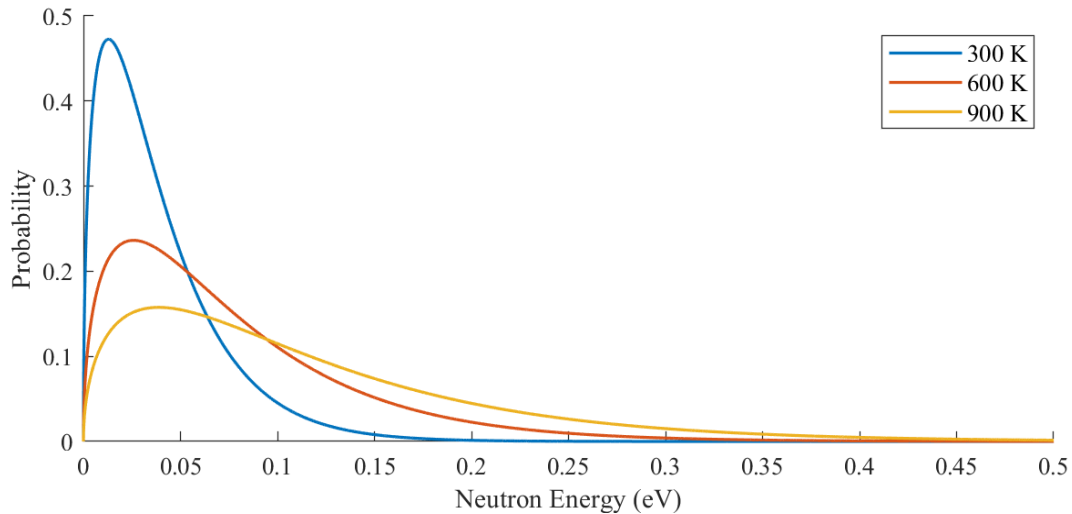


*Fig. 2-8. Thermal neutron energy distribution for various temperatures of surrounding medium.*

### **Additional Information**

During the fission process, the vast majority (approximately 99%) of neutrons are emitted directly following the splitting of the nucleus with the remainder being emitted delayed by daughter isotopes decay. Neutron energies beyond the fast neutron region exist but are typically only the result of high-energy particle physics experiments or cosmic events such as supernovae. Although these high energy neutrons are of interest to industries such as astrophysics and space programs, they are uncommon in nuclear reactor applications and therefore will not be discussed as part of this work. A more thorough examination of such neutron energy regions and their characteristics can be found elsewhere.

### **2.3.2 Beta-Particle & Gamma-ray Environment**

A high flux of gamma rays and beta particles exist near the reactor core as by-products of the fission process. Gamma rays are produced as both prompt by-products of fission and subsequent decay of fission product nuclei. Beta particles are the results of fission product decay only after the initial fission event has occurred. Both particles have the potential of inducing erroneous signals inside instrumentation or causing structural damage, and thus each particle flux must be considered. The gamma-ray and beta-particles fluxes inside the nuclear reactor core are typically within one order of magnitude of the neutron flux, although beta particles are highly localized around the fuel regions of the core due to their short travel distance. Approximately 7-8 prompt gamma rays, 7-8 delayed gamma rays, 6 beta particles are emitted per fission event [26]. More information about these particle fluxes is found in Section 3.1.2.

### **2.3.3 Pressure and Temperature Conditions (Research and Power Reactors)**

Many research reactor configurations utilize an open pool design to reduce cost, complexity and for ease of access for experiment deployments. For such reactors, pressures are limited by the depth of the reactor coolant (most commonly water), and thus are confined to pressures between 1 – 3 atm (100 – 300 kPa) for submerged components. Temperatures in such reactors are also limited by the coolant critical heat flux, at which point the coolant begins to rapidly boil leading to a reduction in heat transfer, which will ultimately result in a full vapor film development which inhibits heat transfer and fuel failure may occur. Commonly, research reactors are kept well below this point for safety purposes and for example, TRIGA coolant temperature limits are typically restricted to 54°C (although temperature may vary depending on fuel material characteristics and forced convection of the coolant). The TRIGA fuel is limited to temperatures below 1150°C for short transients (such as pulsing operations), to prevent hydrogen outgassing, and 750°C for steady state operations [39], [40].

Temperatures and pressures of power reactors far exceed the limitations of research reactors, primarily due to the increased power generation efficiency as a function of higher temperatures, which can only be achieved at higher pressures to avoid coolant critical heat flux. Although exotic working fluids such as gases and liquid metals are utilized, the most common coolant is still water. The critical temperature of water is 375°C, at which point increases in pressure will not inhibit water from undergoing a phase change to the gaseous state. Pressures for commercial power reactors depend on the design of the reactor and the working fluid utilized. As such, pressures vary over a wide range of values spanning from low pressure designs like the boiling water reactor (BWR) at approximately 7.17 MPa to high pressure designs such as the supercritical water-cooled reactor at 22.1 MPa [1].

### **2.3.4 Material Compatibility Considerations**

Reactor instrumentation is often deployed in direct contact with the working fluid or support structure of the reactor. In some reactor designs such as molten salt reactors, instruments may come in direct contact with the fuel. Material compatibility is therefore important when considering reactor instrumentation and careful planning is needed to provide instrument longevity in case it comes contact with corrosive or reactive materials. Commonly, reactor instruments are deployed into a high-temperature and high-pressure water environment (as previously mentioned),

but the future generation of reactor design boasts new and challenging compatibility requirements. High temperature gas-cooled reactors (HTGRs), lithium fluoride thorium reactors (LFTRs), and sodium-cooled fast reactors (SFRs) are among several reactor designs with some of the most volatile environments for instruments to be deployed. Previous studies have examined instrument construction materials such as high nickel or chromium steel alloys, refractory materials, or even ceramic-metal composites as candidates for instrument construction that can survive the harsh, corrosive conditions [41]. While the work contained herein was not focused on extremely adverse reactor conditions, studies were conducted on possible modifications to previous designs, to help increase detector performance in volatile environments.

## **2.4 Materials for Measuring Neutron Flux**

Due to the neutral relative charge of neutrons (as described in Section 2.2.3), most methods of detection rely on the usage of neutron reactive materials which either absorb or scatter neutrons to produce detectable reaction products. Due to the diverse energies of neutrons which exist in the reactor core, common practice is to use materials that are sensitive to various neutron energy regions. As described in Section 2.3.1, neutrons are typically characterized in three categories: thermal, epithermal, and fast. Thermal and fast neutrons are the two main energy groups of interest, as they are indicative of the beginning and ending energy region of neutrons in the core.

Ideal neutron reactive materials require three characteristics to be suitable for use in a reactor to measure neutron flux. First, the material needs to have an absorption cross section that allows for it to readily absorb neutrons and produce reaction products. Second, the released reaction products must have a significant amount of energy, a positive Q-value, which can be deposited into the detection medium. Third, the neutron reaction must be spontaneous.

### **2.4.1 Low-Z Isotopes**

Some of the most common low-Z isotopes used for thermal neutron detection are  $^3\text{He}$ ,  $^6\text{Li}$ , and  $^{10}\text{B}$  due to a high thermal neutron cross section of absorption and large reaction Q-values as illustrated in Fig. 2-9. Each of these isotopes are considered  $1/v$  absorbers for a large span of energies as shown in Fig. 2-10. A  $1/v$  absorber is an isotope with a reaction rate that is independent of the neutron or target nuclei velocity.

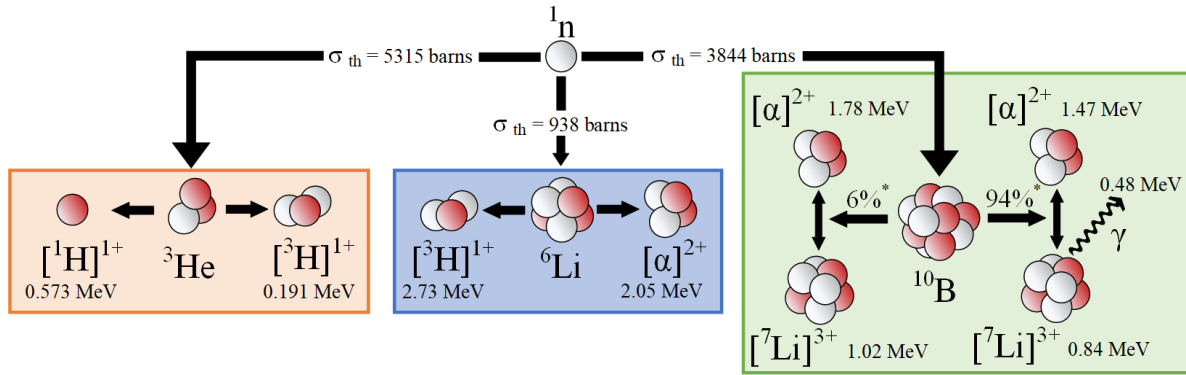


Fig. 2-9. The three most used low Z isotopes for thermal neutron detection. For the  $^{10}\text{B}$  reaction, the branching ratio for the ground state (left) and excited state (right) is not constant at 6:94 and will deviate at higher energies as shown in Fig. 2-11 [34].

These  $1/v$  absorbers have a cross section of absorption which is proportional to the energy of the incident neutron. While these low-Z isotopes are primarily used for thermal neutrons, they can and are commonly used for epithermal neutron detection. Materials such as cadmium or gadolinium, which have high thermal neutron absorption cross sections, can be used as a filter to remove thermal neutrons from interacting with  $^3\text{He}$ ,  $^6\text{Li}$  or  $^{10}\text{B}$ , leading to a preferential response to epithermal neutrons.

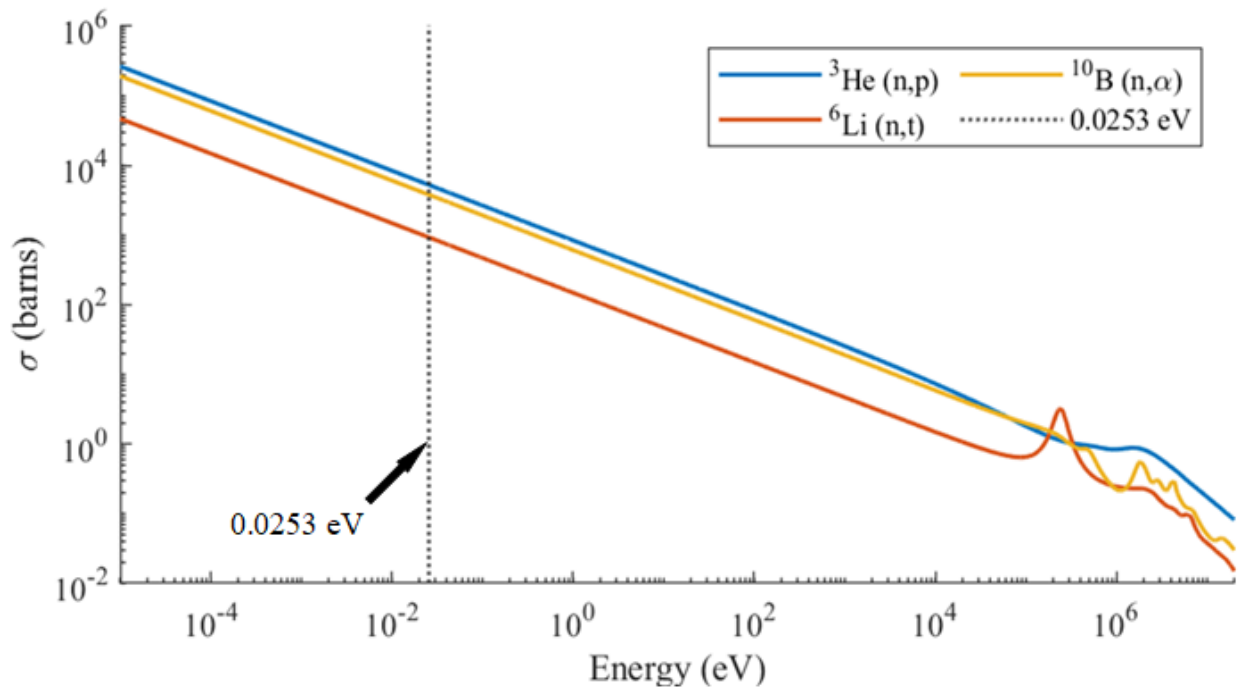


Fig. 2-10. Plot of common low-Z isotopes showing  $1/v$  behavior for most of the neutron energy spectrum.

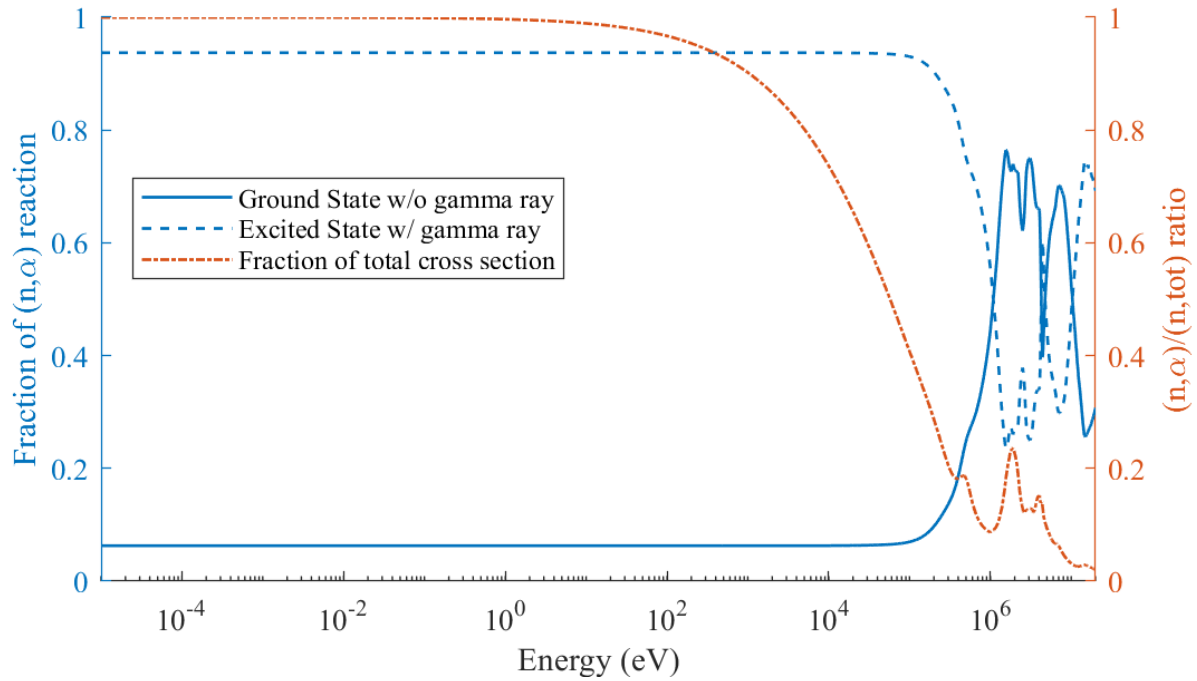


Fig. 2-11. Plot of  $^{10}\text{B}(n,\alpha)^7\text{Li}$  branching ratio with comparison to total cross section.

## 2.4.2 High-Z Isotopes

There are also high-Z isotopes commonly utilized as neutron reactive materials. These materials typically undergo the  $(n,\gamma)$  reaction with subsequent daughter isotopes emitting low energy gamma rays or beta particles. The daughter isotopes commonly have much lower decay Q-values in comparison to  $^3\text{He}$ ,  $^6\text{Li}$ , and  $^{10}\text{B}$ . Among these high-Z isotopes are  $^{59}\text{Co}$ ,  $^{195}\text{Pt}$ ,  $^{103}\text{Rh}$  and  $^{51}\text{V}$  with cross sections shown in Fig. 2-12. A more exhaustive list of high-Z neutron reactive materials can be found in Table 2-4. Because of the larger atomic number, higher density, and the large quantities of mass required to achieve moderate neutron sensitivity, gamma rays commonly interact with these materials resulting in electron emissions. These energetic electrons mimic the response of the daughter isotope decay, and therefore can produce false positive events.

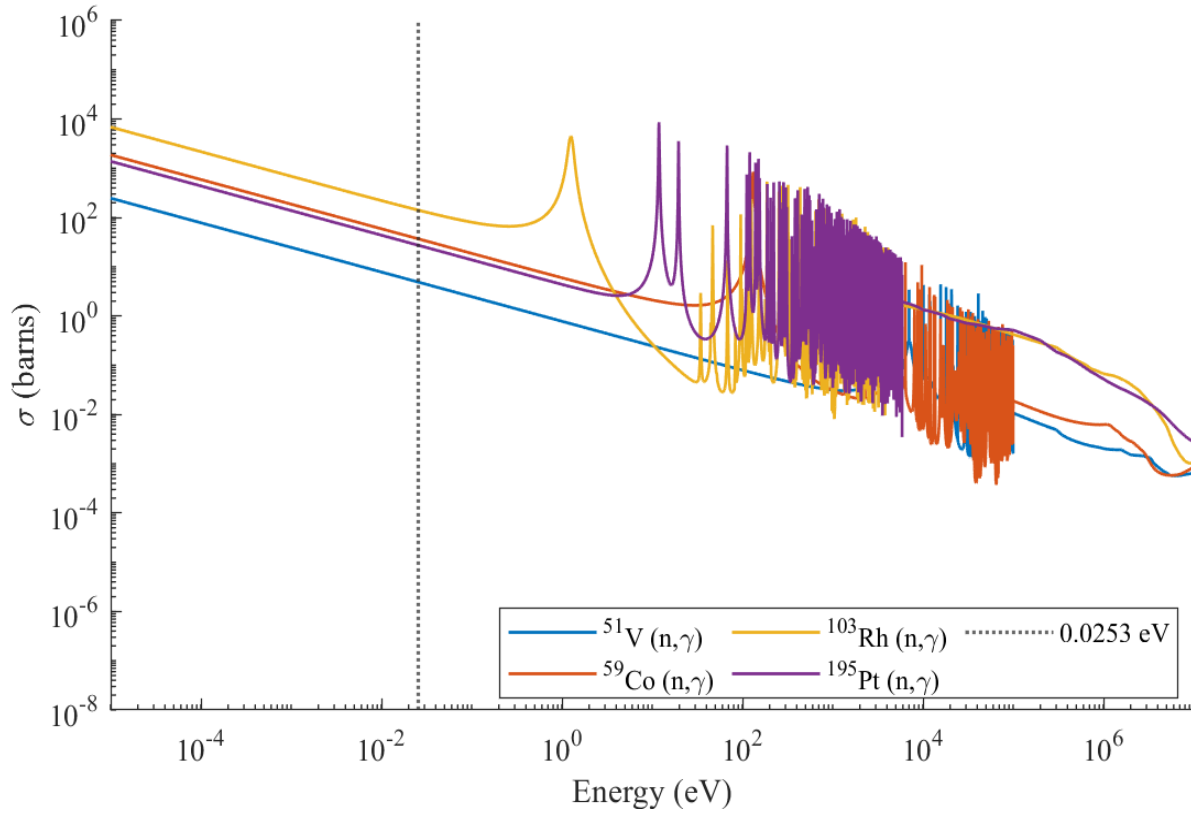


Fig. 2-12. The  $(n,\gamma)$  cross sections for common high-Z isotopes used for neutron detection.

### 2.4.3 Fissioning Isotopes

Another group of high-Z isotopes are considered separate from the aforementioned category, which primarily undergo the  $(n,f)$  reaction. These materials can either undergo fission immediately after absorbing a thermal neutron (fissile), undergo fission after absorbing a fast neutron (fissionable), or can be transmuted into a fissile or fissionable isotope after neutron absorption (fertile). As described in Section 2.3, the fission process commonly produces 200 MeV of energy in the form of two high velocity, ionized fission fragments. Due to the potential of large energy deposition in the detection medium, fission events can easily be distinguished from non-neutron induced events. Additionally, because the fission process occurs also immediately after absorption, with little delay, the resulting fission fragments can be used to detect the neutron flux in real-time. Some of the most common fissile, fertile, and fissionable isotopes and their respective cross sections are shown in Fig. 2-13.

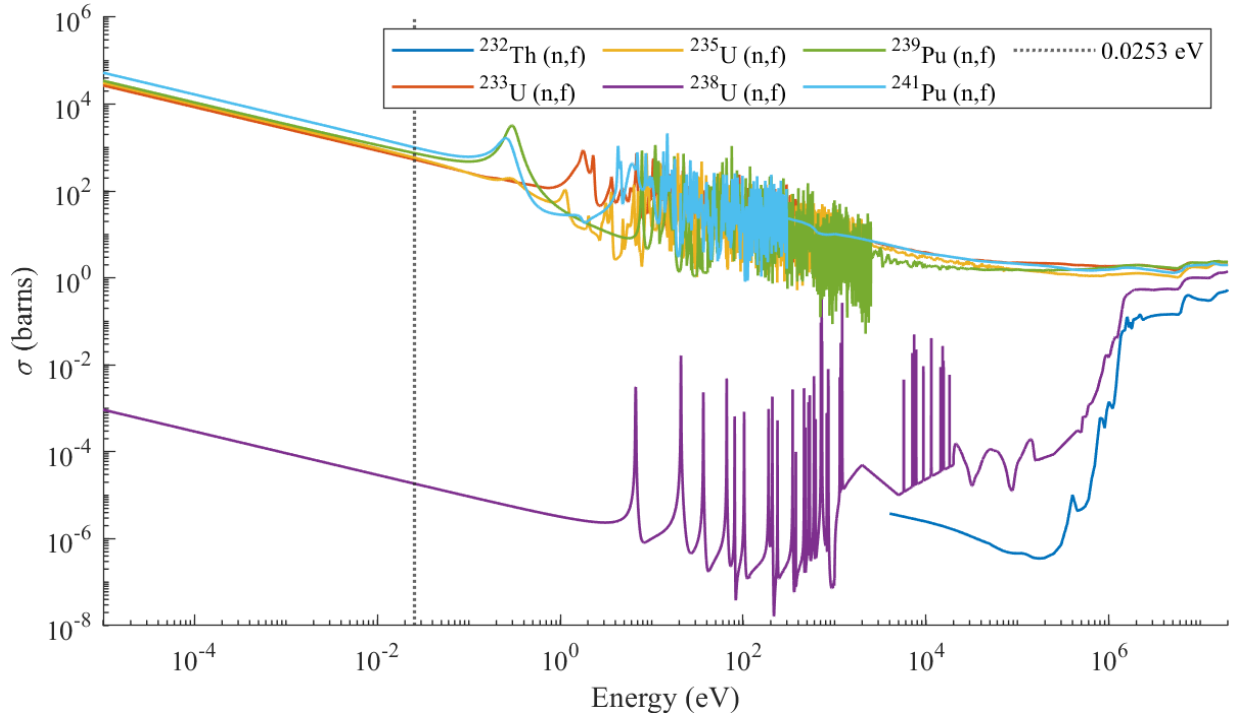


Fig. 2-13. Graph of the fission cross section of important isotopes both for nuclear power and nuclear weapons. Note the clear delineation that exists between fissile isotopes ( $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$ ) and fissionable isotopes ( $^{232}\text{Th}$ ,  $^{238}\text{U}$ ).

#### 2.4.4 Summary

The isotopes described above are only a small selection of materials used for neutron detection. While all are suitable for neutron detection, there are drawbacks to using some of these isotopes for monitoring neutron flux inside a nuclear reactor core. For the low-Z isotopes ( $^3\text{He}$ ,  $^6\text{Li}$ ,  $^{10}\text{B}$ ), large cross sections significantly increase sensitivity to neutrons but results in shortened detector lifetime and can cause local neutron flux perturbation. A neutron flux of  $10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$  for a duration of 1 month will result in both  $^3\text{He}$  and  $^{10}\text{B}$  being fully consumed, and  $^6\text{Li}$  will be 25% consumed. Additionally, due to the low density of low-Z isotopes, neutron sensitivity per volume of material is lower when compared to other neutron reactive material alternatives.

For the listed high-Z isotopes ( $^{59}\text{Co}$ ,  $^{195}\text{Pt}$ ,  $^{103}\text{Rh}$ ,  $^{51}\text{V}$ ), lower cross sections allow for longer detector lifetime (2 – 8 years until full depletion at  $10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ ), but with significantly reduced neutron sensitivity. These materials also suffer from low Q-values of daughter isotope decay and produce reaction products that can escape the detector without depositing their total energy. Additionally, most of these isotopes have delayed decay and emission of reaction products (0.5

minutes  $< t < 5$  hours) which causes residual signal and impacts the detector's ability to track short time scale changes in neutron flux.

Finally, there are fissile and fissionable materials such as the actinides listed above, that have moderately large neutron cross sections and neutron sensitivity. While this moderate neutron cross section will cause quick depletion of the material, fertile isotopes can be added to the material to “breed” more fissionable or fissile isotopes to replenish the material and stabilize the neutron sensitivity [42]. Additionally, the Q-value of a fission event is significantly larger (200 MeV) than all low-Z and high-Z isotopes. This large Q-value allows for simple discrimination techniques to remove signal intrusion from other events such as gamma-ray or beta-particle interactions.

## 2.5 Alternative In-Core Flux Monitoring Technologies

In-core flux monitoring is accomplished by a variety of technologies that can be compiled into two categories: active or passive monitoring platforms. Passive methods have been traditionally used to confirm reactor-physics models and perform experiment dosimetry. Passive neutron flux measurements are commonly accomplished with a “cook & look” method that provides neutron flux information *post facto*. In contrast, active neutron flux monitoring methods have historically been deployed on the periphery of the reactor core and can only observe general reactor dynamics. Recently, active neutron flux monitoring platforms have seen prolific use inside the reactor core as detector form factors have reduced over the course of the last few decades. Both active and passive measurement technologies are utilized in a broad range of neutron flux magnitudes spanning several approximate categories as shown in Fig. 2-14.

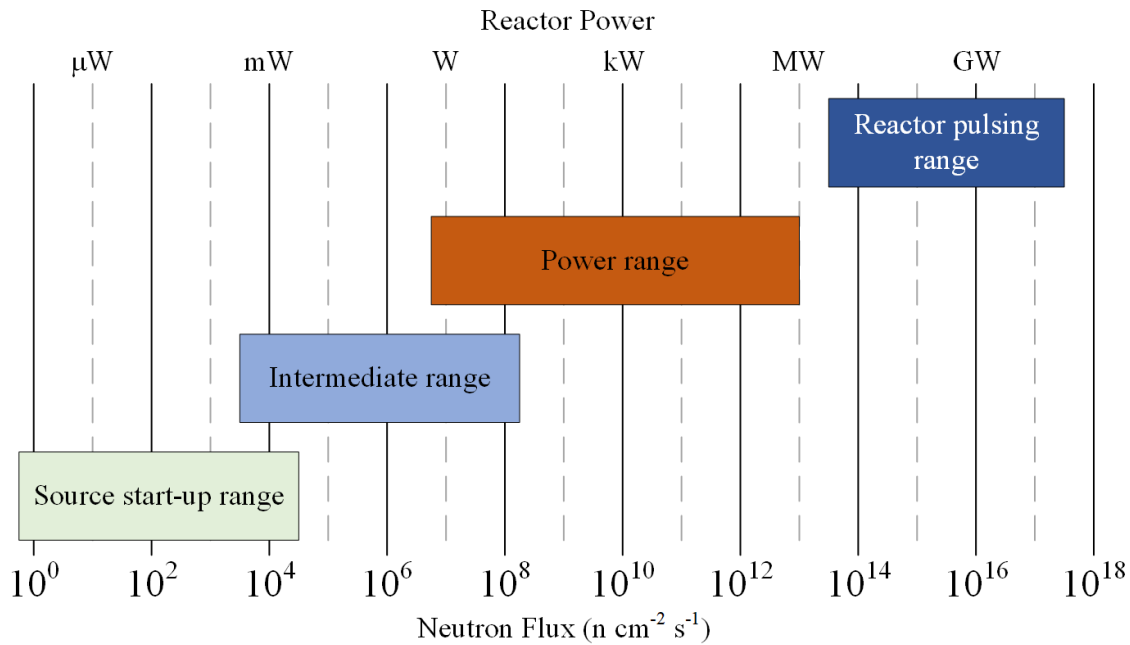


Fig. 2-14. Flux magnitude and reactor power ranges for in-core neutron detectors. Adapted from material previously reported [32], [41].

### 2.5.1 Active Methods

Active flux monitoring devices provide real-time neutron flux information such as the localized neutron population. This category of sensors includes methods that provide continuous signal during operation, and the Micro-Pocket Fission Detector (MPFD) platform is included in this category. The two most common competitive technologies presently available would be the sub-miniature fission chamber (SMFC) and the self-powered neutron detector (SPND).

#### Miniature Fission Chambers (MFC) and Sub-Miniature Fission Chambers (SMFC)

In the past few decades, the French Alternative Energies and Atomic Energy Commission (CEA) has been developing newer versions of traditional coaxial in-core fission chamber. These new detectors have nominal exterior diameters ranging from less than 1 cm (such as the CF8GR, CF3, CFUR) known as Miniature Fission Chambers (MFC) to variants with an outer diameter of 1.5 mm (such as the CFUZ53) known as Sub-Miniature Fission Chambers (SMFC). The SMFC is currently the smallest diameter coaxial fission chamber commercially available and a cross-sectional view of the device is shown in Fig. 2-15.

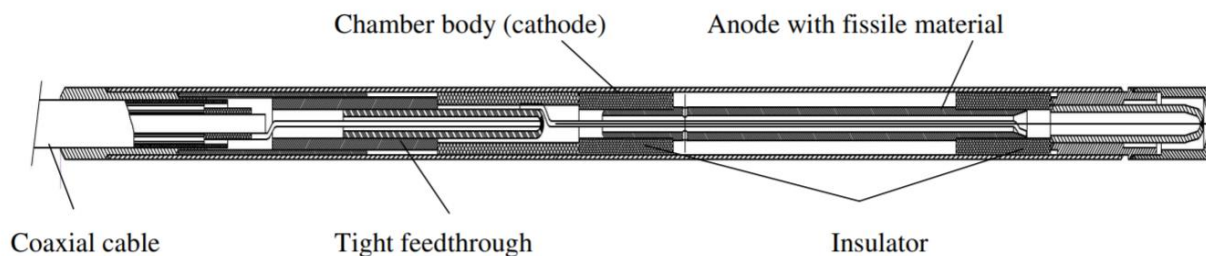


Fig. 2-15. Cross-sectional view of a 1.5-mm diameter cylindrical fission chamber (CFUZ53). Note that this image has a vertical dimension scaling of 1.75 for clarity) [43].

Both the MFC and SMFC detectors are commonly designed to have small inter-electrode spacing, utilizing anodes and cathodes with similar diameters ( $[A/C]_{\phi} \geq 0.5$ ). Because of these design characteristics, these detectors function as ion chambers which can be operated in pulse mode, current mode, or mean square voltage (MSV) mode, also known as Campbell mode. Additionally, the detectors can incorporate a variety of fissile materials including natural and enriched uranium, with fissile masses ranging from  $10^{-5} - 10^{-2}$  g [43]–[45]. Design and operating characteristics of commercial offerings for the CFUR43 and CFUR64 MFC and CFUZ53 SMFC are provided in Table 2-3.

Table 2-3. Design and operating characteristics of commercially available MFC and SMFC detectors manufacturing by Photonis [46]–[49].

| Det. Type | Product Number | Ext. $\phi$ (mm) | Fill gas                       | Thermal Neutron Flux Range ( $\text{n cm}^{-2} \text{s}^{-1}$ )* |                  |                                |
|-----------|----------------|------------------|--------------------------------|------------------------------------------------------------------|------------------|--------------------------------|
|           |                |                  |                                | Pulse Mode                                                       | MSV Mode         | Current Mode                   |
| SMFC      | CFUZ53         | 1.5              | Ar (110 kPa)                   | -                                                                | -                | $2 \times 10^{11} - 10^{14}$   |
| MFC       | CFUR43         | 3                | Ar (110 kPa)                   | -                                                                | -                | $10^{11} - 1.5 \times 10^{14}$ |
| MFC       | CFUR64         | 3                | Ar + 4% $\text{N}_2$ (500 kPa) | $10^7 - 3 \times 10^{11}$                                        | $10^6 - 10^{14}$ | $10^{12} - 10^{14}$            |

Each detector has a sensitive length of 13 mm, and maximum thermal neutron fluence of  $1.5 \times 10^{20} \text{ n cm}^{-2}$

\* Operating characteristics depend on electronics

Both the MFC and SMFC platforms are intended for single point measurement, but can be deployed in an array of chambers to obtain axial neutron flux profiling. Additionally, the SMFC platform has been utilized to map axial neutron flux profiles by moving the detector through the core at incremental steps [50]–[52].

## Self-Powered Neutron Detectors (SPND)

Self-powered neutron detectors (SPND), also referred to as collectrons or Hilborn detectors, rely on neutron activation of the emitter material which releases prompt gamma rays, and transmutes into radioactive isotopes with short half-lives resulting in gamma-ray emission or beta-particle decay. SPNDs are commonly divided into two categories: prompt and delayed response. A wide variety of isotopes have been proposed and tested with some of the most common materials displayed in Table 2-4. For comparison purposes, materials response and burnup was normalized to a common emitter diameter of 500  $\mu\text{m}$ . The neutron reactive material, known as the emitter, is electrically connected to the emitter wire, which is surrounded by insulating materials, such as  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{MgO}$  or  $\text{HfO}_2$ , and encased in an outer conductive sheath known as the collector. This standard geometry is illustrated in Fig. 2-16 including an optional additional conductor inside the insulation which provides a baseline signal of external radiation interaction to compensate for interfering external noise. There are various mechanisms by which an SPND produces neutron-induced signals. The primary mechanism relies on neutron absorption in the emitter material causing transmutation which results in decay, releasing either gamma rays, beta particles or sometimes both. During gamma-ray decay, the gamma rays can interact with the emitter material producing energetic electrons which can exit the emitter, permeating the insulation and producing charge carriers resulting in an induced current. Likewise, during beta-particle decay, the beta particles can exit the emitter producing charge carriers along its path, inducing a current. A femtometer can be connected to the emitter and sheath to form a closed circuit and measure current flow in the system. In an ideal SPND, current flow would be directly proportional the reaction rate (i.e., neutron flux) in the emitter.

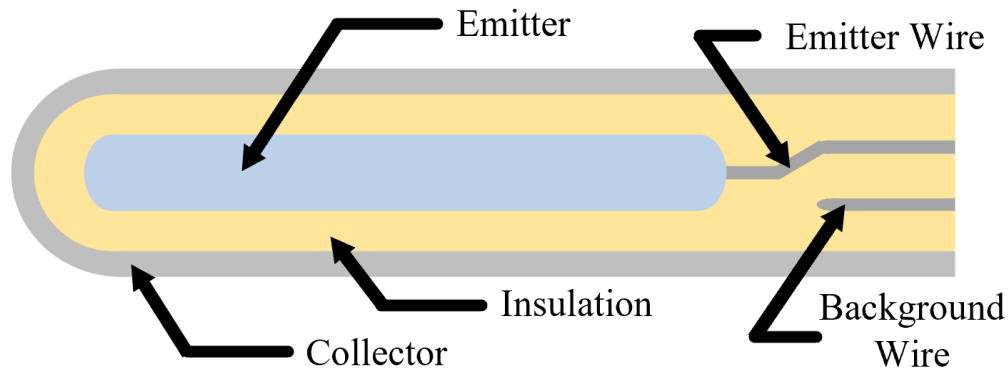


*Fig. 2-16. Standard concept design of SPNDs with the inclusion of a background subtraction wire.*

SPNDs have demonstrated excellent neutron flux monitoring capabilities in real-time or delayed signal setups, but with two key drawbacks. First, SPNDs commonly rely on neutron activation and subsequent decay of the material resulting from transmutation. This process causes a compounded delay effect as the materials used typically have a low neutron cross section of absorption and the resulting unstable isotopes have half-lives on the order of 10 seconds to 2 hours, in comparison to fission reactions which occurs within  $10^{-14}$  seconds [1]. SPND manufacturers commonly must balance this effect when designing the instrument for a specific experimental use or power monitoring application. As such, there is no SPND which is designed as a “one size fits all” format, although rhodium SPNDs have become widely accepted as one of the most versatile designs.

Table 2-4 List of a variety of SPND emitter materials with respective isotope characteristics and burnup longevity.

| Emitter Isotope                                                                  | Isotopic Abundance | Thermal Cross section (barns) | Product Isotope    | Product Half-life   | Contributing Reactions | Avg PG (MeV)   | Max PG (MeV) | Avg E <sub>β</sub> (MeV) | Max E <sub>β</sub> (MeV) | Mon.% Burnup |
|----------------------------------------------------------------------------------|--------------------|-------------------------------|--------------------|---------------------|------------------------|----------------|--------------|--------------------------|--------------------------|--------------|
| <b>Aluminum [Delayed] <math>1.0 \times 10^{-22} \text{ A s cm}^{-3}</math></b>   |                    |                               |                    |                     |                        |                |              |                          |                          |              |
| <sup>27</sup> Al                                                                 | 100.00%            | 0.23                          | <sup>28</sup> Al   | 135 s               | β-                     | 3.52           | 7.72         | 1.24                     | 2.86                     | 0.006        |
| <b>Vanadium [Delayed] <math>8.31 \times 10^{-23} \text{ A s cm}^{-3}</math></b>  |                    |                               |                    |                     |                        |                |              |                          |                          |              |
| <sup>50</sup> V                                                                  | 0.25%              | 44.7                          | <sup>51</sup> V    | stable              | prompt γ               | 3.03           | 9.44         | -                        | -                        | 1.16         |
| <sup>51</sup> V                                                                  | 99.75%             | 4.92                          | <sup>52</sup> V    | 225 s               | β-                     | 3.34           | 7.31         | 1.07                     | 2.54                     | 0.12         |
| <b>Cobalt [Prompt] <math>1.52 \times 10^{-23} \text{ A s cm}^{-3}</math></b>     |                    |                               |                    |                     |                        |                |              |                          |                          |              |
| <sup>59</sup> Co                                                                 | 100.00%            | 37.2                          | <sup>60</sup> Co   | 5.27 y              | prompt γ               | 2.42           | 7.49         | -                        | -                        | 1.00         |
| <b>Nickel [Prompt] <math>4.64 \times 10^{-22} \text{ A s cm}^{-3}</math></b>     |                    |                               |                    |                     |                        |                |              |                          |                          |              |
| <sup>58</sup> Ni                                                                 | 68.08%             | 4.23                          | <sup>59</sup> Ni   | $7.6 \times 10^4$ y | prompt γ               | 5.32           | 9.00         | 0.02                     | 0.05                     | 0.11         |
| <sup>60</sup> Ni                                                                 | 26.22%             | 2.40                          | <sup>61</sup> Ni   | stable              | prompt γ               | 4.94           | 7.82         | -                        | -                        | 0.06         |
| <sup>61</sup> Ni                                                                 | 1.14%              | 2.51                          | <sup>62</sup> Ni   | stable              | prompt γ               | 2.78           | 10.6         | -                        | -                        | 0.07         |
| <sup>62</sup> Ni                                                                 | 3.63%              | 14.9                          | <sup>63</sup> Ni   | 101 y               | prompt γ               | 5.85           | 6.84         | 0.02                     | 0.07                     | 0.39         |
| <sup>64</sup> Ni                                                                 | 0.93%              | 1.48                          | <sup>65</sup> Ni   | 2.52 y              | prompt γ               | 4.00           | 6.03         | 0.63                     | 2.14                     | 0.04         |
| <b>Molybdenum [Prompt] <math>8.50 \times 10^{-24} \text{ A s cm}^{-3}</math></b> |                    |                               |                    |                     |                        |                |              |                          |                          |              |
| <sup>92</sup> Mo                                                                 | 14.84%             | 0.08                          | <sup>93</sup> Mo   | $4.0 \times 10^3$ y | prompt γ               | 3.55           | 8.07         | -                        | -                        | 0.002        |
| <sup>94</sup> Mo                                                                 | 9.25%              | 0.34                          | <sup>95</sup> Mo   | stable              | prompt γ               | 6.65           | 7.17         | -                        | -                        | 0.009        |
| <sup>95</sup> Mo                                                                 | 15.92%             | 13.4                          | <sup>96</sup> Mo   | stable              | prompt γ               | 1.61           | 8.38         | -                        | -                        | 0.350        |
| <sup>96</sup> Mo                                                                 | 16.68%             | 0.60                          | <sup>97</sup> Mo   | stable              | prompt γ               | 1.12           | 6.14         | -                        | -                        | 0.016        |
| <sup>97</sup> Mo                                                                 | 9.55%              | 2.20                          | <sup>98</sup> Mo   | stable              | prompt γ               | 1.61           | 7.91         | -                        | -                        | 0.057        |
| <sup>98</sup> Mo                                                                 | 24.13%             | 1.30                          | <sup>99</sup> Mo   | 65.9 h              | β-                     | 0.79           | 5.93         | 0.39                     | 1.22                     | 0.034        |
| <sup>100</sup> Mo                                                                | 9.63%              | 0.20                          | <sup>101</sup> Mo  | 14.6 m              | β-                     | 1.06           | 5.40         | 0.52                     | 2.62                     | 0.005        |
| <b>Rhodium [Prompt] <math>1.0 \times 10^{-21} \text{ A s cm}^{-3}</math></b>     |                    |                               |                    |                     |                        |                |              |                          |                          |              |
| <sup>103</sup> Rh                                                                | 100.00%            | 134                           | <sup>104</sup> Rh  | 42.3 s              | β-                     | 4.92           | 7.00         | 0.99                     | 2.44                     | 3.9          |
|                                                                                  |                    | 11                            | <sup>104m</sup> Rh | 4.34 m              | prompt γ               | 0.22           | 0.98         | -                        | -                        |              |
| <b>Silver [Delayed] <math>3.55 \times 10^{-22} \text{ A s cm}^{-3}</math></b>    |                    |                               |                    |                     |                        |                |              |                          |                          |              |
| <sup>107</sup> Ag                                                                | 51.84%             | 0.35                          | <sup>108m</sup> Ag | 438 y               | prompt γ               | 0.28           | 1.57         | -                        | -                        | 0.9          |
|                                                                                  |                    | 37.6                          | <sup>108</sup> Ag  | 143 s               | β-                     | 6.22           | 7.27         | 0.62                     | 1.65                     |              |
| <sup>109</sup> Ag                                                                | 48.16%             | 4.4                           | <sup>110m</sup> Ag | 250 d               | prompt γ               | 0.18           | 0.79         | 0.07                     | 0.53                     | 2.3          |
|                                                                                  |                    | 87                            | <sup>110</sup> Ag  | 24.6 s              | β-                     |                |              | 1.19                     | 2.89                     |              |
| <b>Gadolinium [Prompt] <math>1.56 \times 10^{-22} \text{ A s cm}^{-3}</math></b> |                    |                               |                    |                     |                        |                |              |                          |                          |              |
| <sup>152</sup> Gd                                                                | 0.20%              | 735                           | <sup>153</sup> Gd  | 240 d               | prompt γ               | 0.69           | 6.25         | -                        | -                        | 19.1         |
| <sup>154</sup> Gd                                                                | 2.18%              | 85.2                          | <sup>155</sup> Gd  | stable              | prompt γ               | 1.02           | 6.44         | -                        | -                        | 2.21         |
| <sup>155</sup> Gd                                                                | 14.80%             | $6.07 \times 10^4$            | <sup>156</sup> Gd  | stable              | prompt γ               | 1.44           | 8.45         | -                        | -                        | >100         |
| <sup>156</sup> Gd                                                                | 20.47%             | 1.80                          | <sup>157</sup> Gd  | stable              | prompt γ               | 3.62           | 6.36         | -                        | -                        | 0.05         |
| <sup>157</sup> Gd                                                                | 15.65%             | $2.53 \times 10^5$            | <sup>158</sup> Gd  | stable              | prompt γ               | 1.44           | 7.86         | -                        | -                        | >100         |
| <sup>158</sup> Gd                                                                | 24.84%             | 2.20                          | <sup>159</sup> Gd  | 18.5 h              | prompt γ               | 1.52           | 5.94         | 0.30                     | 0.97                     | 0.06         |
| <sup>160</sup> Gd                                                                | 21.86%             | 1.41                          | <sup>161</sup> Gd  | 3.66 m              | β-                     | 2.89           | 5.32         | 0.56                     | 1.64                     | 0.04         |
| <b>Hafnium [Prompt] <math>9.98 \times 10^{-23} \text{ A s cm}^{-3}</math></b>    |                    |                               |                    |                     |                        |                |              |                          |                          |              |
| <sup>174</sup> Hf                                                                | 0.16%              | 548                           | <sup>175</sup> Hf  | 70.0 d              | prompt γ               | 1.50           | 6.58         | -                        | -                        | 14.2         |
| <sup>176</sup> Hf                                                                | 5.26%              | 21.4                          | <sup>177m</sup> Hf | stable***           | prompt γ               | 0.41           | 0.62         | -                        | -                        | 0.55         |
| <sup>177</sup> Hf                                                                | 18.60%             | 374                           | <sup>178</sup> Hf  | stable***           | prompt γ               | 4.60           | 7.53         | -                        | -                        | 9.69         |
| <sup>178</sup> Hf                                                                | 27.28%             | 80                            | <sup>179m</sup> Hf | stable***           | prompt γ               | 1.05           | 5.72         | -                        | -                        | 2.07         |
| <sup>179</sup> Hf                                                                | 13.62%             | 0.43                          | <sup>180m</sup> Hf | 5.53 h              | prompt γ               | 5.32           | 7.30         | -                        | -                        | 1.18         |
|                                                                                  |                    | 45                            | <sup>180</sup> Hf  | stable              |                        |                |              |                          |                          |              |
| <sup>180</sup> Hf                                                                | 35.08%             | 13.1                          | <sup>181m</sup> Hf | 42.4 d              | β-                     | 2.65           | 5.70         | 0.12                     | 1.03                     | 0.34         |
| <b>Platinum [Prompt] <math>1.28 \times 10^{-22} \text{ A s cm}^{-3}</math></b>   |                    |                               |                    |                     |                        |                |              |                          |                          |              |
| <sup>194</sup> Pt                                                                | 32.97%             | 0.1                           | <sup>195m</sup> Pt | 4.01 d              | prompt γ               | 0.39           | 1.09         | -                        | -                        | 0.03         |
|                                                                                  |                    | 1.1                           | <sup>195</sup> Pt  | stable              |                        |                |              | -                        | -                        |              |
| <sup>195</sup> Pt                                                                | 33.83%             | 27.5                          | <sup>196</sup> Pt  | stable              | prompt γ               | 1.60           | 7.92         | -                        | -                        | 0.71         |
| <sup>196</sup> Pt                                                                | 25.24%             | 0.04                          | <sup>197m</sup> Pt | 1.59 h              | prompt γ               | 1.84           | 5.85         | 0.22                     | 0.71                     | 0.003        |
|                                                                                  |                    | 0.06                          | <sup>197</sup> Pt  | 19.9 h              | prompt γ               |                |              | 0.20                     | 0.72                     |              |
| <sup>198</sup> Pt                                                                | 7.16%              | 0.3                           | <sup>199m</sup> Pt | 13.6 s              | IT                     |                |              | -                        | -                        |              |
|                                                                                  |                    | 3.5                           | <sup>199</sup> Pt  | 30.8 m              | β-                     | Delayed Gammas |              | 0.54                     | 1.70                     | 0.10         |

\* Sensitivities based on aggregate of previous works and normalized to a 0.5 mm diameter

\*\* Monthly burnup given for flux of  $10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ , ( $\Phi = 2.6 \times 10^{20} \text{ n cm}^{-2}$ )

\*\*\* Isomeric transitions

Sources: [1], [32], [33], [53]–[65]

The second drawback which results as a solution to the first effect is the loss of localized neutron population monitoring. SPND manufacturers will compensate for low signal current by increasing the total mass of the emitter material to increase the sensitivity of the device. Unfortunately, the increase in neutron reactive material mass causes the form factor of the SPND to increase. Due to self-shielding concerns, the diameter of the emitter is limited, thus mass of the emitter is modified by varying the length. This increase in length leads to loss of spatial neutron flux monitoring fidelity. Spiralizing the sensitive region of the SPND is a common method of mitigating the loss of spatial sensitivity as shown in Fig. 2-17, but requires a larger deployment location inside the reactor to accommodate the larger detector envelope.

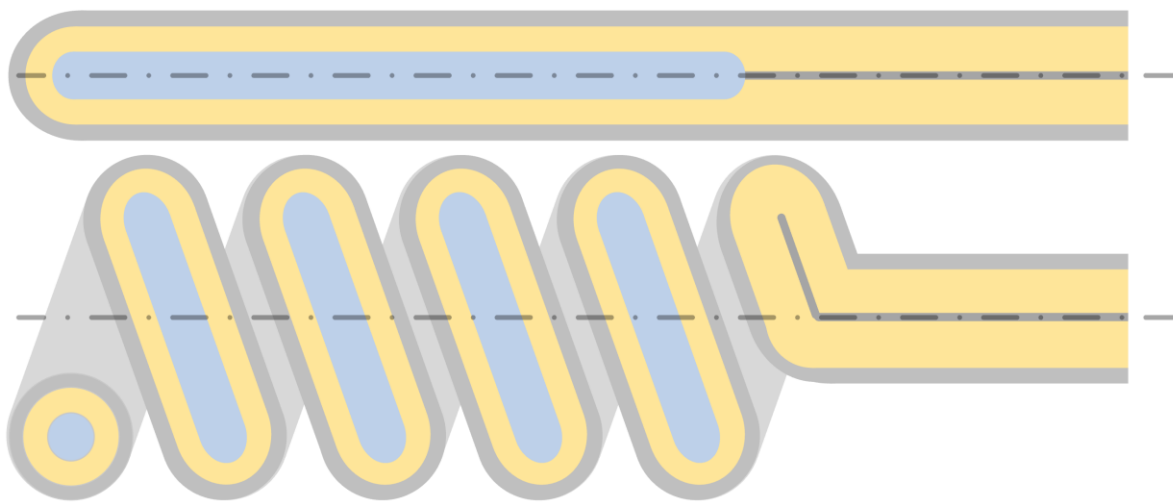


*Fig. 2-17. Diagram illustrating geometry differences between the standard design (top) and a spiralized compact-form design (bottom).*

### **2.5.2 Passive Methods**

Passive flux monitoring methods rely on a “cook and look” approach to determine radiation field conditions. Neutron reactive materials are first inserted into a radiation field for a specific duration of time and then removed. Once removed, the device is measured through various forms of interrogation such as gamma-ray spectrometry, optical stimulation, thermal stimulation, and film development. These devices rely on exposure to the radiation field that either induces damage in the material, excites electrons, or transmutes material into new isotopes. Passive flux monitoring methods are sensitive to both gamma rays and neutron but have differing interaction mechanisms,

and thus can be distinguished via the use of filter material or through different processing techniques after exposure. Passive methods operate as integral dosimetry which can be used to determine average neutron flux given the known duration of exposure and steady radiation field magnitude conditions. If a variety of materials with different interaction mechanisms and response characteristics are used, the passive dosimetry can be used to determine additional information such as the energy spectrum of the radiation field.

### Neutron Activation Analysis

Neutron activation analysis is based on the process of neutron absorption by material as outlined in Section 2.2.3. To determine the neutron flux inside a nuclear reactor, a small mass of material (typically < 10 mg) is inserted into or near the reactor core to absorb or be excited by neutrons. These materials often come in the form of a foil or wire and are known as activation dosimetry. Neutron bombardment of the matter has a high probability of transmuted the target atoms into new radioactive isotopes. Various interaction types are highly dependent on the incident neutron energy and thus can be used to determine information regarding the neutron energy spectrum of the reactor environment. To perform neutron activation analysis (NAA), the target atoms must transmute into a radioactive isotope which releases characteristic gamma rays. Reactions such as (n,2n), (n,n') and (n,α) can also be used to determine the epithermal and fast portions of the neutron energy spectrum, as these reactions prominently occur when target atoms interact with higher energy neutrons. To determine the thermal neutron flux magnitude, Eq. (6) & (7) can be coupled with information about the activation dosimetry. Fig. 2-18 illustrates the buildup and decay of the daughter isotope with components of the time factor,  $T$ .

$$\varphi_{th} = \frac{CA\lambda}{N_a \bar{\sigma}_a \mu m p f \eta} T^{-1} \quad (6)$$

$$T = (1 - e^{-\lambda t_0}) (e^{-\lambda(t_a - t_0)}) (1 - e^{-\lambda(t_b - t_a)}) \quad (7)$$

$C$  = counts under energy peak during measurement

$A$  = atomic mass of sample

$\lambda$  = decay constant of daughter isotope

$N_a$  = Avogadro's number ( $6.022 \times 10^{23}$  atoms/mol)

$\bar{\sigma}_a$  = avg. thermal neutron cross section

$m$  = mass of sample

$p$  = natural abundance of target isotope

$f$  = branching ratio of specific gamma ray

$\eta$  = detector efficiency

$T$  = time factor

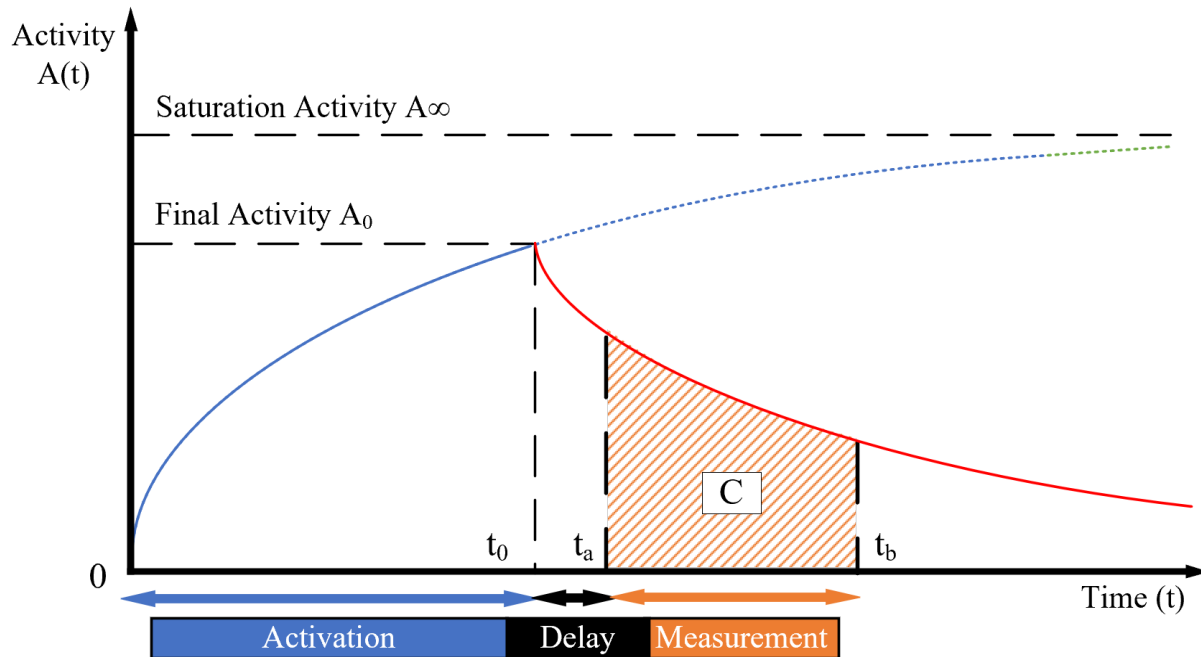


*Fig. 2-18. Build-up activation of materials during neutron irradiation and subsequent decay of activity. Note that given sufficient time, the material will reach saturation activity.*

Eq. (6) & (7) can be used to determine the approximate thermal neutron flux with a few key assumptions. The mass and form factor of the sample needs to be sufficiently small to avoid self-absorption, and the reactor power must remain constant during the duration of the irradiation. This method operates as an integral dosimeter and therefore will only be able to determine the average neutron flux magnitude within the irradiation duration. Minimum mass thresholds for elements to use NAA is listed in Table 2-5.

Table 2-5. Mass sensitivity limits for various elements using NAA. Note that these values are generalized for natural isotopic abundances.

| El. | Min. Mass<br>( $\mu\text{g}$ ) | El. | Min. Mass<br>( $\mu\text{g}$ ) | El. | Min. Mass<br>( $\mu\text{g}$ ) | El. | Min. Mass<br>( $\mu\text{g}$ ) |
|-----|--------------------------------|-----|--------------------------------|-----|--------------------------------|-----|--------------------------------|
| Ag  | $1 \times 10^{-4}$             | Eu  | $9 \times 10^{-7}$             | Mo  | $1 \times 10^{-2}$             | Sm  | $9 \times 10^{-5}$             |
| Al  | $1 \times 10^{-3}$             | Fe  | $1 \times 10^{-1}$             | Na  | $4 \times 10^{-4}$             | Sn  | $2 \times 10^{-2}$             |
| Ar  | $1 \times 10^{-4}$             | F   | $2 \times 10^{-1}$             | Nb  | $1 \times 10^{-1}$             | Sr  | $9 \times 10^{-4}$             |
| As  | $2 \times 10^{-4}$             | Ga  | $4 \times 10^{-3}$             | Nd  | $2 \times 10^{-2}$             | Ta  | $9 \times 10^{-3}$             |
| Au  | $2 \times 10^{-4}$             | Gd  | $2 \times 10^{-3}$             | Ne  | $4 \times 10^{-1}$             | Tb  | $5 \times 10^{-3}$             |
| Ba  | $5 \times 10^{-3}$             | Ge  | $5 \times 10^{-5}$             | Ni  | $4 \times 10^{-3}$             | Te  | $1 \times 10^{-2}$             |
| Bi  | $5 \times 10^{-2}$             | Hf  | $7 \times 10^{-3}$             | Os  | $4 \times 10^{-3}$             | Th  | $7 \times 10^{-3}$             |
| Br  | $9 \times 10^{-3}$             | Hg  | $2 \times 10^{-4}$             | Pd  | $2 \times 10^{-4}$             | Ti  | $1 \times 10^{-3}$             |
| Ca  | $5 \times 10^{-1}$             | Ho  | $4 \times 10^{-5}$             | Pr  | $1 \times 10^{-4}$             | Tl  | $5 \times 10^{-1}$             |
| Cd  | $9 \times 10^{-3}$             | I   | $1 \times 10^{-3}$             | Pt  | $4 \times 10^{-3}$             | Tm  | $2 \times 10^{-3}$             |
| Ce  | $2 \times 10^{-2}$             | In  | $5 \times 10^{-6}$             | Rb  | $4 \times 10^{-2}$             | U   | $4 \times 10^{-4}$             |
| Cl  | $2 \times 10^{-3}$             | Ir  | $2 \times 10^{-5}$             | Re  | $4 \times 10^{-5}$             | V   | $1 \times 10^{-4}$             |
| Co  | $9 \times 10^{-4}$             | K   | $2 \times 10^{-3}$             | Rh  | $2 \times 10^{-5}$             | W   | $2 \times 10^{-4}$             |
| Cr  | $2 \times 10^{-1}$             | Kr  | $1 \times 10^{-3}$             | Ru  | $2 \times 10^{-3}$             | Xe  | $1 \times 10^{-2}$             |
| Cs  | $4 \times 10^{-4}$             | La  | $4 \times 10^{-4}$             | Sb  | $9 \times 10^{-4}$             | Y   | $2 \times 10^{-4}$             |
| Cu  | $2 \times 10^{-3}$             | Lu  | $9 \times 10^{-6}$             | Sc  | $2 \times 10^{-3}$             | Yb  | $4 \times 10^{-4}$             |
| Dy  | $4 \times 10^{-8}$             | Mg  | $5 \times 10^{-2}$             | Se  | $2 \times 10^{-2}$             | Zn  | $2 \times 10^{-2}$             |
| Er  | $4 \times 10^{-3}$             | Mn  | $5 \times 10^{-6}$             | Si  | $9 \times 10^{-4}$             | Zr  | $2 \times 10^{-1}$             |

Source: Data from Shultis 2008 pg. 443 [1]

### Thermoluminescent Dosimetry

Thermoluminescent dosimeters (TLD) are an entire class of radiation detectors that record radiation dose and utilize materials such as LiF and  $\text{Li}_2\text{B}_4\text{O}_7$ . As radiation interacts in a TLD, electrons are excited into the conduction band and then de-excite much like the scintillation process, but instead of emitting light, these electrons fall into energy trap sites in the band gap of the material and remain trapped until thermal stimulation. This process allows for an integral dose record of the radiation environment the TLD experienced, which can then be readout after the irradiation via heating the chip and measuring the light signal it emits as the trapped electrons de-excite back into the valence band. For neutron-flux monitoring purposes, a pair of TLDs such as the Harshaw<sup>TM</sup> TLD-600s and TLD-700s, are commonly irradiated simultaneously. TLD-600s are LiF based chips which incorporate 96.5% enriched  $^6\text{Li}$  during manufacturing and have a higher neutron sensitivity of the natural enrichment TLD-700s. After irradiation, the signal difference between the TLD-700 can be subtracted from the TLD-600 to determine the neutron dose [12].

## Periodic Dosimetry (Aeroballs)

Aeroballs are high vanadium content steel spheres that are deployed into interstitial reactor locations during operation via a gas deployment tube. Periodically, the series of spheres are expelled from the core via a N<sub>2</sub> carrier gas at high pressure and then pushed into a counting tray with a line of congruent gamma-ray detectors which perform NAA on each sphere. A complete circuit of irradiation, expulsion, counting and return to reactor can occur within 15 minutes, with irradiation durations typically spanning 3 minutes. An illustration of the deployment setup is shown in Fig. 2-19. The Aeroballs allow for experimenters and reactor operators to profile the entire length of the fuel region to determine both the magnitude and distribution of the axial neutron flux. The Aeroballs have a 1.7 mm diameter and the shroud tube has a diameter of 6 mm [66], [67]. One drawback of the Aeroball platform is the residual signal from previous irradiations. Because <sup>51</sup>V has a 3.75-minute half-life (see Table 2-4), and the duty cycle of the detection system is 15 minutes, 6.25% of the previous irradiation signal will still exist within the consecutive measurement, causing a compounding error if not considered and compensated.

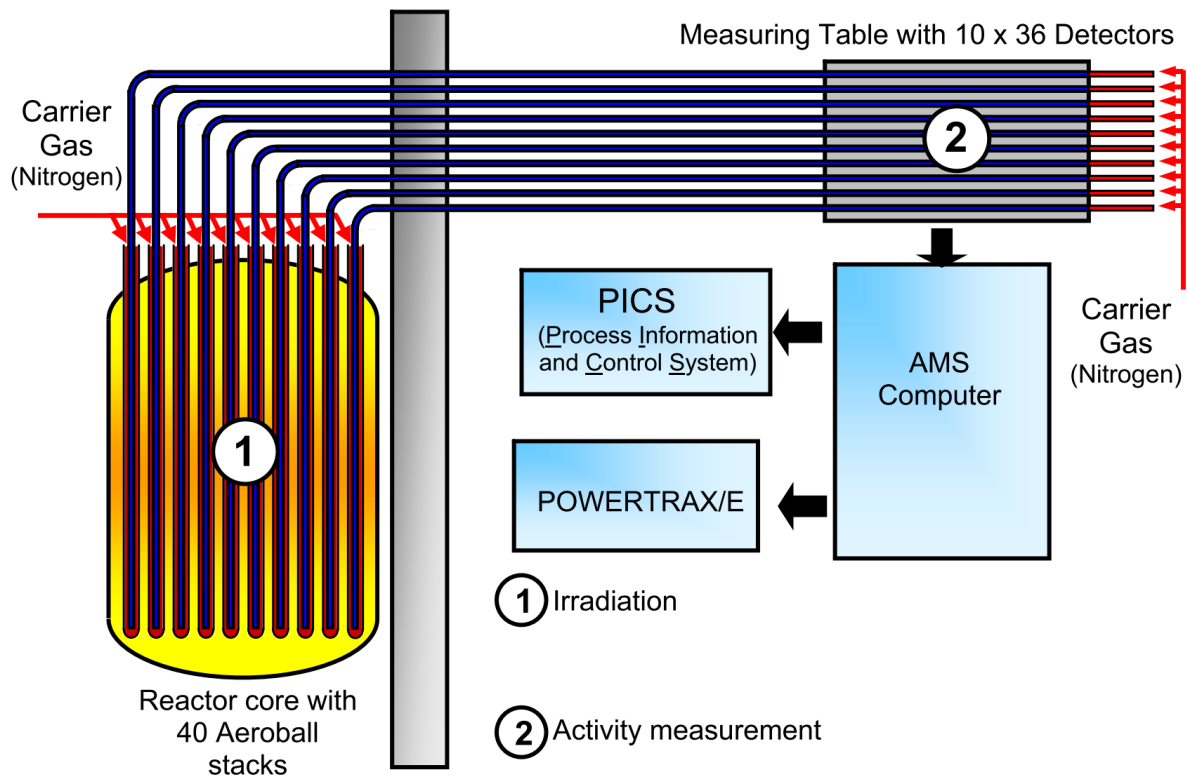


Fig. 2-19. Diagram illustrating Aeroball deployment and measurement setup [53].

A summary of simulation work to determine the MPFD radiation sensitivity will be provided. Simulation work to determine the MPFD signal response for deployment in a research reactor will also be provided. Additional discussion will focus on material selection and design changes to enhance MPFD performance.

### 3.1 Device Design Characteristics

The MPFD platform has been under development with numerous modifications since 2005. Manufacturing, assembly, geometry, and even operating parameters have changed, but the fundamental operating principles have remained the same [10], [42], [68]. Additionally, the core characteristics that make MPFDs novel, such as the small form factor, simplistic design, and spatially discreet flux monitoring capabilities, have not changed during its ongoing development. Changes to the design primarily revolve around optimizing already defined characteristics and reducing impact of drawbacks in the platform.

#### 3.1.1 Detector Operating Principles

An MPFD, like traditional fission chambers, is a gas-filled radiation detector that incorporates a fissile material layer as a neutron reactive coating. The chamber contains at least two electrodes, an anode and cathode, with an applied voltage between them that forms an electric field. When a neutron enters the chamber and is absorbed by the fissile material, a (n,f) reaction can occur resulting in subsequent emission of fission fragments, gamma rays, neutrons and other charge particles (such as alpha and beta particles; refer to Section 2.3). For detection purposes in this application, only fission fragments are desirable, and all other particles contribute to background signals. There is a significant probability that one of the two fission fragments will leave the fissile layer and deposit a fraction of its kinetic energy in the chamber gas by ionization. Charge carriers such as electrons and ions are produced as the fission fragment ionizes nearby gas molecules while traversing the chamber. The electrons and ions are then mobilized under the influence of the electric field and drift towards opposing polarity electrodes (positively charged ions to the cathode; electrons and negatively charged ions to the anode). This drift of charges

induces current across the anode-cathode circuit due to image charge development on the surface of the corresponding electrodes. This process is conceptually represented in Fig. 3-1.

Complementary electronics supply voltage to the anode-cathode pair to produce the electrical field in the chamber. These electronics are also used to measure the current produced in the MPFD chamber directly (current-sensitive electronics) or capacitively (voltage-sensitive electronics). Throughout this work, the designation of pulse-mode electronics will be used to indicate the usage of voltage-sensitive electronics, whereas the designation of current-mode electronics will refer to the use of current-sensitive electronics. Based on the magnitude and frequency of changes in the current produced in the chamber, experimenters can deduce that a neutron-induced fission event has occurred. Ultimately, this process allows the MPFD to detect the number of neutrons within a given area of space and time. This quantity of neutrons per area per time [ $\text{n cm}^{-2} \text{s}^{-1}$ ] is known as flux and is an important parameter to observe for experimental purposes and safe operations of nuclear reactors. Additional information about the neutron flux, as well as other forms of radiation, are provided in Section 2.3.

Due to the small chamber geometry, the entire process, from fission to electron collection occurs within a relatively small amount of time ( $< 200 \text{ ns}$  for standard chamber geometries and operating parameters) [69]. The same ionization process can occur for the other fission by-products such as gamma rays, alpha particles and beta particles, but due to the small size of the chamber, these other particles deposit significantly less energy (refer to Section 3.1.2 for further detail). This difference is another advantage of the MPFD design and allows for preferential detection of neutron-induced events.

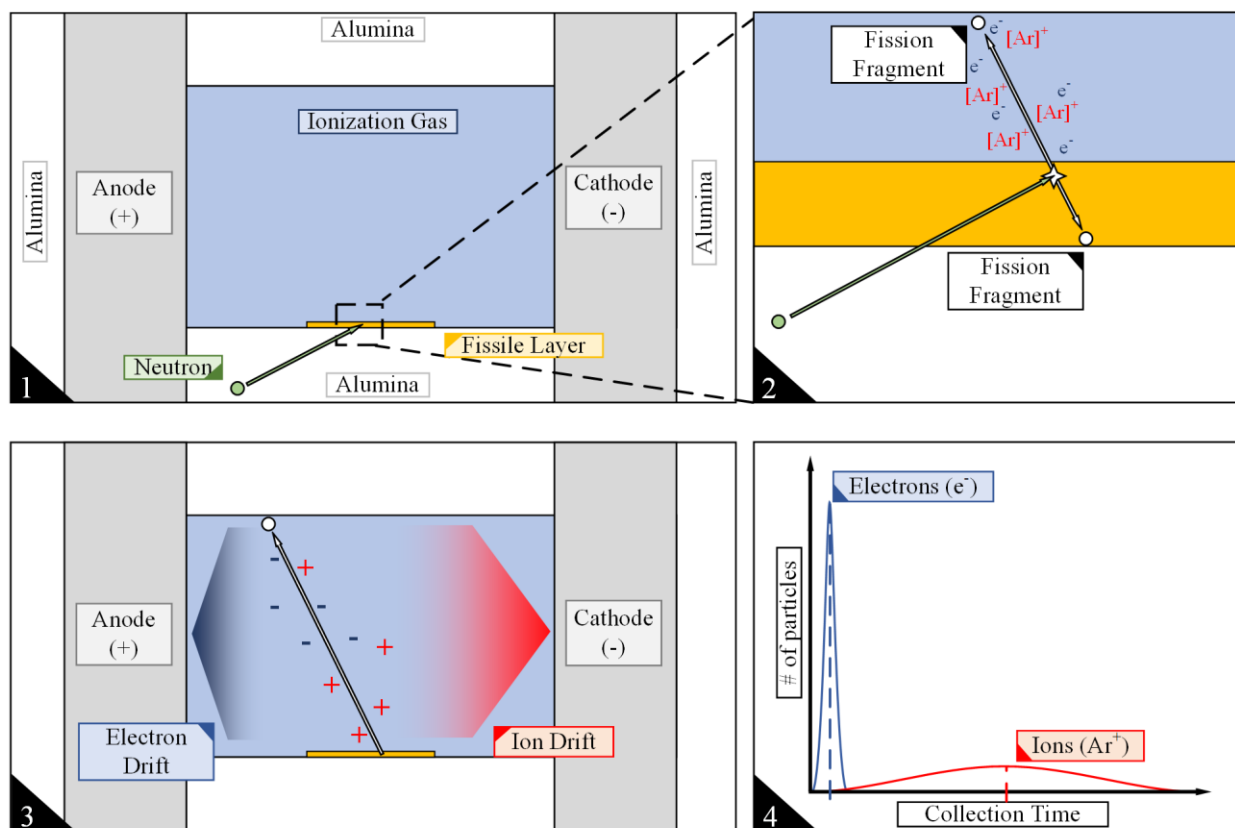


Fig. 3-1. (1) A neutron enters the detector and causes a fission event in the fissile layer. (2) As the nucleus splits, the fission fragments are commonly ejected in opposite directions. One of these fission fragments may escape the fissile layer and enter the ionization gas. (3) As the fission fragment traverses the chamber, it coulombically interacts with the gas molecules producing charge carriers (electrons and ions). These charge carriers, under the influence of the electric field between the anode and cathode, drift to the electrodes. (4) Graphical representation of the electron and ion collection time distributions.

### 3.1.2 Radiation Sensitivity

Simulations of the most current MPFD design were conducted to better understand the MPFD energy deposition sensitivity to various forms of radiations such as described in Section 2.3. For the purposes of these simulations, the following parameters (shown in Table 3-1) were used in a clover geometry chamber (discussed further in Section 3.3) and is shown in Fig. 3-2.

Table 3-1. Detector design parameters and simulation radiation characteristics.

| Dimensions                                 |                       |                                    |
|--------------------------------------------|-----------------------|------------------------------------|
| Chamber Height                             | 0.5                   | mm                                 |
| Chamber Volume                             | 0.41                  | mm <sup>3</sup>                    |
| Fissile Diameter                           | 250                   | μm                                 |
| Detector Composition                       |                       |                                    |
| Fissile Layer (U)                          | 93                    | <sup>235</sup> U % enr.            |
| Fissile Mass ( <sup>235</sup> U)           | 0.886                 | μg                                 |
| Chamber gas (Ar)                           | 60                    | psig                               |
| Radiation Conditions (700W <sub>th</sub> ) |                       |                                    |
| Thermal-Neutron Flux                       | $6.82 \times 10^8$    | n cm <sup>-2</sup> s <sup>-1</sup> |
| Fast-Neutron Flux                          | $2.90 \times 10^9$    | n cm <sup>-2</sup> s <sup>-1</sup> |
| Gamma-ray Flux                             | $1.13 \times 10^{10}$ | g cm <sup>-2</sup> s <sup>-1</sup> |

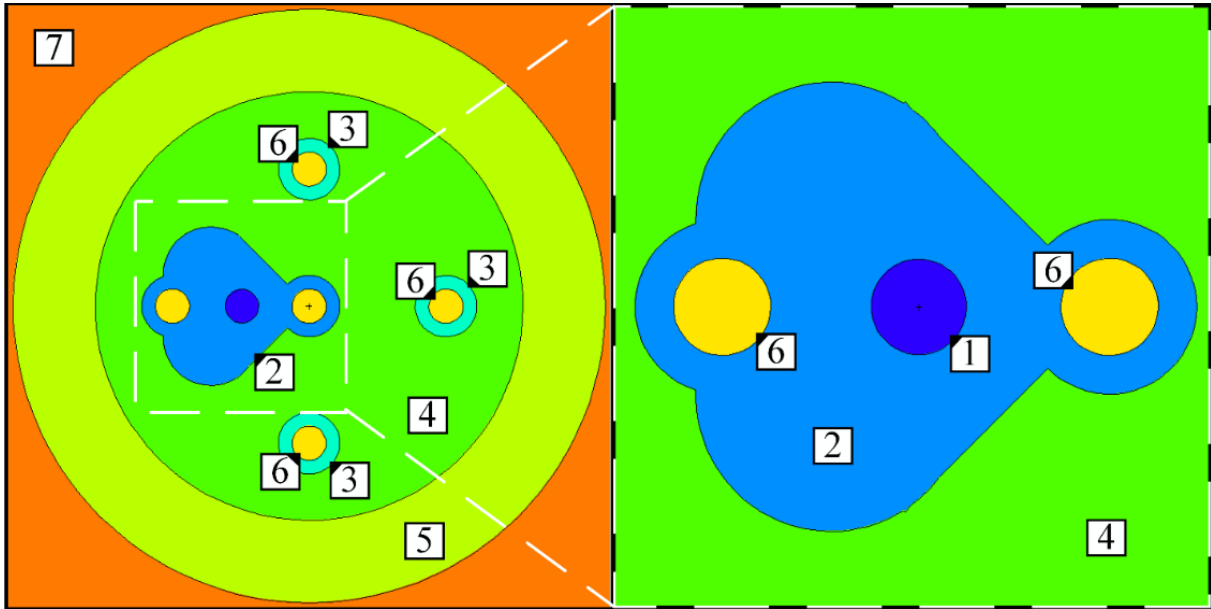


Fig. 3-2. MCNP model top-down view of a single MPFD chamber with fissile layer (1), argon gas (2, 3), alumina chamber piece (4), stainless steel sheath/wires (5, 6), surrounding water (7). The MPFD is simulated to be deployed in a water column with a height of 1 in and a diameter of 0.349 in.

While previous studies have been conducted for arbitrary gas volumes, a more systematic and geometry-driven approach was needed [70]. Of primary concern are alpha particles, beta particles, and gamma rays which are each produced by the nuclear reactor core, and inside the chamber as a fission by-product of the fissile layer. If these particles originate from outside the MPFD, they can cause ionization in the chamber gas that produce added induced current in the MPFD chamber,

which will add uncertainty the detector neutron response. Alpha particles, which have a relatively high specific energy deposition, have a relatively low production rate as a by-product of the fission process, whereas gamma rays and beta particles have a relatively low specific energy deposition but a higher production rate from fission fragments and products. These radiation particles add to the signal generated by fission fragments.

### **Alpha Particles**

Alpha particles are born as a fission by-product by two main methods. First, alpha particles can be generated at the initial fission event as ternary compounds that are ejected from the fractured nucleus and therefore share a considerable fraction (0-30 MeV) of the total kinetic energy imparted on the various fission fragments [71]. Commonly, fission is classified as a binary fracturing of the nucleus, given that most fission events occur with nucleons of the original atom splitting between two large fractional clusters. Ternary fission results in additional nucleon emission clusters such as protons, deuterons, tritons, and alpha particles, but are far less common. The second main mechanism of alpha-particle production is from alpha-particle decay of the fission fragment daughters. Fission fragments are commonly neutron-rich and decay by  $\beta^-$  emission [1], [72]. Although some decay chains with alpha-particle emission exist, they are far less common compared to  $\beta^-$  decay and are often times considered negligible when analyzing the whole of energy released during fission [73]. Additionally, alpha-particle production is limited to locations in the reactor with fissile material. Although MPFDs can be placed in proximity to nuclear fuel, the alpha particles produced inside the fuel cannot penetrate the fuel cladding, reactor moderator, or MPFD sheath to deposit energy in the MPFD chamber.

Alternatively, the fissile material used in the chamber for neutron sensitivity commonly emits alpha particles due to its own radioactive decay. Alpha-particle energies and intensities for the various uranium isotopes used for MPFD fabrication are included in Table 3-2 for reference.

Table 3-2. Summary of most common uranium isotopes with empirical equations for determining isotopic mass using  $^{235}\text{U}$  enrichment ( $0.007 < x < 0.93$ ). Each isotope is listed with the three highest emission intensity alpha particles. Alpha-particle emission is the primary decay mode, therefore fission decay is given for comparison [1], [34], [74].

| Isotope | Isotope Mass<br>$m_i = m_U * [ax^2 + bx + c]$ |         |         | Fission prob. Per<br>decay (%) | Specific Activity<br>(Bq $\mu\text{g}^{-1}$ ) | Energy<br>(MeV)         | Emission<br>Intensity  |
|---------|-----------------------------------------------|---------|---------|--------------------------------|-----------------------------------------------|-------------------------|------------------------|
|         | a                                             | b       | c       |                                |                                               |                         |                        |
| U-234   | 0.0053                                        | 0.0058  | 0       | $1.73 \times 10^{-9}$          | 230                                           | 4.603<br>4.722<br>4.774 | 0.2%<br>28.4%<br>71.4% |
| U-235   | 0                                             | 1       | 0       | $2.0 \times 10^{-7}$           | 0.08                                          | 4.215<br>4.364<br>4.395 | 6.0%<br>18.9%<br>57.7% |
| U-236   | 0.0004                                        | 0.0045  | 0.00002 | $9.6 \times 10^{-8}$           | 2.4                                           | 4.332<br>4.445<br>4.494 | 0.2%<br>26.0%<br>74.0% |
| U-238   | -0.0056                                       | -1.0103 | 1       | $5.4 \times 10^{-5}$           | 0.012                                         | 4.038<br>4.151<br>4.198 | 0.1%<br>21.0%<br>79.0% |

Alpha-particle emissions of uranium are therefore confined to a narrow window of expected energies. Simulations were performed to determine expected alpha-particle energy deposition in the chamber using these energies. The resulting energy deposition spectrum for alpha particles is shown in Fig. 3-3, with an average energy of deposition determined to be 301 keV. For the prescribed 93% enrichment uranium layer, the total activity was determined to be 2.228 Bq. Because each isotope primarily decays by alpha-particle emission, it is assumed that the fissile layer will emit 2.228 alpha particles each second, with only a fraction of those emitted entering the chamber gas. Using this activity, the interaction rate for alpha particles was determined to be 1.1 interactions/second after factoring in the solid angle correction.

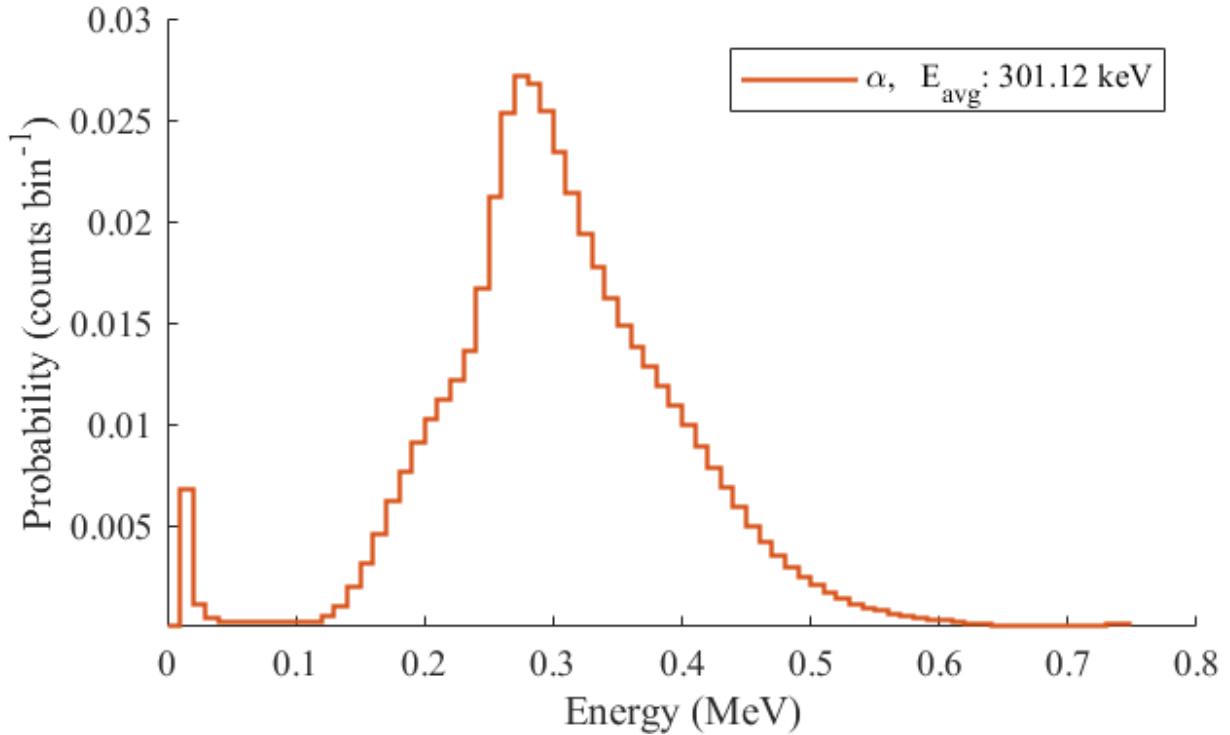


Fig. 3-3. Alpha-particle energy deposition spectrum for 93% enriched uranium coating with 2  $\mu\text{m}$  thickness.

### Beta Particles

The beta-particle population inside the reactor is primarily determined by the fission rate of the fuel ( $5.5 \beta^-/\text{fission}$  where  $E_\beta < 13 \text{ MeV}$ ), but other methods contribute to or degrade the flux spectrum. Neutron-induced activation of fission products can both increase and decrease certain energy regions of beta-particle energy spectrum ( $E_\beta < 3.54 \text{ MeV}$ ). Additionally, there is beta-particle production due to  $(n,\gamma)$  and  $(n,2n)$  reactions with heavy nuclei ( $E_\beta < 1.3 \text{ MeV}$ ) and neutron capture in structural materials of the reactor core ( $E_\beta < 2 \text{ MeV}$ ) or in water ( $E_\beta \approx 10 \text{ MeV}$ ) due to  $^{16}\text{O}(n,p)^{16}\text{N}$  reaction [75]. Due to the half-lives of many beta-particle emitters, the beta-particle flux approaches its asymptotic limit after 50 days of operations at steady state conditions with a production rate of  $6.6 \beta^-$  per fission.

Most of the in-core beta-particle population is sequestered to the inside of the reactor fuel where fission fragments and their daughter isotopes are retained inside the fuel meat. MPFDs are deployed congruent to fuel elements, but typically not in direct contact, therefore they are separated from the fuel meat of fuel elements by the external cladding and a small distance of moderator

(typically water). For example, TRIGA Mark II fuel elements have 0.02-in. of stainless-steel cladding, which would preclude beta particles with  $E_\beta < 1$  MeV from exiting the fuel. To conservatively estimate the energy deposition spectrum for beta particles in the MPFD, the raw beta-particle spectrum (shown in Fig. 3-4) without attenuation was utilized to develop an MCNP simulation.

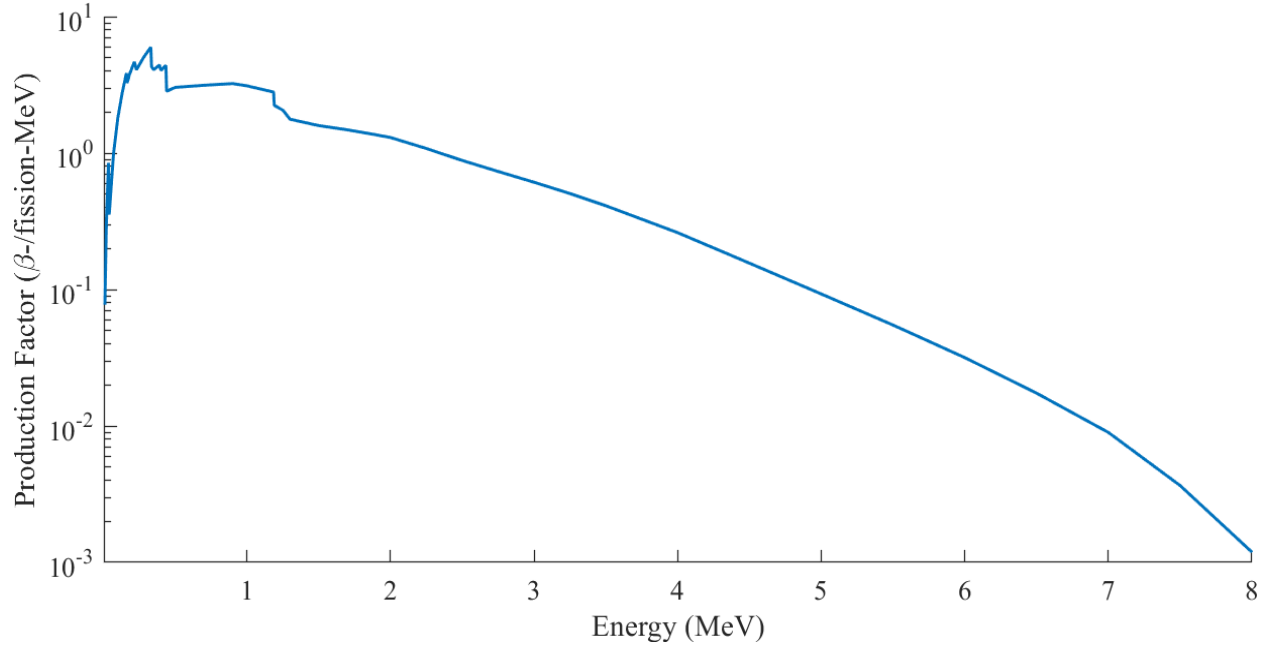


*Fig. 3-4. In-core beta-particle spectrum normalized to per fission and per energy [75].*

Based on the developed beta-particle source term, an energy deposition spectrum was produced and is shown in Fig. 3-5, with an average energy deposition of 1.1 keV. This result is normalized to the fission rate of the reactor. Assuming 700 W<sub>th</sub> reactor power for the ATRC core, the estimated scaling factor for the B-8 position fuel center plane was determined to be  $3.61 \times 10^9$ , which constitutes a total beta-particle interaction rate of  $1.4 \times 10^5$  s<sup>-1</sup>.

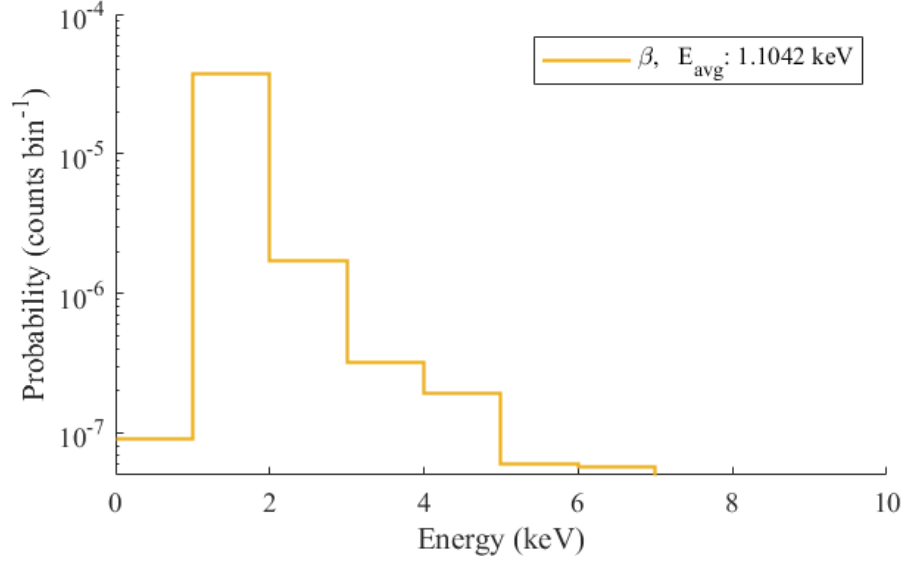


Fig. 3-5. Beta-particle energy deposition spectrum.

### Gamma Rays

The energy spectra of fission gamma rays, represented by an analytic solution and experimental data, is shown in Fig. 3-6. An analytical solution for gamma-ray energy spectra for thermal-neutron induced fission is provided in Eq. (8) and shown in Fig. 3-6 [76].

$$N(E) = \begin{cases} 38.13(E - 0.085)e^{1.648E} & E < 0.3 \text{ MeV} \\ 26.8e^{-2.30E} & 0.3 < E < 1.0 \text{ MeV} \\ 8.0e^{-1.10E} & 1.0 < E < 8.0 \text{ MeV} \end{cases} \quad (8)$$

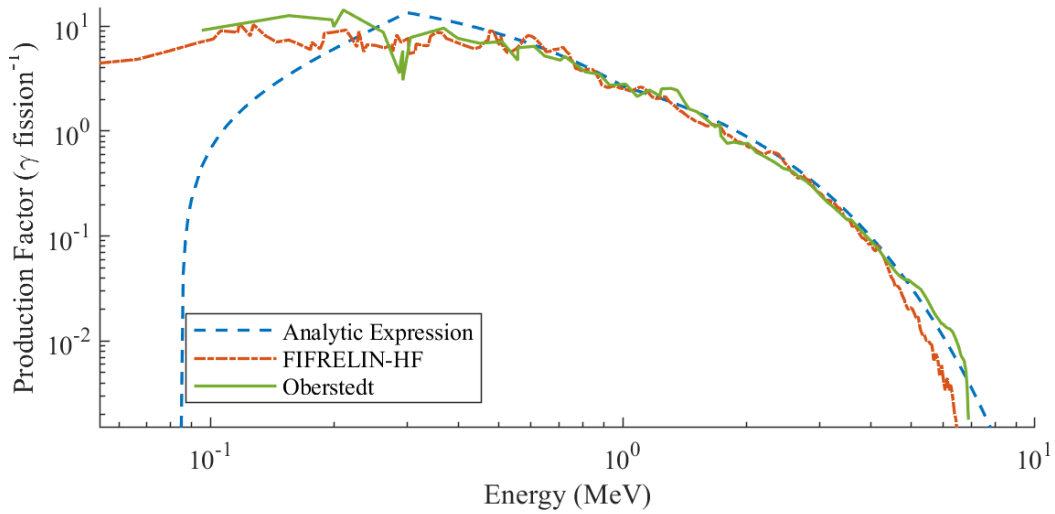


Fig. 3-6. Fission gamma-ray energy spectra including an analytical expression [77], [78].

Using the analytical solution provided above, a photon source term for MCNP was created and used to simulate energy deposition inside the MPFD chamber gas. Most of the gamma-ray flux is produced in the fuel meat of the reactor. Based on this simulation, the resulting energy deposition spectrum was produced (shown in Fig. 3-7) with an average energy deposition of approximately 1.0 keV. Using gamma-ray flux values for the ATRC B-8 position, the expected gamma-ray interaction rate is  $1.627 \times 10^7 \text{ s}^{-1}$ .

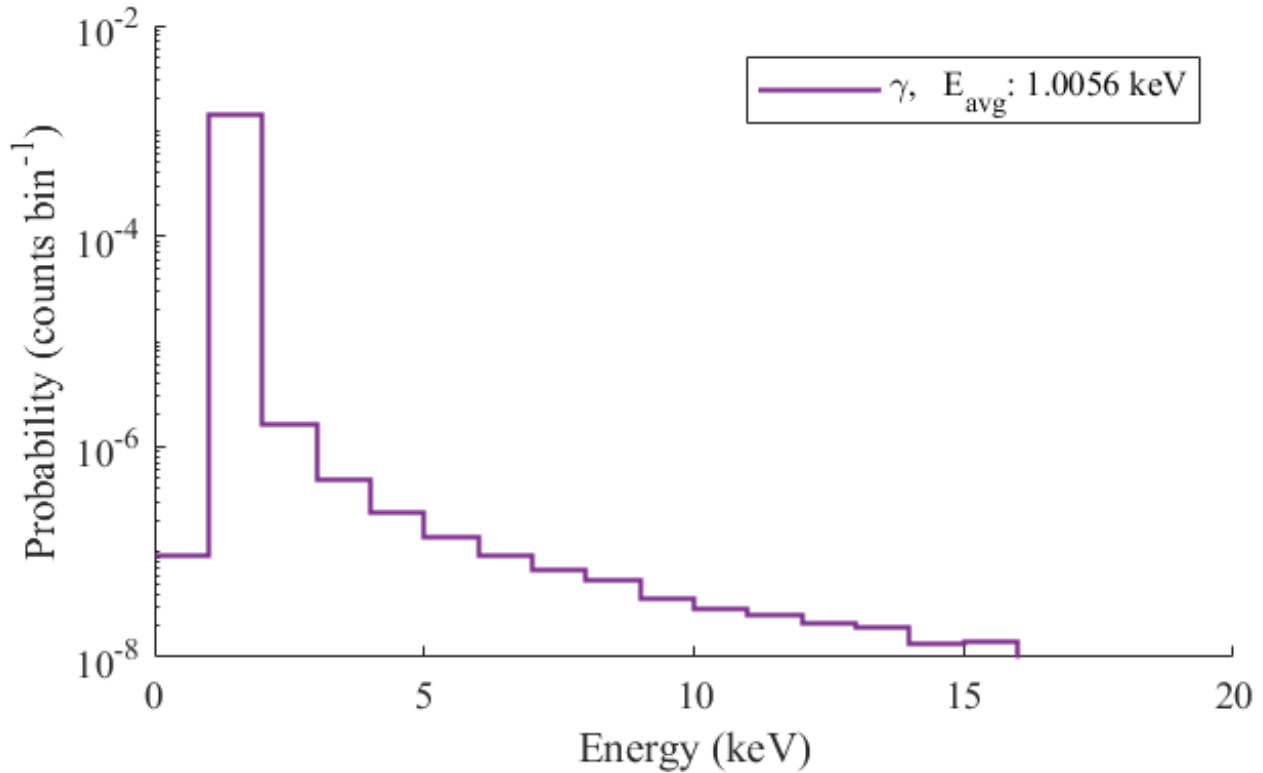


Fig. 3-7. Energy deposition spectrum for gamma rays incident on MPFD gas volume.

### Fission Fragments

The efficiency and efficacy of the MPFD platform is directly coupled to fission fragment energy deposition magnitude and rate. MPFDs, like most fission chambers, require a sufficient signal from neutron-induced fission events and the resulting fission fragments to detect neutron flux. If fission fragments deposit a comparable magnitude of energy and at a similar interaction rate as other forms of radiation, neutron-induced events will be almost impossible to distinguish amongst other radiation-inducing events. As such, it is paramount to the design of the MPFD that fission fragments (signal) versus other forms of radiation (background) have a high signal-to-noise

ratio both with interaction rate and energy/event characteristics. Due to the small sensitive volume of the MPFD, there is an inherent discrimination against other forms of radiation. To demonstrate this effect, a fission fragment energy deposition simulation was performed using MCNP6.2.

A fission fragment source input was developed for use in MCNP which initially incorporated only 34 heavy ions but was further expanded to a source input utilizing 505 unique heavy ions. MCNP6.2 presently has a limited library of heavy ions to simulate, therefore an algorithm was developed to cross correlate which ions are available for simulation and which isotopes have known fission product yields. The outcome results in a high-fidelity fission fragment model, with 505 isotopes (equating to 99.1% integral fission product yields), that can simulate the expected emission rates and energy depositions during MPFD operation. Fig. 3-8 is provided to show the cross correlation between fission product isotopes and the available ions in MCNP6.2. Fig. 3-9 shows the fission product yields for isotopes as identified in ENDF B/VIII.0 [72].

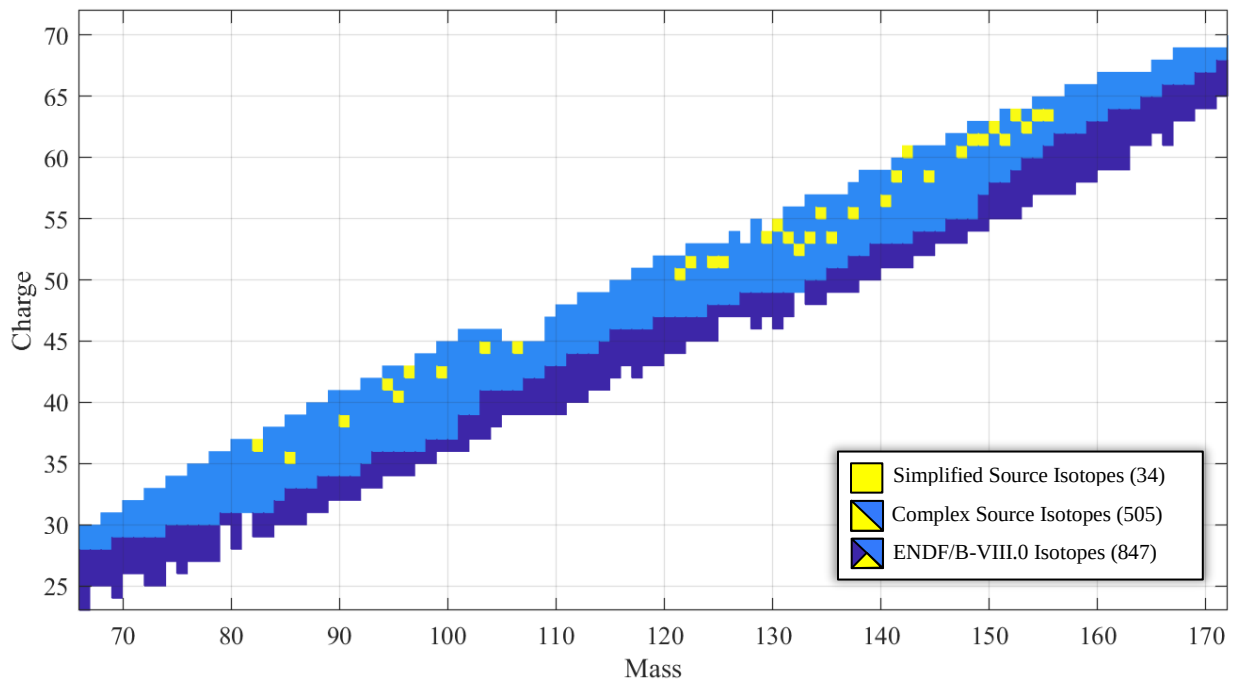


*Fig. 3-8. Graphical representation of heavy ion list used to simulate fission fragments. Note that MCNP6.2 has a limited heavy ion library, therefore an algorithm was written to cross-correlated available isotopes for simulation and isotopes with known fission yields.*

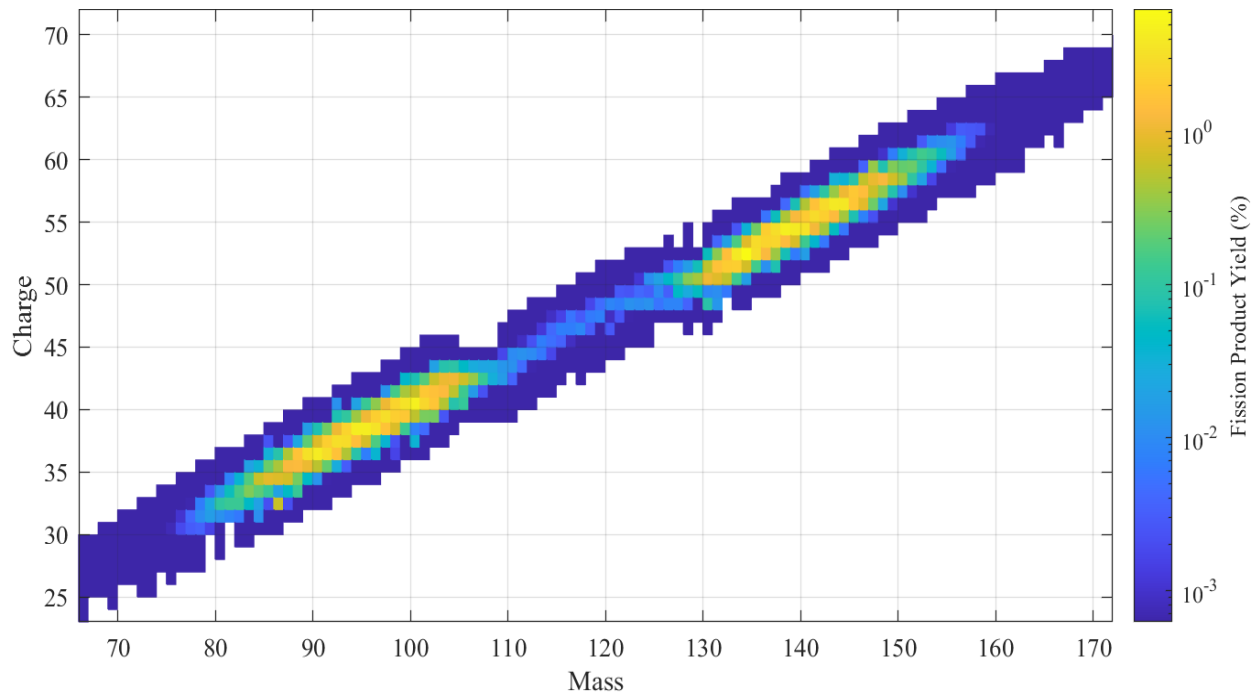


*Fig. 3-9. Raw ENDF B/VIII.0 fission-product yield data [72].*

Much like the other radiation particles listed in previous sections (gamma rays, alpha particles, and beta particles), fission fragments were simulated using the standard MPFD geometry, with final energy deposition spectrum shown in Fig. 3-10. These results show that the average energy deposition for fission fragments in the described chamber is 14.25 MeV. Additionally, the fission rate was determined to be  $5700 \text{ s}^{-1}$  during deployment in the B-8 position of the ATRC at  $700 \text{ W}_{\text{th}}$ .

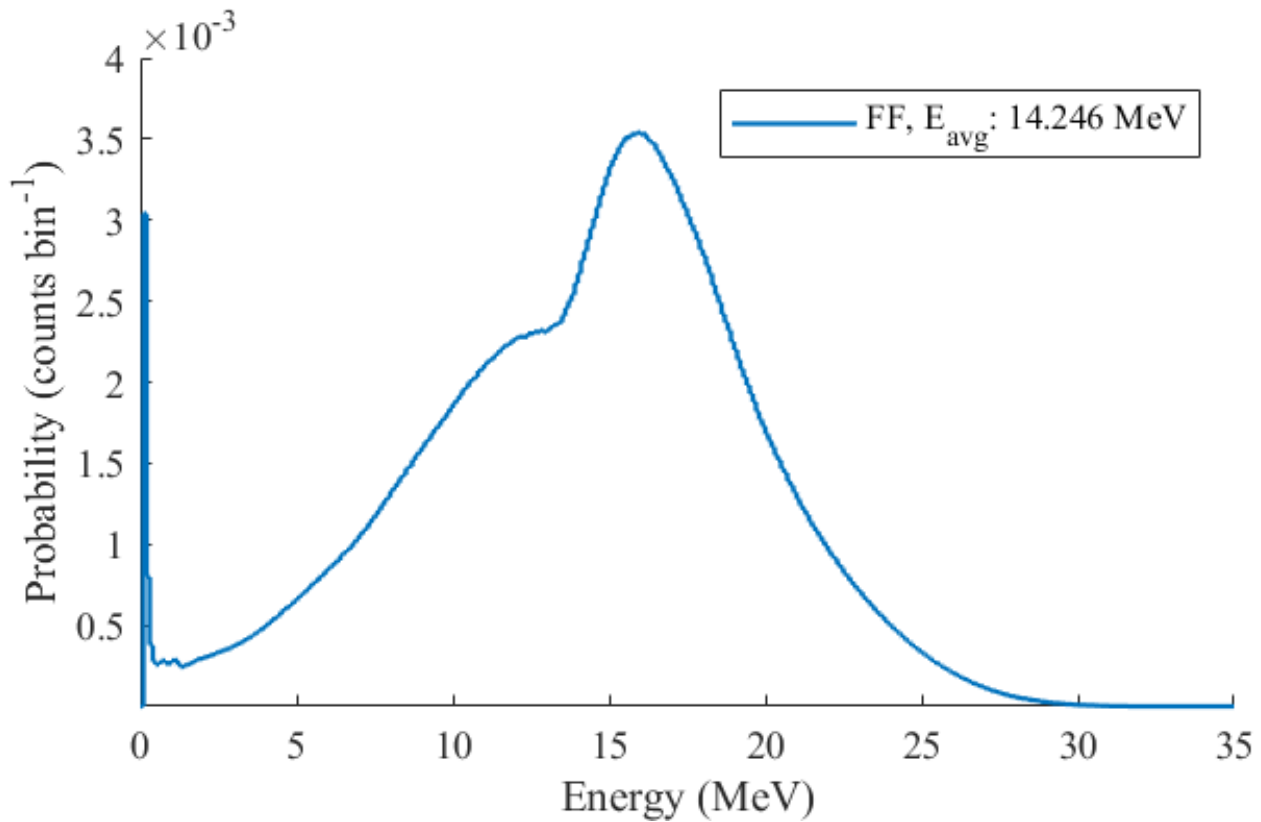


Fig. 3-10. Energy deposition spectrum for fission fragments in the MPFD gas volume.

### Summary Comparison

Various forms of radiation were simulated, both for internal and external influence on the MPFD gas volume. Based on the results described above, it was determined that fission fragments have a significant interaction rate ratio, and energy deposition rate in comparison to the other forms of radiation. These results are summarized for comparison in Table 3-3, with spectra comparisons shown in Fig. 3-11.

Table 3-3. Summary of average deposited energy, interaction rate, and comparison values.

|                          | Energy<br>(keV) | Interaction<br>Rate (s <sup>-1</sup> ) | FF Energy<br>Ratio | FF Interaction<br>Ratio | Energy Deposition<br>Rate (MeV s <sup>-1</sup> ) |
|--------------------------|-----------------|----------------------------------------|--------------------|-------------------------|--------------------------------------------------|
| <i>fission fragments</i> | 14,246          | $5.7 \times 10^3$                      | -                  | -                       | 81,202                                           |
| <i>α particles</i>       | 301.12          | 1.10                                   | 47.3               | 5182                    | 0.33                                             |
| <i>β particles*</i>      | 1.1042          | $1.40 \times 10^5$                     | $1.30 \times 10^4$ | $4.07 \times 10^{-2}$   | 154                                              |
| <i>γ rays</i>            | 1.0056          | $1.63 \times 10^7$                     | $1.42 \times 10^4$ | $3.50 \times 10^{-4}$   | 16,270                                           |

\* This is a maximum estimate due to the fuel congruency assumption, therefore, practical values should be lower.

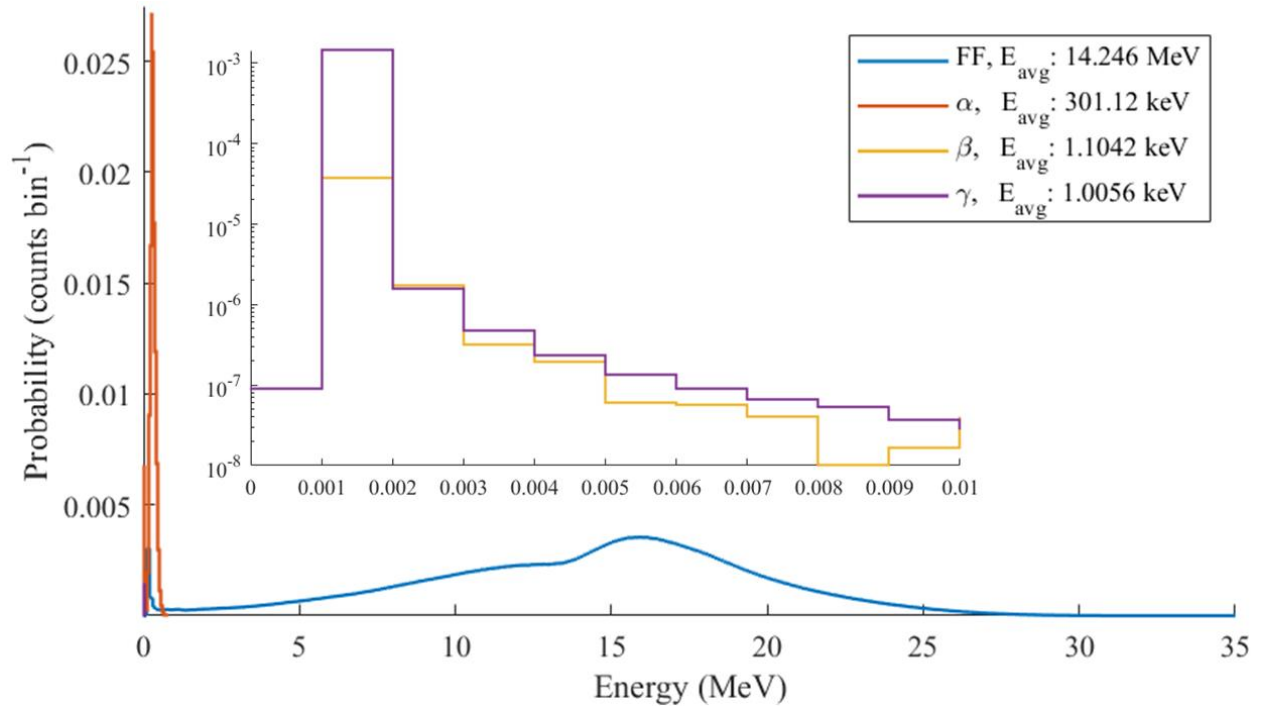


Fig. 3-11. Plot comparing energy deposition spectra for each of the particle types with the average energy deposition for each particle noted.

### 3.1.3 Estimated Flux Response

In connection to the radiation sensitivity study discussed in the previous section, an estimate of the MPFD flux response was determined by simulations for deployment in the Advanced Reactor Critical Facility (ATRC) core. For simulation purposes, the MPFD was modeled in the B-8 irradiation port which is situated between the outside of the core boundary and the hafnium outer shim control cylinders (OSCC). In this location, an MCNP k-code model was used to tally the neutron and gamma-ray flux spectra for 69 axial tally zones as depicted in Fig. 3-12.

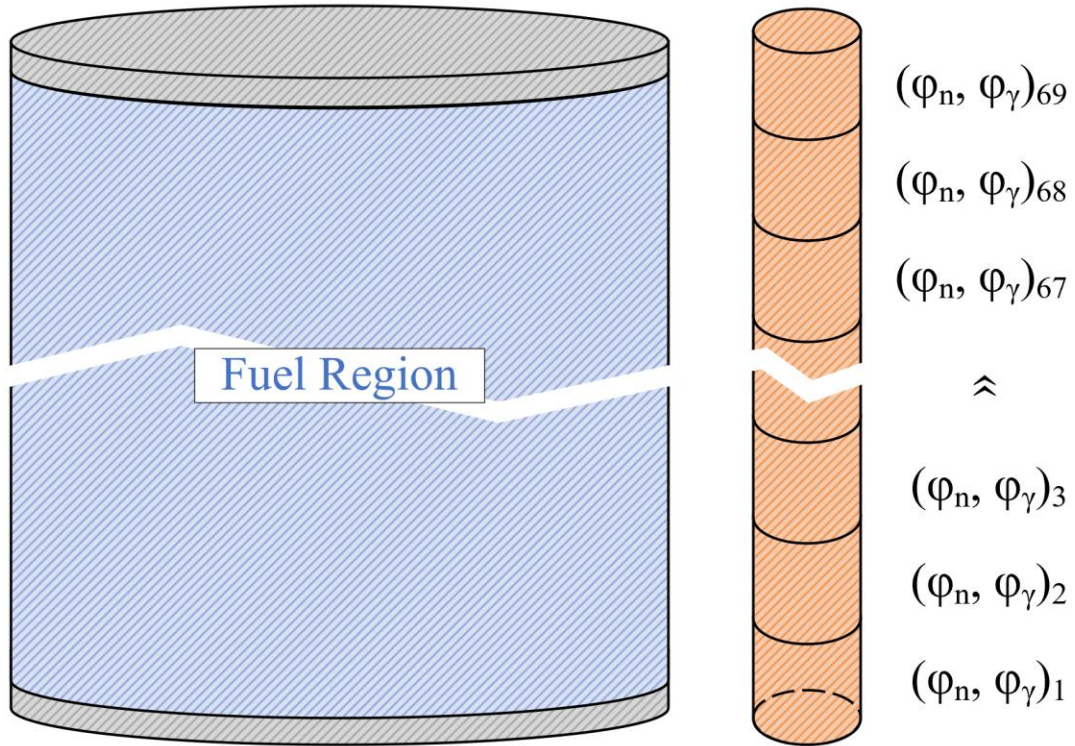


*Fig. 3-12. Diagram depicting the series of gamma-ray and neutron flux tallies performed to simulation detector performance at various core altitudes [79].*

These tallies spanned the length of the fuel region and extend into the top and bottom beryllium reflector portions of the core. An example of the resulting neutron and gamma-ray flux spectra for the fuel center plane are shown in Fig. 3-13 and Fig. 3-14. Using these flux spectra as source inputs, an MCNP model was developed for the proposed MPFD geometry to determine the expected fission and gamma ray interaction rate. The MCNP model geometry is shown in Fig. 3-2.

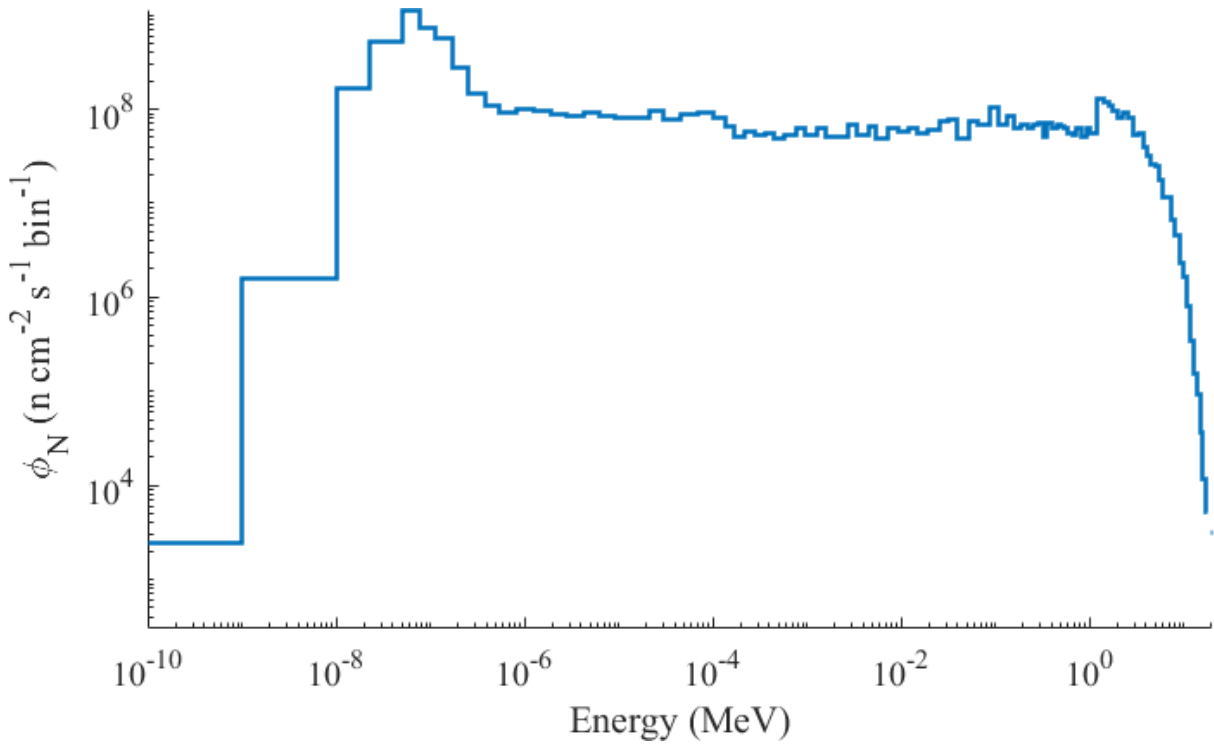


Fig. 3-13. 140-group integral neutron energy spectrum from ATRC B-8 position at fuel center plane for a power level of  $700 W_{th}$ .

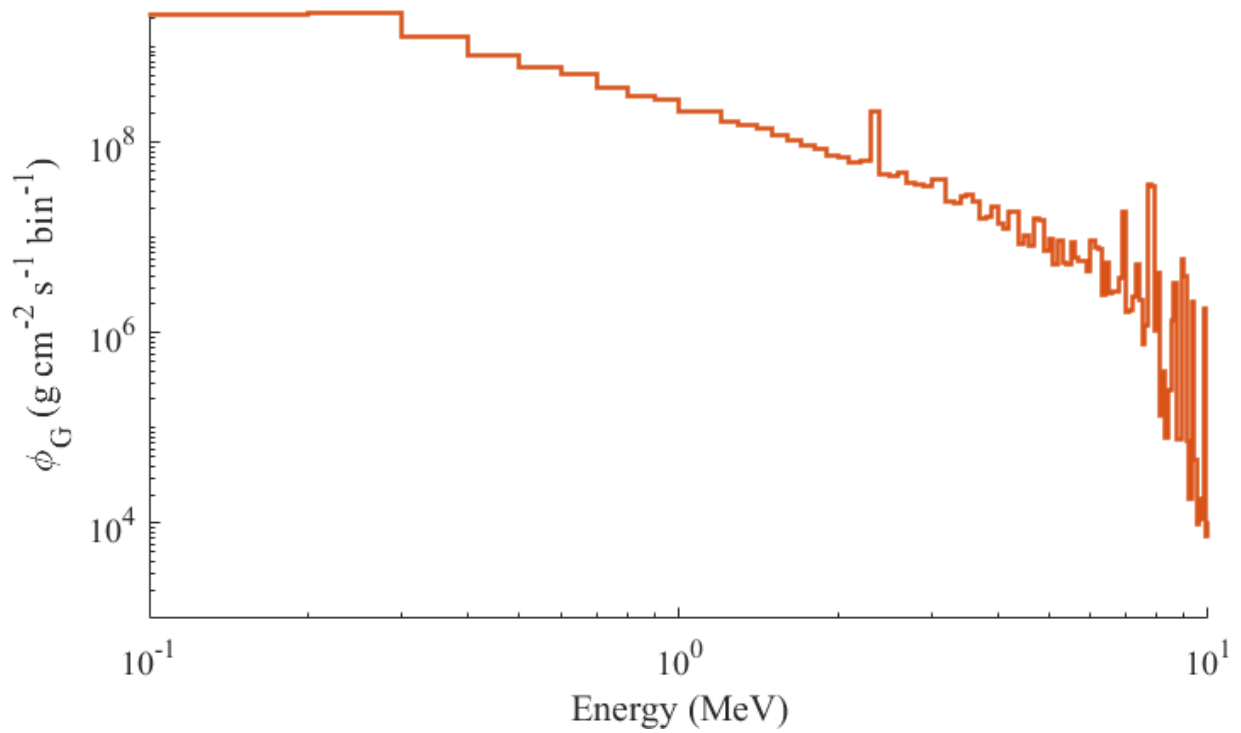


Fig. 3-14. 99-group integral gamma-ray energy spectrum from the same ATRC B-8 position.

The interaction rate results using the previously mentioned model were then used to determine the expected detector response by simulating individual fission reaction events and tracking the heavy ions (fission fragments) produced as a result. By utilizing the 69 axial neutron tallies, an expected axial neutron flux profile was determined as a function of nearby drum angle positioning and is shown in Fig. 3-15.

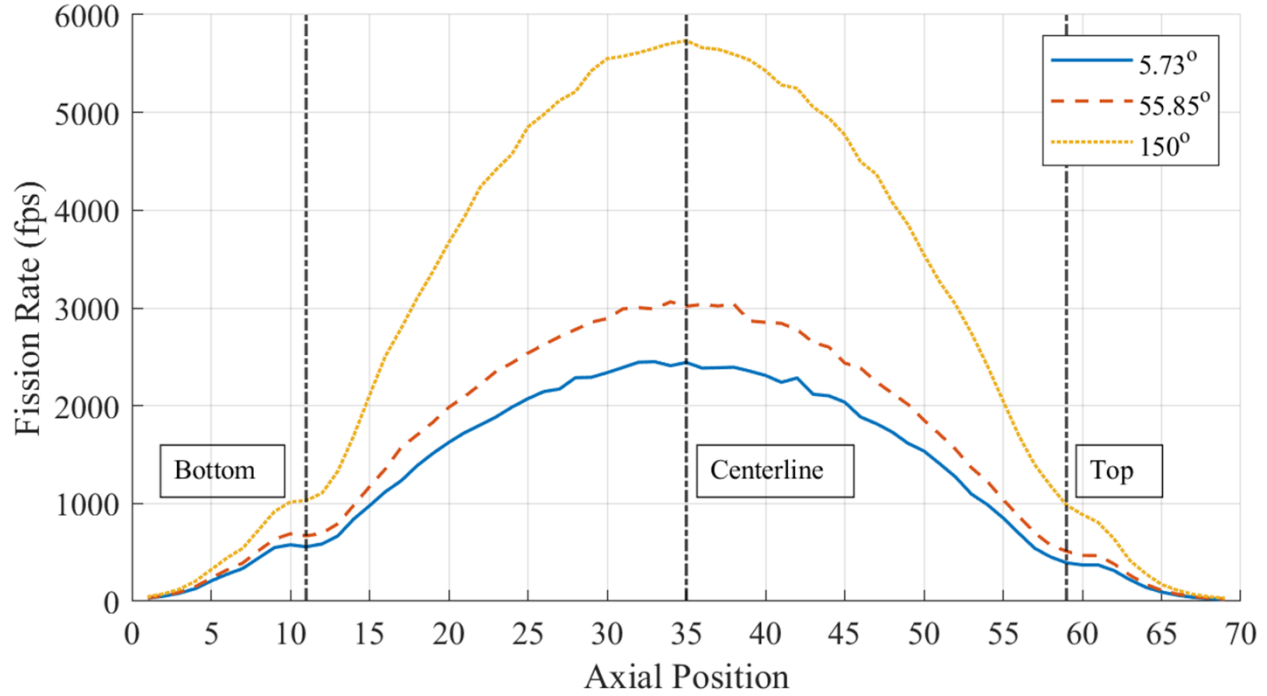


Fig. 3-15. Estimated axial fission rate of the MPFD during deployment in the ATRC B-8 position for various OSCC positions.

The goal of this project was to determine the impact on the localized neutron flux by the nearby OSCC. The OSCCs act as control drums which can be used to shim the reactor power or tilt the flux magnitude in the core. This simulation shows that as the OSCC is rotated away from the B-8 position in the ATRC core, the localized neutron flux spectrum shifts to a thermalized spectrum. At the fuel centerline for an OSCC angle of 150°, the neutron sensitivity of the MPFD was determined to be  $6.47 \times 10^{-7} \text{ cm}^2$ .

## 3.2 Construction Material Selection

As described in Section 2.3.4, material selection is important for the MPFD design for the neutron activation, electrical conductivity, and structure. During this research process, MPFDs

were primarily deployed in non-volatile reactor environments with open pool designs (MITR, UWNR, KSU TRIGA, TREAT).

### 3.2.1 Electrode Wire and Sheath Selection

As part of the construction of the MPFD, technical specification analysis of the electrode and sheath materials was conducted to determine thermal, mechanical, electrical characteristics, and radiation interaction effects. Traditional materials for MPFD construction have included stainless steel 316 (SS316) and Titanium CP2 for the sheath, and Alumel, Chromel, SS316 and Ti-6Al-4V alloy for the signal wires. Inconel-600 is included for consideration as the sheath material due to its prolific use throughout the nuclear industry for both reactor structural components and instrumentation. A summary of the material characteristics is provided in Table 3-4.

*Table 3-4. Summary of sheath and wire material characteristics of interest.*

| <b>Wires &gt;</b> |                               | Alumel | Chromel | Ti6Al4V | SS316 | Ti-CP2 | Inconel-600 | <b>&lt; Sheath</b>                 |
|-------------------|-------------------------------|--------|---------|---------|-------|--------|-------------|------------------------------------|
| Therm.            | <i>Specific Heat</i>          | 523    | 448     | 526     | 500   | 523    | 444         | J kg <sup>-1</sup> K <sup>-1</sup> |
|                   | <i>Conductivity</i>           | 29.7   | 19.2    | 6.7     | 16.3  | 16.4   | 14.9        | W m <sup>-1</sup> K <sup>-1</sup>  |
|                   | <i>Melting Point</i>          | 1260   | 1430    | 1604    | 1370  | 1660   | 1354        | °C                                 |
|                   | <i>CTE</i>                    | 12     | 13.1    | 8.6     | 16    | 8.6    | 13.3        | 10 <sup>-6</sup> K <sup>-1</sup>   |
| Mech.             | <i>Density</i>                | 8.6    | 8.7     | 4.4     | 8     | 4.5    | 8.5         | g cm <sup>-3</sup>                 |
|                   | <i>Hardness-Vickers</i>       | 130    | 150     | 349     | 155   | 280    | 196         | MPa                                |
|                   | <i>Yield Tensile Strength</i> | 586    | 655     | 880     | 240   | 450    | 310         | MPa                                |
| Elec.             | <i>Resistivity</i>            | 0.29   | 0.71    | 1.78    | 0.74  | 0.54   | 1.03        | 10 <sup>-4</sup> ohm-cm            |

Material properties can vary by manufacturer, thus the values provided are used for general comparison.

### Thermal Characteristics

The MPFD platform has been deployed primarily into research reactors that do not experience high temperatures more than 150°C, thus all materials are compatible for use due to their high melting point (> 1250°C). The MPFD internal structure has been manufactured using alumina to form the ionization chamber and to provide insulation between the anode, cathode, and exterior sheath. For this reason, the coefficient of thermal expansion (CTE) of the metals used should be well matched to the alumina ( $8.2 \times 10^{-6}$  K<sup>-1</sup>) to avoid stress development as the detector undergoes heating.

### Mechanical Characteristics

When MPFDs are deployed into a reactor core, they often come in direct contact with the reactor structural components which are commonly made of aluminum and stainless steel. The sheath material hardness, therefore, can impact the damage it causes to the reactor during deployment as materials harder than the reactor components have the potential to scratch or gouge during insertion and removal. The tensile strength of the material determines the maximum pressure the sheath can endure either from internal or external force.

Although not included in Table 3-4, the chemical resistance of each metal is important. As part of the MPFD research, no deployments have occurred in exotic reactors such as molten salt reactors or high temperature gas-cooled reactor, where chemical resistance of MPFD components is of high concern. Of the materials shown, both titanium alloys (Ti-6Al-4V and Ti-CP2), as well as SS316 and Inconel-600, have high chemical resistance, and therefore should be considered for special reactor deployments.

### **Electrical Characteristics**

Both the wires and the sheath play integral roles in the detection process. The sheath provides an electrically grounded case to mitigate radiofrequency pickup by the anode and cathode. The wires are utilized as the anode and cathode to produce an electric field inside the chamber to collect charge carriers and propagate the induced current to the detector electronics located outside the reactor primary vessel. To generate the electric field in the chamber, voltage must be supplied through the anode-cathode pair.

### **Radiation Interactions**

Each of these proposed materials were examined for neutron activation and gamma-ray interaction during irradiation. Neutrons and gamma rays will cause damage to the materials from lattice displacements and neutron activation. They also create secondary charged particles that can cause background signals in the detector. The neutron cross section of the material is a measure of the magnitude that the localized neutron flux affects the efficacy of the MPFD and can impose a negative reactivity burden on the reactor core. Any neutron that is absorbed by these materials will remove neutrons from the reactor that can be observed by the MPFD or utilized by the reactor to sustain the chain reaction. Additionally, any neutron absorbed by the MPFD material can cause neutron activation and transmutation to radioactive isotopes which can increase post-irradiation handling complexity. Optimal materials for use as the sheath and wires would have low neutron

and gamma-ray cross sections, and with neutron transmutation, would have short half-life daughter products. Gamma-ray cross sections for the most common elements in each alloy composition are shown in Fig. 3-16.

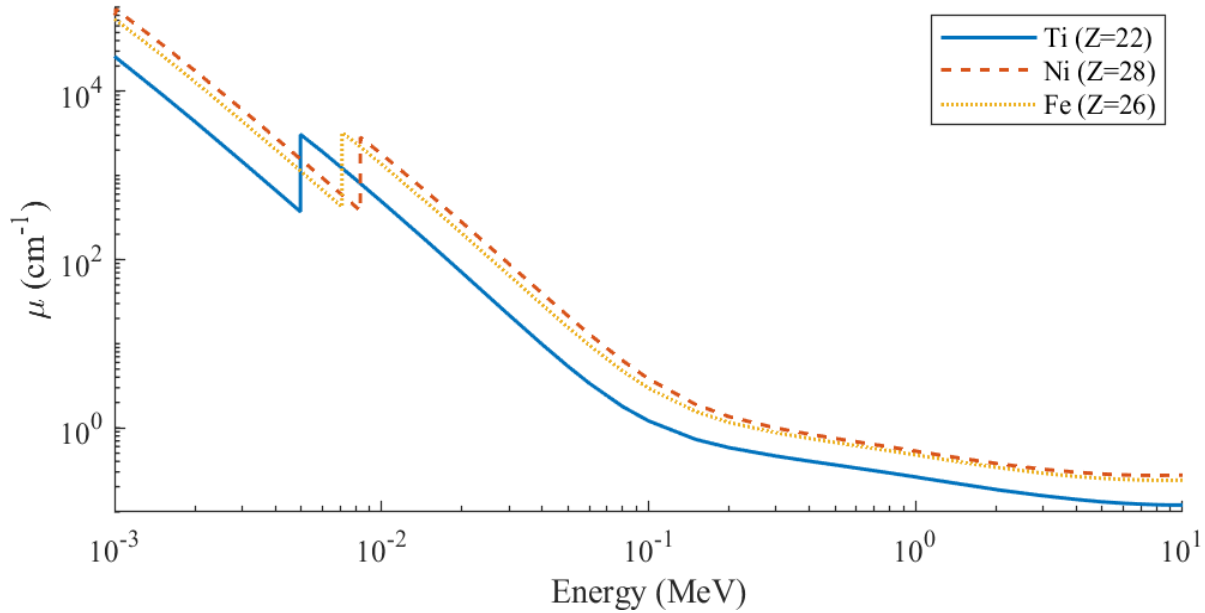


Fig. 3-16. Gamma-ray macroscopic cross sections for titanium, nickel and iron [31].

### 3.2.2 Fill-Gas Selection (Ar, Ar-N<sub>2</sub>, P-10, Neon)

Due to the nature of the in-core environment, only certain gases should be considered for use based on operability and longevity concerns. To determine which gas is suitable for usage in an MPFD, a list of gases was considered including Ar, Ar-N<sub>2</sub>, P-10, and Ne. Each gas was investigated for its ionization characteristics, charge carrier mobility, neutron activation, optimal pressure, operating voltage, and lifetime.

#### Ionization Potential, Stopping Power and Drift Velocity

After charge carriers are produced in the chamber gas, they will begin to drift towards the anode and cathode under the influence of the electric field. The number of charge carriers produced per ionization is determined by the gas average ionization energy,  $\omega$ . The transit time of these charge carriers is affected by the electric field strength, the gas pressure, and the distance to the corresponding electrodes (anode or cathode). Various properties of the gases under consideration are summarized in Table 3-5.

Table 3-5. Properties of the detector gases under consideration.

| Gas             | $Z_{\text{eff}}$ | $A_{\text{eff}}$ | $\rho_0$ (1 atm)<br>$10^{-3} \text{ g cm}^{-3}$ | $I_0$<br>eV | $\omega$<br>eV/(e-ion pair) | $(dE/dx)/\rho_0$<br>MeV $\text{cm}^2 \text{g}^{-1}$ | $dE/dx$<br>keV/cm |
|-----------------|------------------|------------------|-------------------------------------------------|-------------|-----------------------------|-----------------------------------------------------|-------------------|
| Ar              | 18               | 39.9             | 1.66                                            | 15.8        | 26                          | 1.47                                                | 2.44              |
| CH <sub>4</sub> | 10               | 16               | 0.67                                            | 13.1        | 28                          | 2.21                                                | 1.48              |
| N <sub>2</sub>  | 14               | 28               | 1.17                                            | 15.5        | 35                          | 1.68                                                | 1.96              |
| Ne              | 10               | 20.2             | 0.84                                            | 21.6        | 36                          | 1.68                                                | 1.41              |

Source: Table from McGregor and Shultis (2020) pg. 308 [12].

These gases will exhibit drift velocities as shown in Fig. 3-17, according to the electric field strength and the fill gas pressure inside the MPFD. While ion drift velocities are not included, they commonly travel at a factor of  $10^{-3}$  slower than their electron counterparts for a given gas. In the case of P-10 [Ar:CH<sub>4</sub> 90%:10%] and Ar-N<sub>2</sub> [Ar:N<sub>2</sub> 98.2%:1.8%], the lower mass molecules (either CH<sub>4</sub> or N<sub>2</sub>) act as a higher mobility gas to increase the average drift velocity of the mixture in comparison to pure argon.

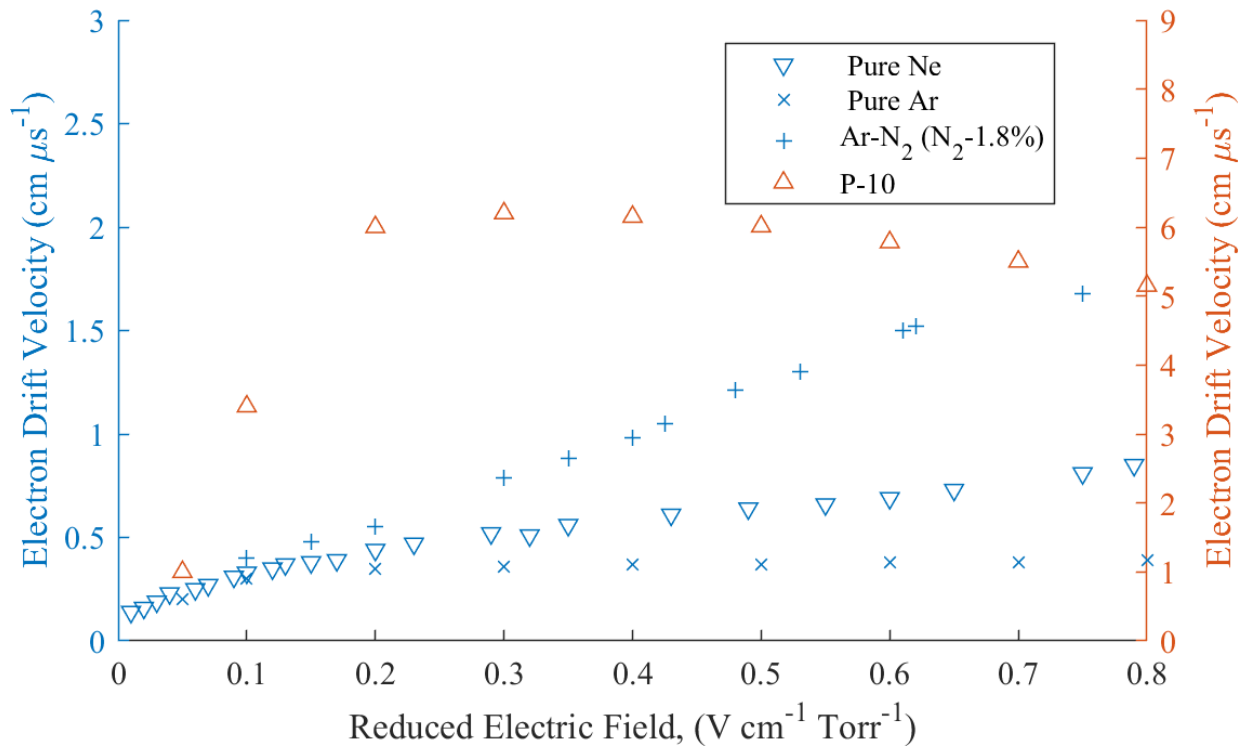


Fig. 3-17. Electron drift velocities for various gases for comparison. Data from: [80]–[82].

Argon, being the majority constituent of the gas mixtures of interest, will absorb most of the energy deposited by the fission fragments as they enter and coulombically interact with the gas. Due to

the lower initial ionization energy of the methane and nitrogen molecules, the argon ion will transfer its charge to these smaller molecules. These newly formed low-mass positive ions become the main charge carriers with higher velocities, rapidly drifting to the cathode.

### Pressure and Operating Voltage Impact

Previous MPFD designs had electrode spacings varying between 1.0 – 3.0 mm, with gas pressures ranging between 5 – 60 psig, and voltages ranging between 0 – 400 V. Because of the non-planar chamber geometry (discussed in Section 3.3), the electric field strength inside the ionization volume is not uniform. For example, electric field strength along the center plane bisecting the chamber shown in Fig. 3-18 are plotted in Fig. 3-19.

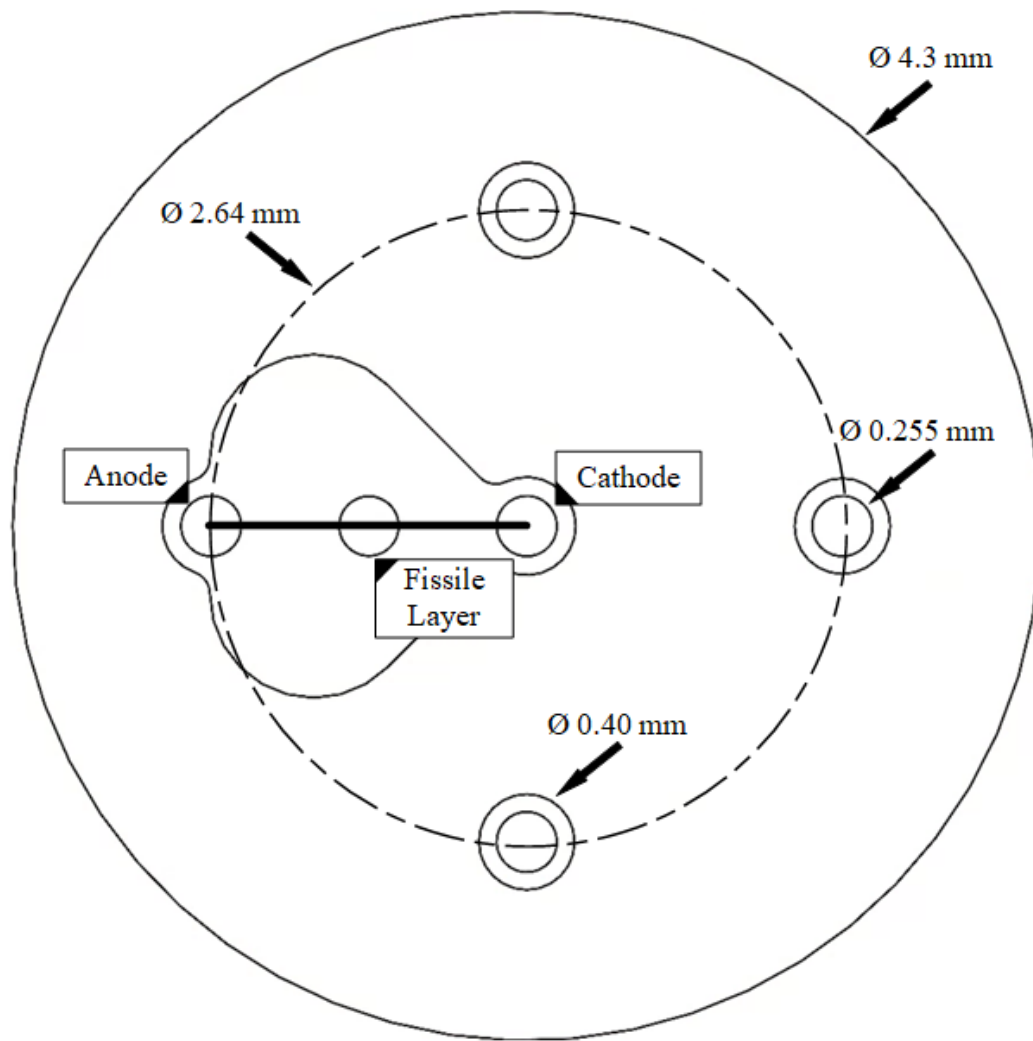


Fig. 3-18. Chamber bisecting line (solid black line) depicting the segment from which data shown in Fig. 3-19 was collected. The segment was positioned at half the height of the chamber.

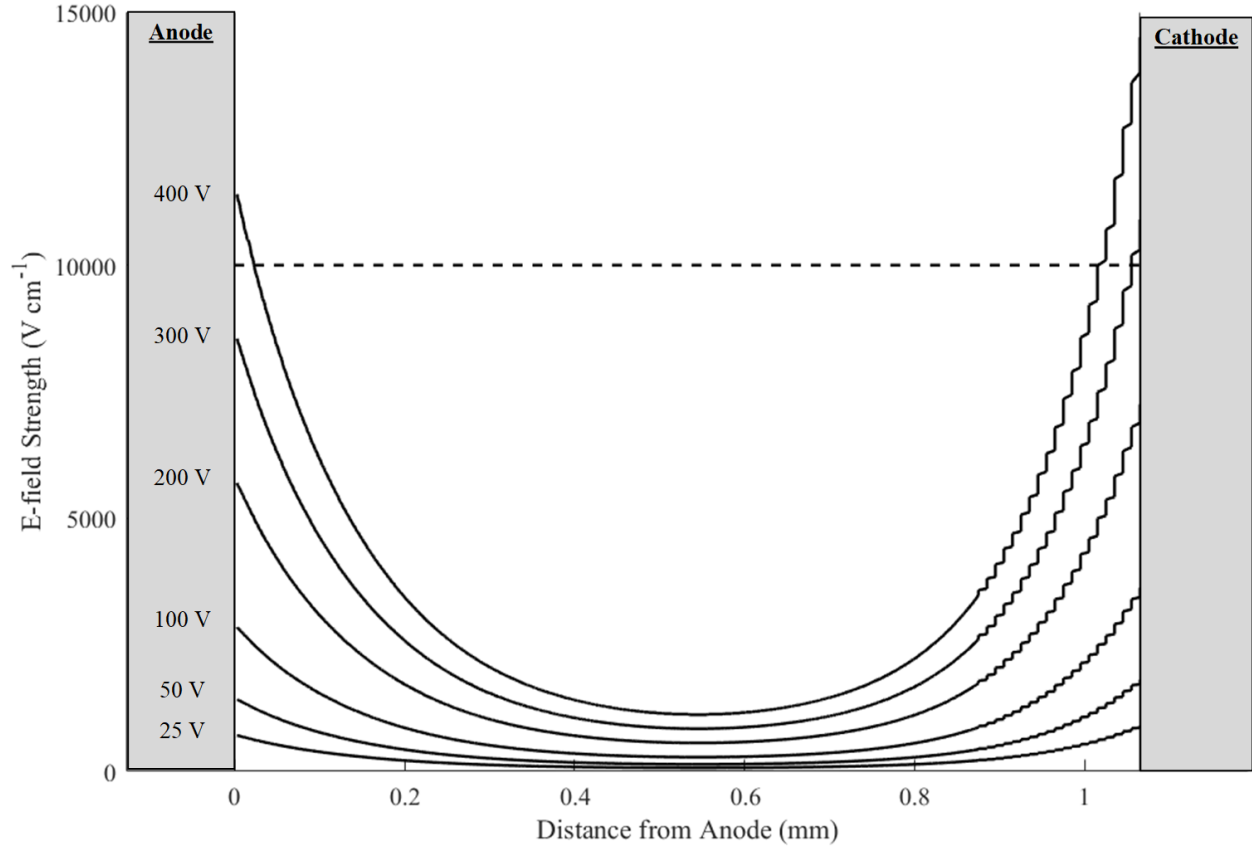


Fig. 3-19. Various operating voltages and the subsequent electric field strength between the anode and cathode. Dashed line indicates  $10 \text{ kV cm}^{-1}$  at which gas multiplication begins to occur for a fill gas pressure of 1 atm (760 Torr).

Based on the results shown in Fig. 3-19, for the clover chamber design, operating voltage should be limited to below 300 V to reduce probability of gas multiplication near the cathode wire if operated at 1 atm pressure. Utilizing these results, coupled with the electron drift velocity results shown in Fig. 3-17, it is determined that a minimum voltage of 75V is required to achieve saturation velocity across the chamber center plane for pure argon gas. It is important to note that the electric-field strength along the chamber center plane is the shortest distance between the anode and cathode, and thus represents the line with the highest average electric field strength magnitude. Due to the geometry of the clover chamber design, electric-field strength magnitude varies significantly further from the chamber center plane as shown in Fig. 3-20.

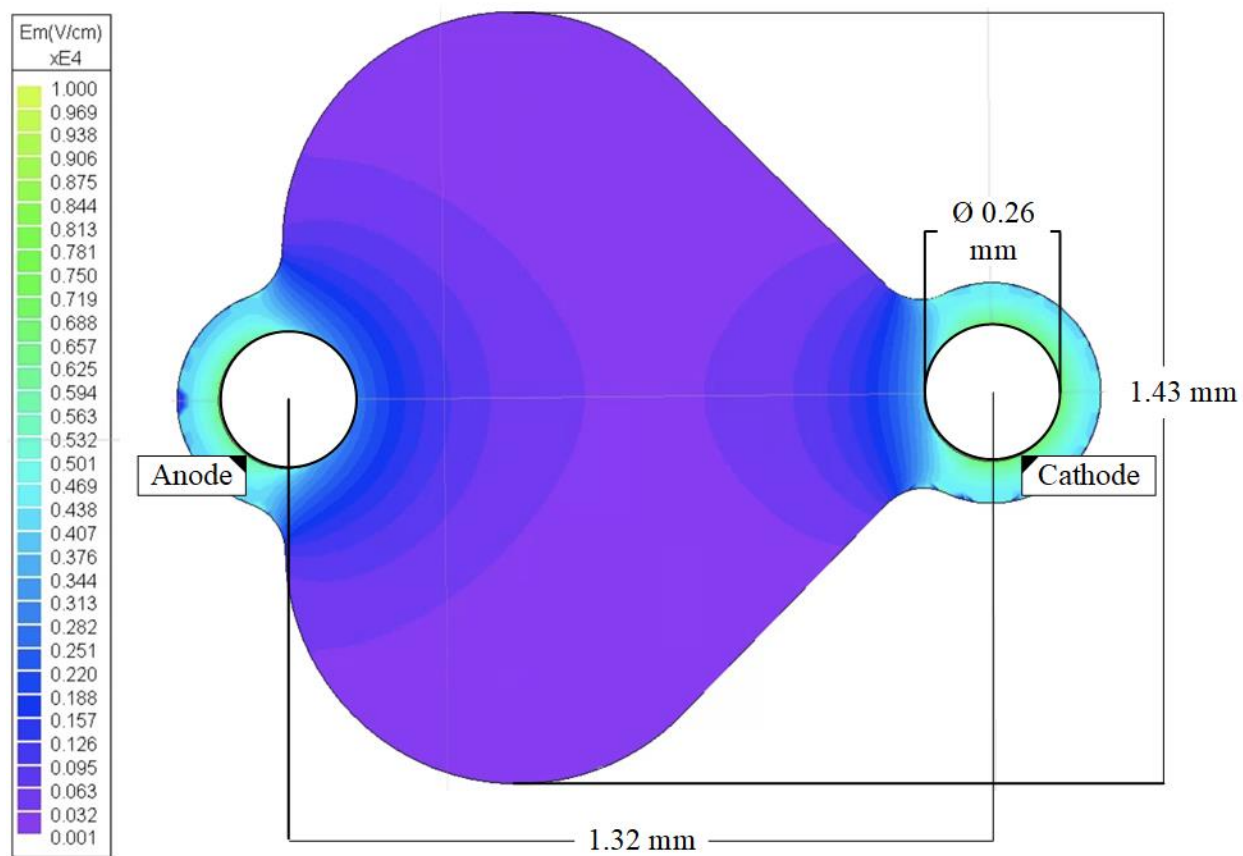


Fig. 3-20. The electric field magnitude inside the clover chamber with 100 V bias on all four anodes, with the central cathode and sheath set to 0 V.

### Charge Carrier Drift and Diffusion

As part of the electric field simulations, a preliminary study of charge carrier drift compared to diffusion was performed. Values for charge carrier drift and mobility were determined using equations and tabulated values from literature and are shown in Table 3-6 [12].

Table 3-6. Literature and calculated values for electron and Ar ion mobility and diffusion.

|                                                          | Electrons | Ar ions |
|----------------------------------------------------------|-----------|---------|
| $n_0$ (rel. conc.)                                       | 1         | 1       |
| $\epsilon$ (V cm <sup>-1</sup> )                         | 480       | 480     |
| $D$ (cm <sup>2</sup> s <sup>-1</sup> )                   | 113.4     | 0.008   |
| $\mu$ (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> ) | 153.38    | 0.34    |

Using Eq. (9), an estimated spatial charge carrier density was determined as a function of time and is shown in Fig. 3-21 the average thermal speed of free electrons after the initial ionization event is:

$$n_z(x, t, \varepsilon) = \frac{n_0}{\sqrt{4\pi D_z t}} \exp\left(\frac{-(x - \mu_z \varepsilon t)^2}{4 D_z t}\right) \quad (9)$$

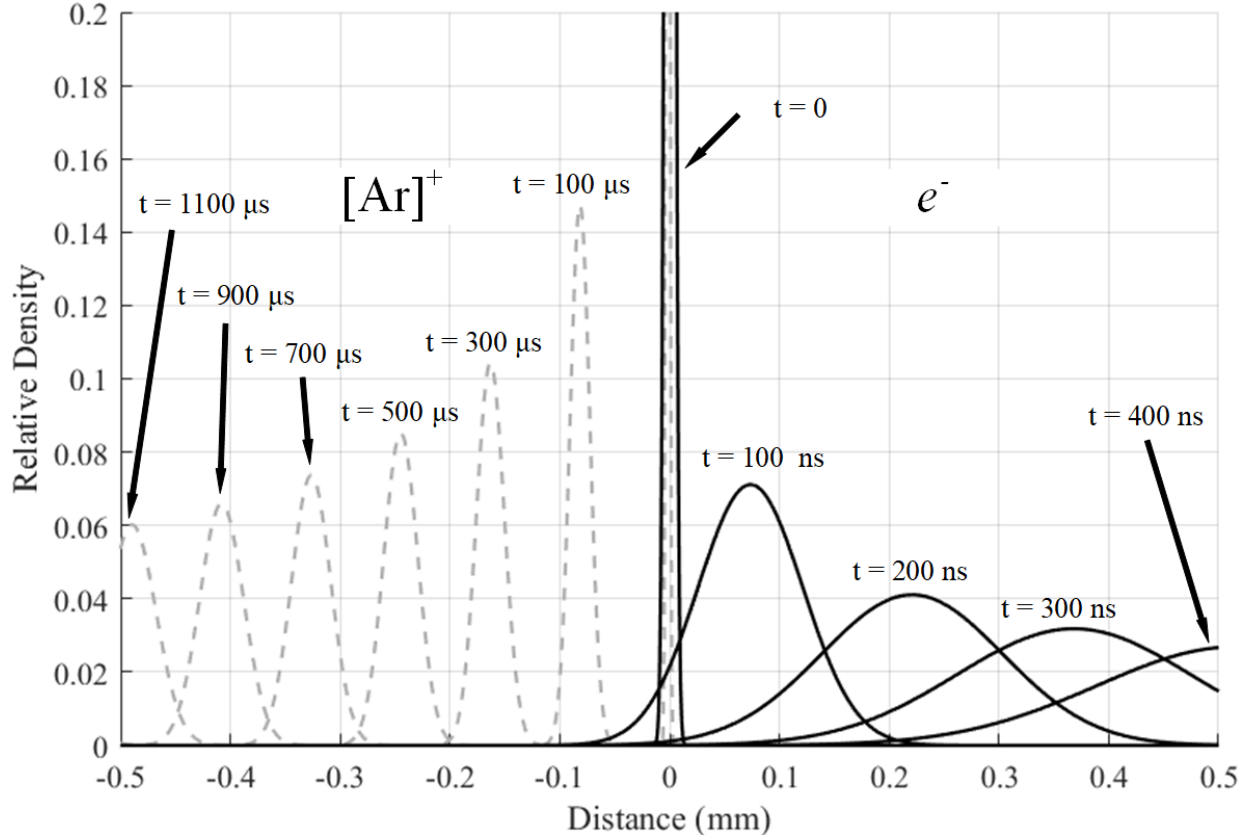


Fig. 3-21. Plot showing electron and ion spatial drift and diffusion of the charge carriers within the detection gas with a 100 V bias. The left and right sides of the plot represent the cathode and anode respectively.

This result shows that electrons experience more lateral diffusion in comparison to the Ar ions as they mobilize towards the respective electrodes. This lateral diffusion will cause some of the initial electron cloud to reach the chamber wall prior to reaching the anode, thus resulting in a reduced charge collection efficiency ultimately reducing the total charge measured.

## Radiation Effects

Each of these examined gases will absorb neutrons and can transmute into isotopes which will no longer aid the detection process. A special consideration is given to the methane included in P-10 gas mixtures. Methane serves as a quenching gas included with argon to form a mixture (Ar/CH<sub>4</sub> 90:10%) and aides in increasing charge carrier mobility (refer to Fig. 3-17). The bond energy between the attached hydrogen atoms and the central carbon atom (see Fig. 3-22) have bond strengths of 18.7, 20.5, 19.6, 14.6 eV for the first, second, third and fourth bond in the sequence [83].

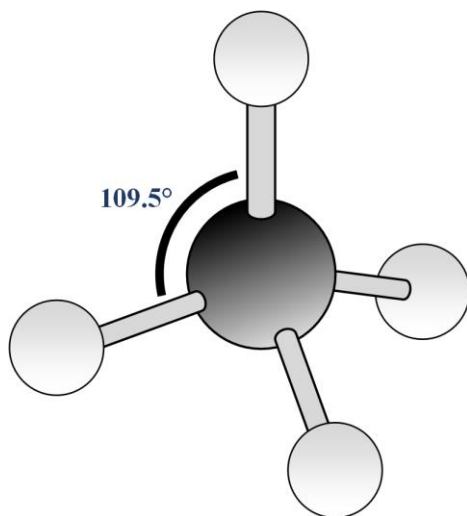


*Fig. 3-22. The tetrahedral methane molecule showing the bond angle.*

Because of the low energy required to separate a bonded hydrogen from the central carbon atom, methane readily dissociates and can form long polymeric chains with other dissociated hydrocarbons. This process leads to the precipitation of hydrocarbon chains and the subsequent polymers coating the walls of the ionization chamber including the electrodes, causing a build-up of electrically-insulating material [84]. While methane can significantly increase charge carrier mobility (see Fig. 3-17), it should not be used in high neutron and gamma-ray fluence environments due to its degradation and potential of negatively impacting gas detector efficacy.

Neutron activation of the various gas molecules will lead to transmutation and can impact detector performance. The nitrogen gas will undergo  $^{14}\text{N}(\text{n},\text{p})^{14}\text{C}$  and  $^{15}\text{N}(\text{n},\gamma)^{16}\text{N}$  reactions, with the  $^{16}\text{N}$  quickly decaying ( $T_{1/2} = 7.13$  s) into  $^{16}\text{O}$  which can pollute the gas causing an increase in electron attachment phenomena. It is for this reason, that nitrogen gas should be used with caution,

as high concentrations of oxygen will contaminate the detector gas causing a reduction in efficiency. A thorough explanation of electron attachment can be found elsewhere [12]. The argon gas will undergo  $^{40}\text{Ar}(n,\gamma)^{41}\text{Ar}$  and decays ( $T_{1/2} = 109.61 \text{ m}$ ) into  $^{41}\text{K}$  and the neon gas will undergo  $^{22}\text{Ne}(n,\gamma)^{23}\text{Ne}$  and decays ( $T_{1/2} = 37.25 \text{ s}$ ) into  $^{23}\text{Na}$ . Both  $^{41}\text{K}$  and  $^{23}\text{Na}$  are non-radioactive alkali metal isotopes.

### 3.2.3 Substrate Selection

The substrate material used for the internal components of the MPFD must be carefully selected to withstand the intense radiation, heat, and pressure conditions of in-core deployment and usage. These requirements significantly limit the materials that can be considered for use as the fissile layer substrate. High temperature ceramics with high strength and low electric conductivity are considered. During the course of the MPFD research, only two materials, aluminum oxide ( $\text{Al}_2\text{O}_3$ ) and aluminum nitride ( $\text{AlN}$ ) were considered and examined, although other materials have been proposed and considered elsewhere, such as silicon dioxide ( $\text{SiO}_2$ ) and silicon carbide ( $\text{SiC}$ ) [41], [85].

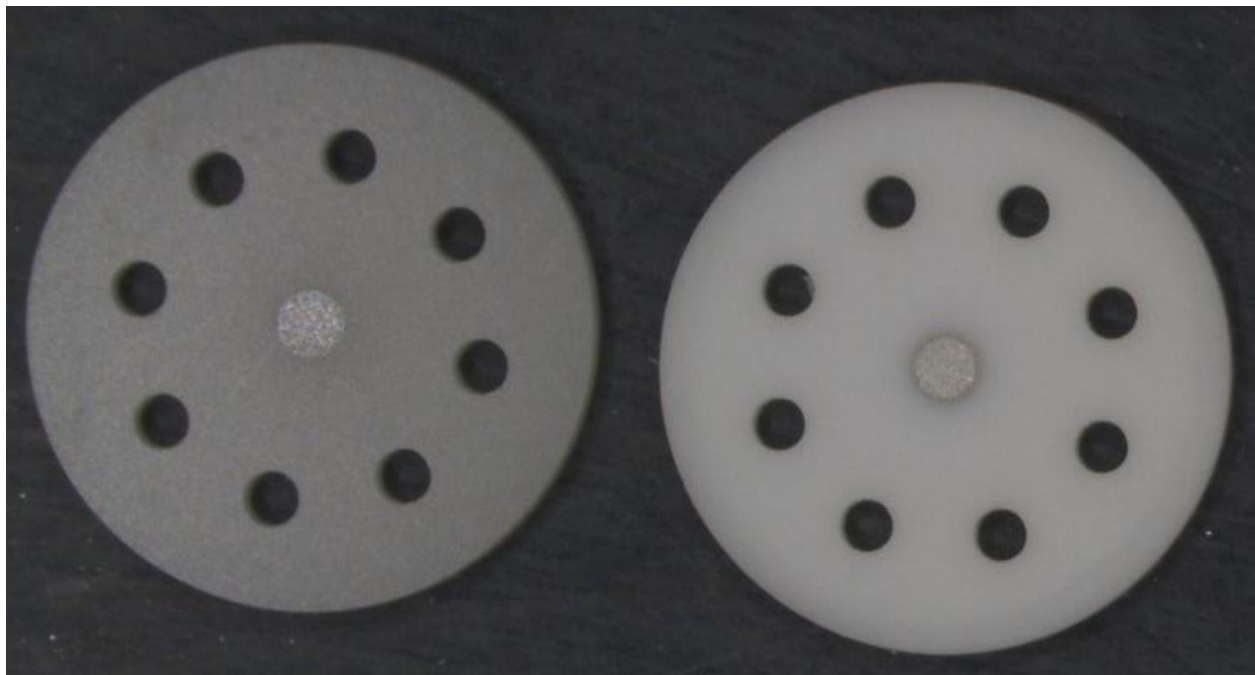


*Fig. 3-23. Microscope image of aluminum nitride (left) and alumina (right).*

## Summary of Characteristics

Three categories of material characteristics were considered which can affect device construction and performance, those being thermal, mechanical, and electrical. The substrate fulfills several key roles as part of the MPFD device, such as a thermal sink during fast transients, the base material to which the fissile layer is applied, and as an insulator between the voltage biased anode and cathode. The substrate also separates the electrode wires from the outer sheath and provides structural support. A summary of various material properties of interest for the two considered materials is given in Table 3-7. Additional detail about each relevant characteristic is provided in the following sections.

*Table 3-7. Material properties for alumina oxide and aluminum nitride.*

|        |                                           | <b>Al<sub>2</sub>O<sub>3</sub></b> | <b>AlN</b>       |                                    |
|--------|-------------------------------------------|------------------------------------|------------------|------------------------------------|
| Therm. | <i>Specific Heat</i>                      | 880                                | 740              | J kg <sup>-1</sup> K <sup>-1</sup> |
|        | <i>Conductivity</i>                       | 31                                 | 175              | W m <sup>-1</sup> K <sup>-1</sup>  |
|        | <i>CTE</i>                                | 8.2                                | 4.6              | 10 <sup>-6</sup> K <sup>-1</sup>   |
| Mech.  | <i>Density</i>                            | 3.92                               | 3.26             | g cm <sup>-3</sup>                 |
|        | <i>Hardness-Vickers</i>                   | 14.1                               | 11.8             | GPa                                |
|        | <i>Surface Roughness (R<sub>a</sub>)*</i> | 6.33                               | 24.35            | nm                                 |
| Elec.  | <i>Dielectric Constant @ 1 MHz</i>        | 9.8                                | 8.9              | -                                  |
|        | <i>Resistivity @ 300 K</i>                | 10 <sup>14</sup>                   | 10 <sup>13</sup> | ohm-cm                             |

\* Valley Design Corp. materials measured using a Veeco Dektak 150 profilometer

## Heat Capacity and Thermal Conductivity

As the fissile layer undergoes fission inside a nuclear reactor, a significant amount of heat can be deposited inside the chamber. This heat load is shared amongst the various internal components, chief amongst these being the fissile layer, titanium and platinum adhesion layer, and the substrate. Although some heat is transferred to the gas, a significant fraction is deposited in the base substrate. During the MPFD research, concern developed that fast transients could cause a significant amount of localized heating inside the MPFD chamber due to the high fission rate. Given the short duration of reactor pulsing (10 – 100 ms FWHM), enough heat could be generated and deposited in the fissile layer to cause degradation or delamination of the Ti/Pt adhesion layer from the substrate. For example, during fast power transients at TREAT, a natural enrichment uranium fissile layer (0.5 µg) could experience a 20 GW<sub>th</sub> pulse (100 ms duration). This would

result in 60  $\mu\text{J}$  of energy produced inside the MPFD fissile layer and surrounding material. Given that the specific heat of uranium dioxide is  $294 \text{ J kg}^{-1} \text{ K}^{-1}$  and the melting point is 3138 K, the fissile layer temperature would rise 408 K. This heat must be dissipated through the Ti/Pt layer and into the substrate. It is for this reason that the specific heat and thermal conductivity of the substrate is important when selecting a material.

### **Surface Roughness Effect on Electrode Adhesion**

As part of the manufacturing process, (see Section 4.1), a Ti/Pt adhesion layer is evaporated onto the substrate to provide a conductive surface to which the fissile layer is applied during the electrodeposition process (discussed further in Section 4.2). Surface roughness of the base substrate will impact the roughness of the Ti/Pt layer and can affect the layer uniformity of the electrodeposited fissile layer. It is therefore important that the substrate selected for manufacturing has a fine surface roughness with a lapped or polished finished. Additionally, any defects in the substrate such as pitting or scratches, should be minimized as it will cause defects in the fissile layer as shown in Fig. 3-24.

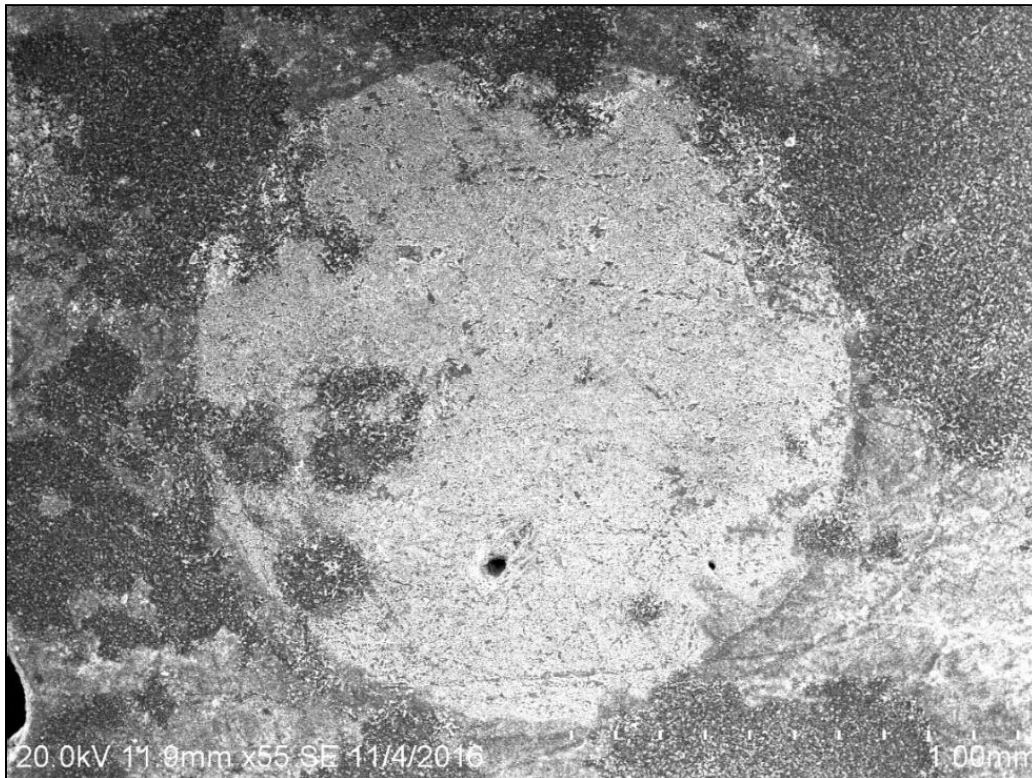


*Fig. 3-24. SEM image capture of the fissile layer pitting caused by a substrate defect.*

Legacy MPFD substrates were manufactured by cutting annular capillary tubing stock to the desired thickness with a diamond wire saw and then sent to a third-party vendor for micromachining and polishing. To increase the reliability of substrate material, current MPFD substrates are procured from third-party vendors which supply high purity, high polish alumina and aluminum nitride wafers with surface roughness listed in Table 3-7. Vendors offer a wide variety of thicknesses (0.125 – 1 mm) and can provide laser water jet cutting to produce custom shapes.

### **Resistivity and Dielectric Constant**

The chamber substrate is used to form the gas volume and electrically isolate the anode, cathode, and sheath. Because of this requirement, it is important that the substrate is a strong insulator with high resistivity and a high dielectric constant (also known as relative permittivity) to reduce leakage current. Both the resistivity and dielectric constant will decrease as temperature of the material increases which should also be considered for high temperature applications of the MPFD.

## **3.3 Geometric Considerations**

Previous designs of the MPFD utilized a 4-node configuration where each node comprises a unique fissile layer, chamber, and anode-cathode pair. Originally this design had a trench machined in the alumina substrate to allow for charge carriers to drift toward their respective anodes, but a later iteration of the design utilizes a diamond shaped chamber to form the gas volume [6], [86]. As part of the design optimization process, various chamber shapes and electrode configurations were simulated and ultimately experimentally tested for performance. As a preliminary step to determining chamber shape impacts on detector performance, electrostatic simulations were performed.

### **3.3.1 Electric-field Simulations**

Over the course of the MPFD research presented herein, three different MPFD designs were tested experimentally; colloquially known as the trench, diamond, and clover design. Early in the research there was a transition from the trench to diamond design which was a result of better manufacturing methods and an effort to decrease production costs. The trench design was not included as part of the electric-field simulations as it was no longer utilized after the adoption

of the diamond design. An illustration of each design with top-down and cross-sectional views is presented in Fig. 3-25. More detailed illustrations of the diamond and clover design are provided in Fig. 3-26 and Fig. 3-27. Three characteristics of the chamber shape were examined as part of these simulations: the electric field line orientation, the electric field strength, and the weighting potential. As a result of the study, it was observed that the diamond design had certain drawbacks when utilized in a multi-node MPFD array. Additionally, the clover design was determined to be a more suitable candidate design for multi-node arrays due to its electric field streamline alignment for each detection chamber.

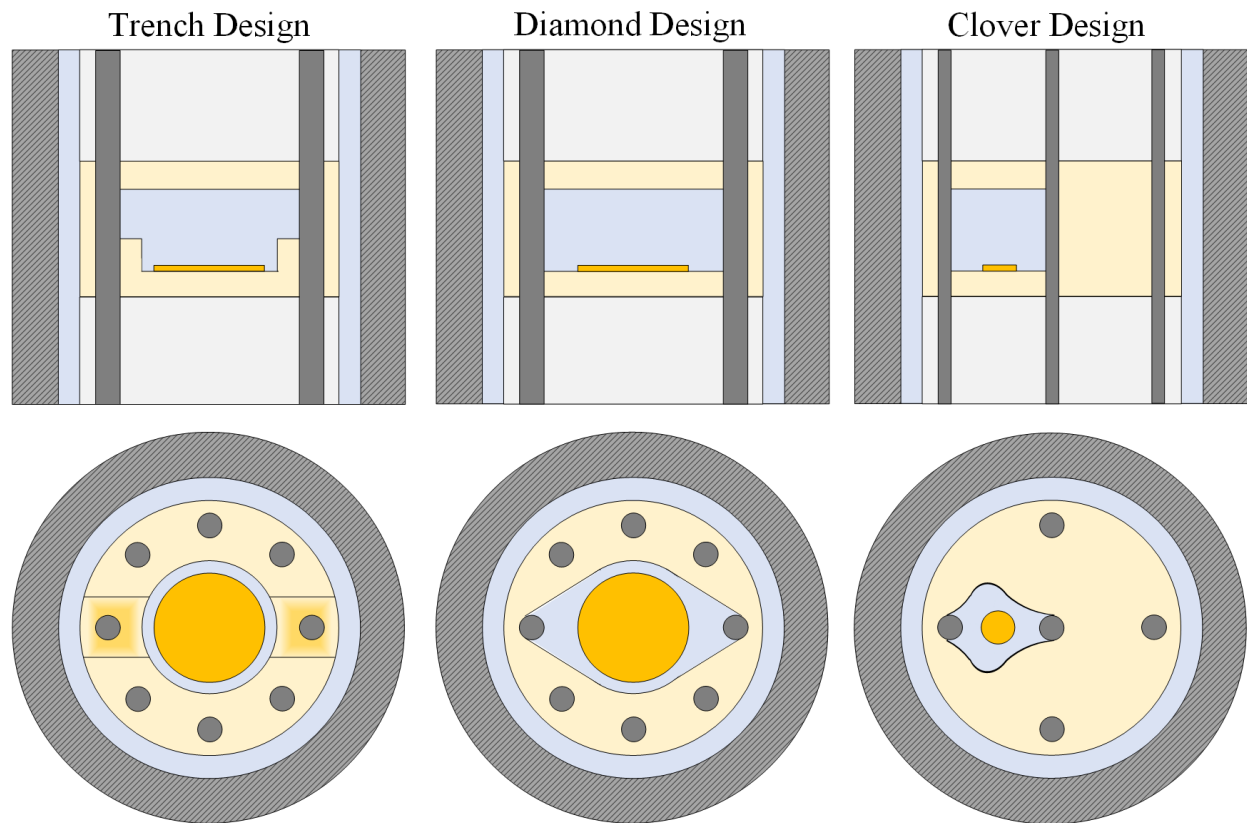


*Fig. 3-25. An illustration of each of the design changes during the MPFD development, culminating in the final clover design.*

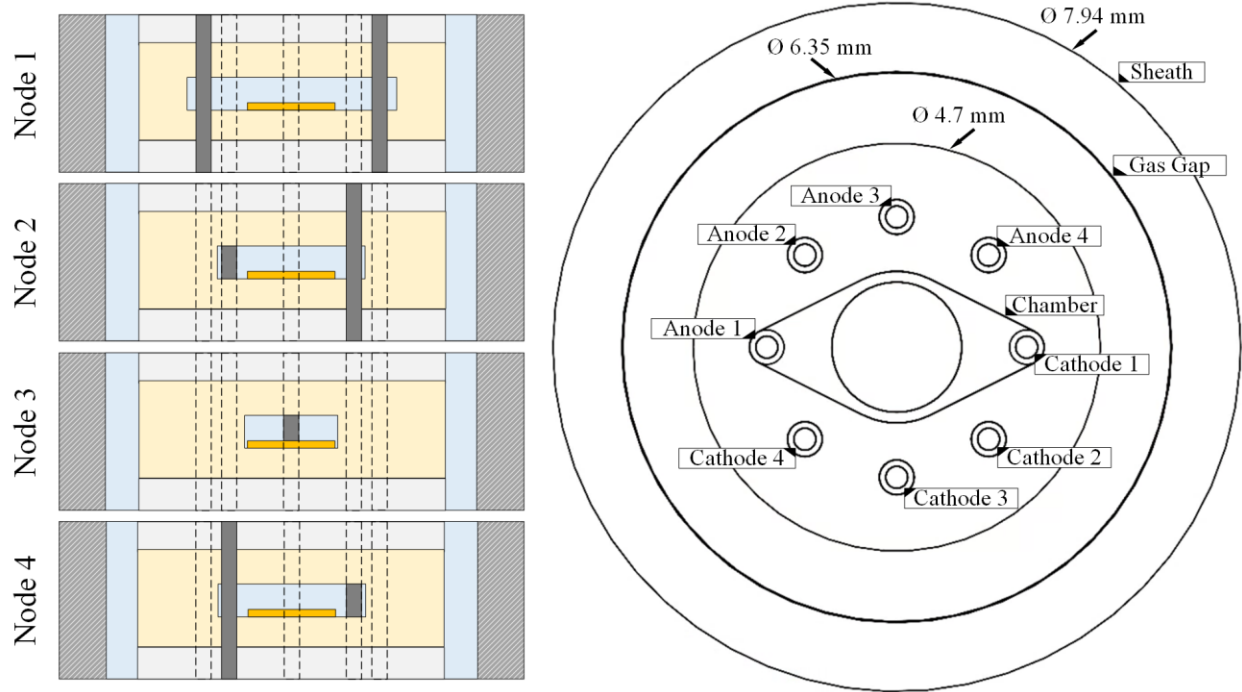


Fig. 3-26. Side cross section slices of each node chamber (left) of the diamond chamber design, and a top-down view of node 1 (right) with dimensions and labeled features.

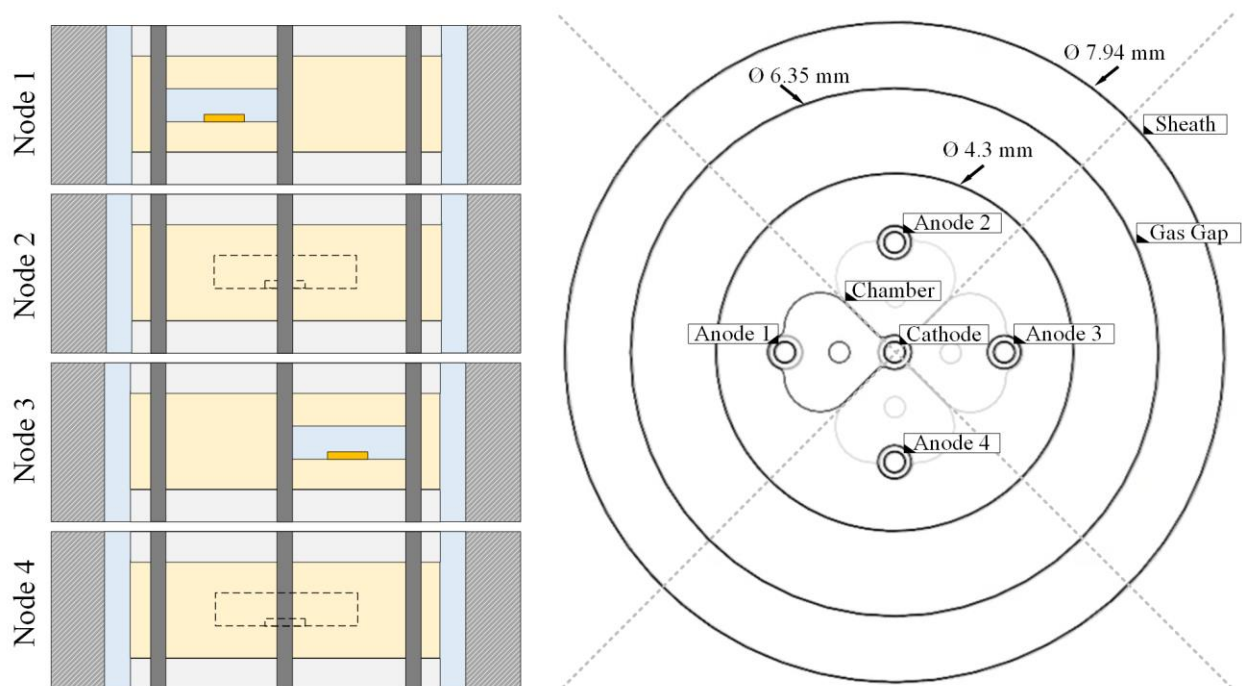


Fig. 3-27. Side cross section slices of each node chamber (left) of the clover chamber design, and a top-down view of node 1 (right) with dimensions and labeled features. Dashed lines in the top-down view show how each node is a copy of the previous node rotated 90° clockwise.

## Electric-field Orientation

Traditional ion-chamber designs utilize parallel-plate configurations which result in a uniform electric-field strength and mono-directional electric-field lines, as shown in Fig. 3-28. Electric-field lines represent the theoretical direction charge carriers will follow when influenced by an electric field. Parallel-plate designs result in charge carriers being oriented directly towards the opposing polarity electrodes (ions to the cathode, and electrons to the anode) with straight trajectories. Parallel-plate designs also have a constant weighting field gradient between the anode and cathode as shown in Fig. 3-29. In contrast, MPFDs are constructed with wires located at opposing ends of the chamber resulting in electric field line curvature as seen in Fig. 3-28. This is known as a parallel wire configuration which leads to a non-constant weighting potential across the chamber as shown in Fig. 3-29.

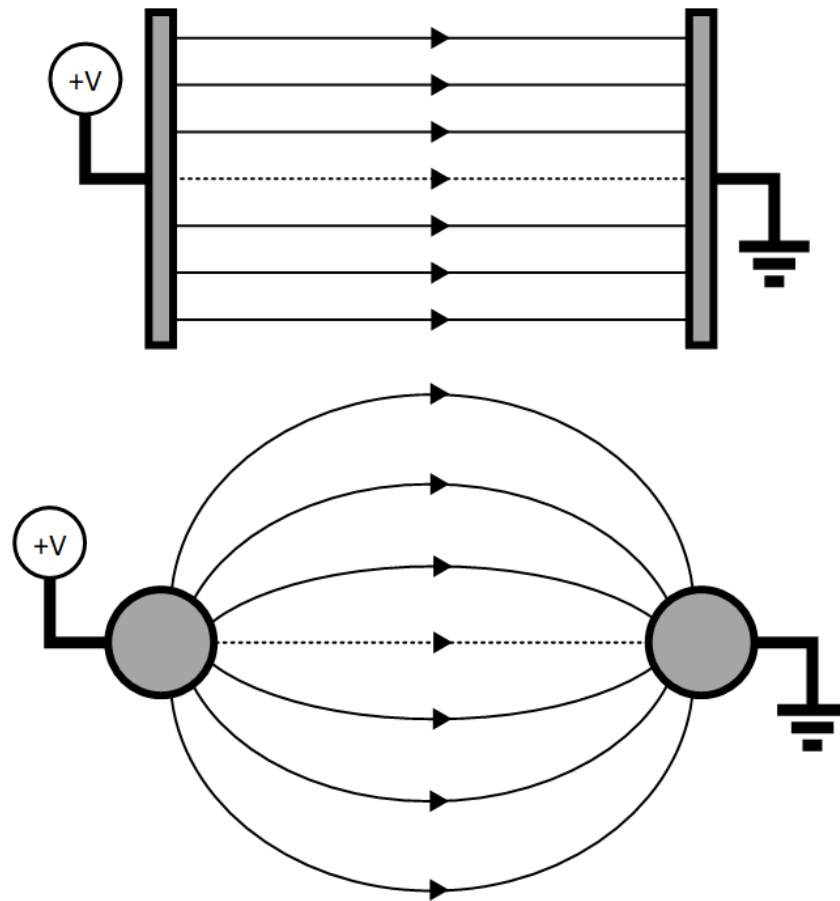


*Fig. 3-28. Comparison of electric-field streamlines for idealized parallel plate (top) and parallel wire (bottom) configurations. Note the parallel and straight lines in the parallel plate configuration, whereas in the parallel wire configuration where streamlines curve toward the electrodes. Additionally, in the parallel wire configuration, streamline density is highest near each electrode.*

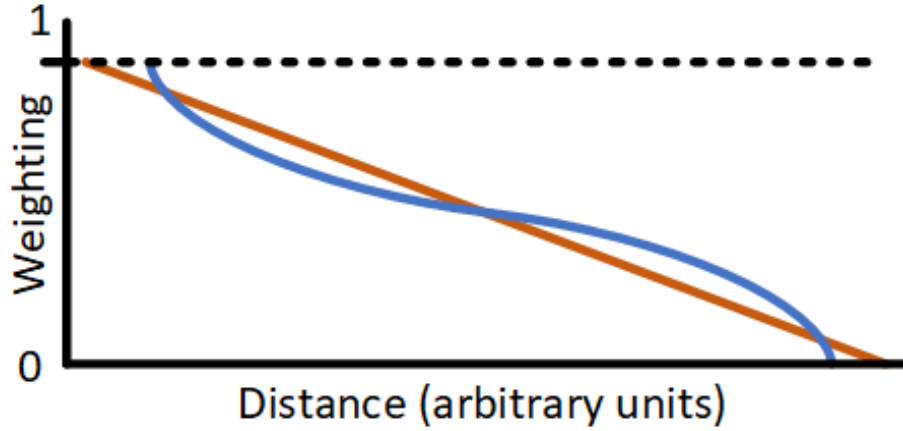


*Fig. 3-29. Theoretical weighting distributions for the parallel plate (orange) and parallel wire (blue) configurations along the chamber center line (dotted line in Fig. 3-28) between the electrodes.*

Both cases presented above represent ideal conditions for the parallel plate and parallel wires configurations. One of the drawbacks of the multi-node MPFD diamond design is that the anode-cathode pairs of the other chambers can impact electric-field orientation which can cause charge carriers to sweep into the chamber walls prematurely terminating their contribution to current induction. This scenario leads to lower voltage pulse amplitudes as measured in the coupled electronics and reduces the detection efficiency of the device. The electric-field lines for each chamber of a 4-node MPFD diamond design are shown in Fig. 3-30. As observed, due to the pattern of anode and cathode pairs, both node 1 (top of the array) and node 4 (bottom of the array) suffer from the misaligned electric-field lines. In contrast, the MPFD clover design was selected due to its rotational symmetry about the central cathode, whereby each of the 4 nodes had identical intra-chamber electric-lines as shown in Fig. 3-31. The electric field line alignment of the clover design sweeps charge carriers towards the respective electrodes with a lower probability of being swept into the chamber walls. Additionally, the rotational symmetry of the design leads to identical weighting field and electric field strength conditions in each of the chambers in a multimode design. For these reasons the clover design was determined to be the preferred chamber shape for multi-node MPFD array fabrication. The diamond chamber shape was determined to only be suitable for single-node MPFD assemblies.

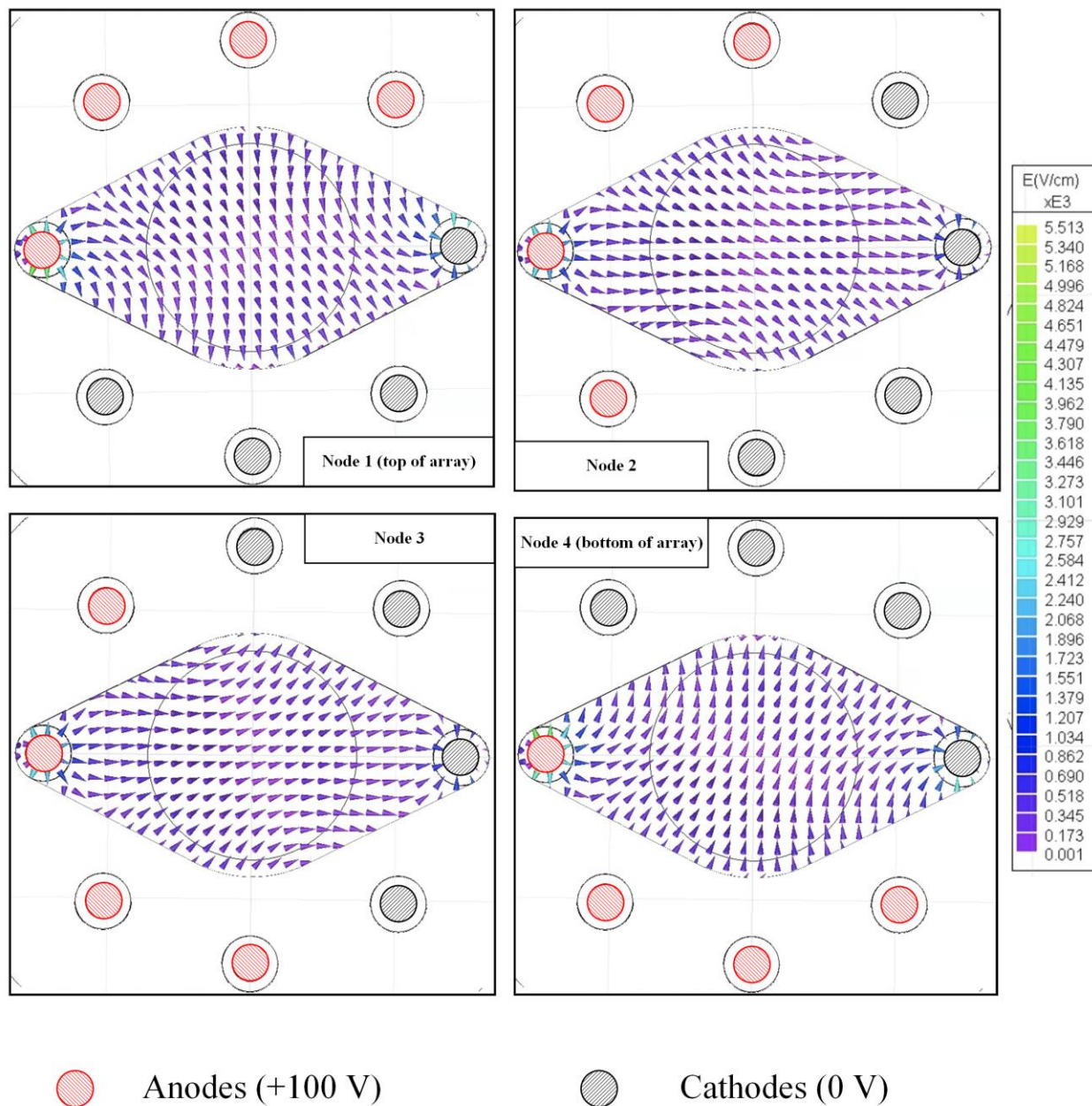


Fig. 3-30. Diagram of electric field lines inside the diamond shaped chamber for each of the four nodes. Note that for nodes 2 and 3 (top right and bottom left), the electric field is primarily oriented toward the anode cathode pair, whereas in nodes 1 and 4 (top left and bottom right), the electric field is primarily oriented towards the walls. The sheath is include in the simulation, but not shown, and was set to 0 V potential.

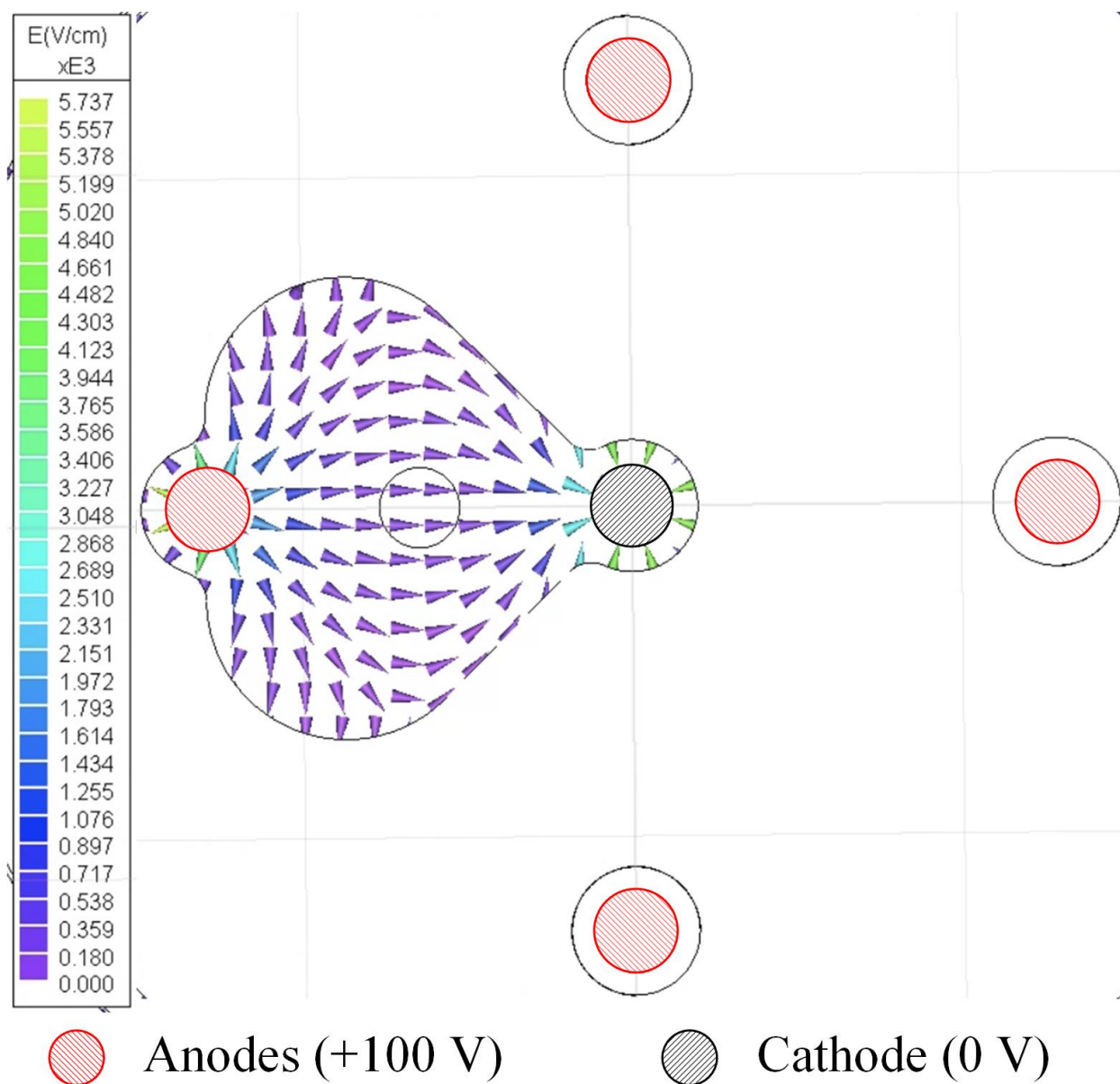


Fig. 3-31. Diagram of electric field lines inside the clover shaped chamber. Note how the chamber shape conforms to most of the electric field lines. Because of the symmetry about the center for each node, the electric field lines depicted in this figure is representative of all four nodes. The sheath is include in the simulation, but not shown, and was set to 0 V potential.

## Electric-field Strength

The electric-field strength throughout the chamber determines the electron and ion drift velocities. As shown in Fig. 3-17, electron drift velocities for various reduced electric field strengths are provided. For a gas pressure of 60 psig, the electron drift velocity at an electric field strength of  $0.032 \text{ V cm}^{-1}$  in the chamber is  $0.266 \text{ cm } \mu\text{s}^{-1}$  which is approximately 69% of the maximum drift velocity. Electron's will achieve a drift velocity of  $0.355 \text{ cm } \mu\text{s}^{-1}$  (approximately 92.1% of the maximum drift velocity), at an electric field strength of  $0.095 \text{ V cm}^{-1}$ . By comparing electric-field strengths for both the diamond-shaped chamber (shown in Fig. 3-32) and the clover-shaped chamber (shown in Fig. 3-33) it shows that the majority of the chamber of each design has electron drift velocities close to the maximum which reduces charge carrier collection time.

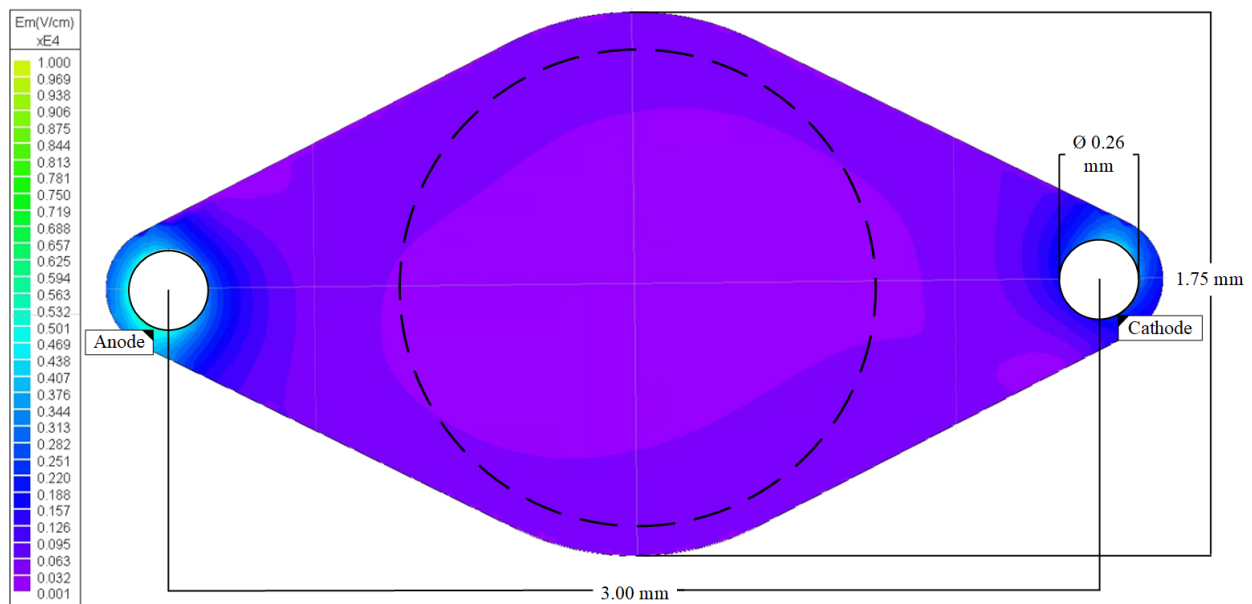


Fig. 3-32. The electric field magnitude inside the diamond chamber (node 1) with 100 V bias on all four anodes (west, northwest, north and northeast), with all cathodes (east, southeast, south, and southwest) and the sheath set to 0 V. The fissile layer (1.5 mm diameter) is indicated as the dotted line circle.

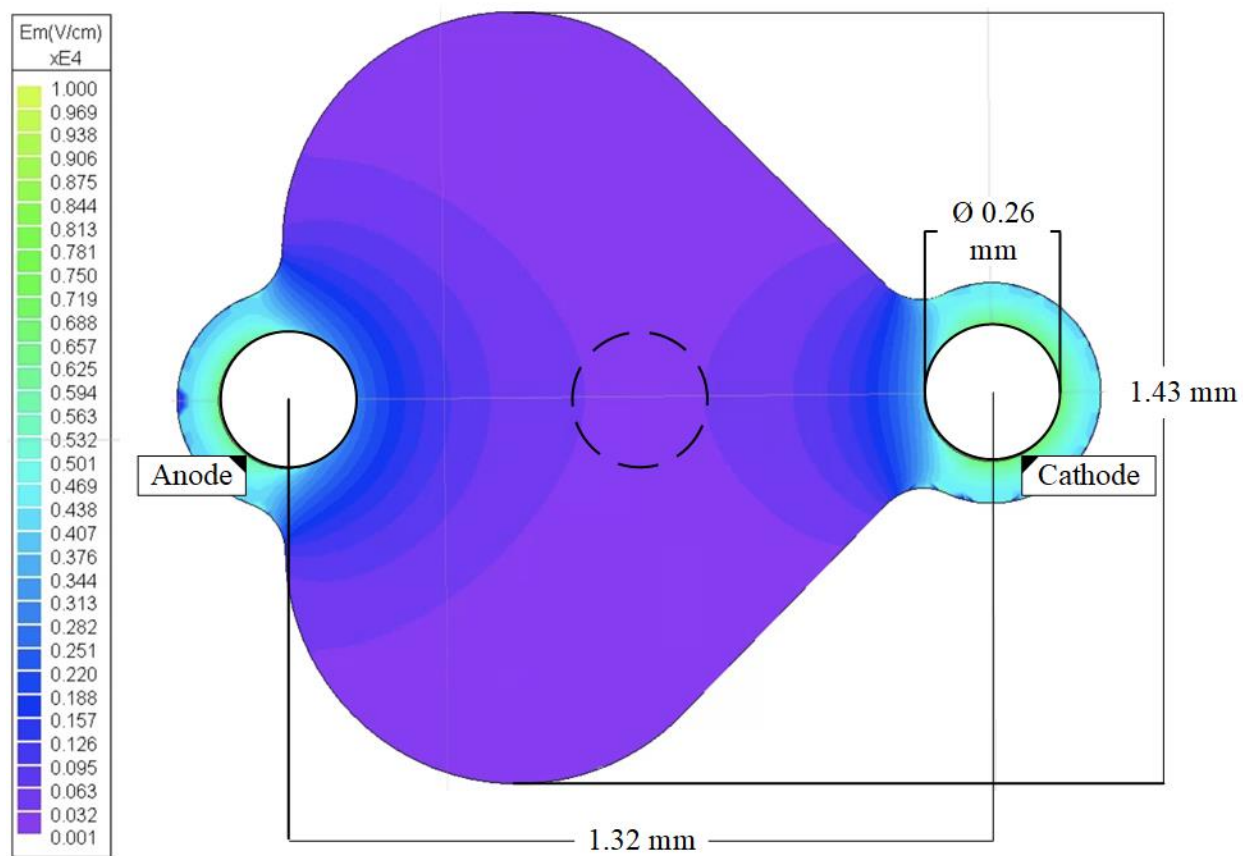


*Fig. 3-33. The electric field magnitude inside the clover chamber with 100 V bias on all four anodes (north, east, south, and west), with the central cathode and the sheath set to 0 V. The fissile layer (0.25 mm diameter) is indicated as the dotted line circle.*

### **Weighting Potential**

The weighting potential for both the diamond- and clover-shaped chambers were simulated and compared. In both chamber geometries the weighting field has the largest gradient nearest to the anode. In the clover-shaped chamber, the gradient of the weighting field is larger in comparison to the diamond-shaped chamber due to the weighting field compression because of the central cathode. This effect results in voltage pulse rise times during pulse-mode operation.

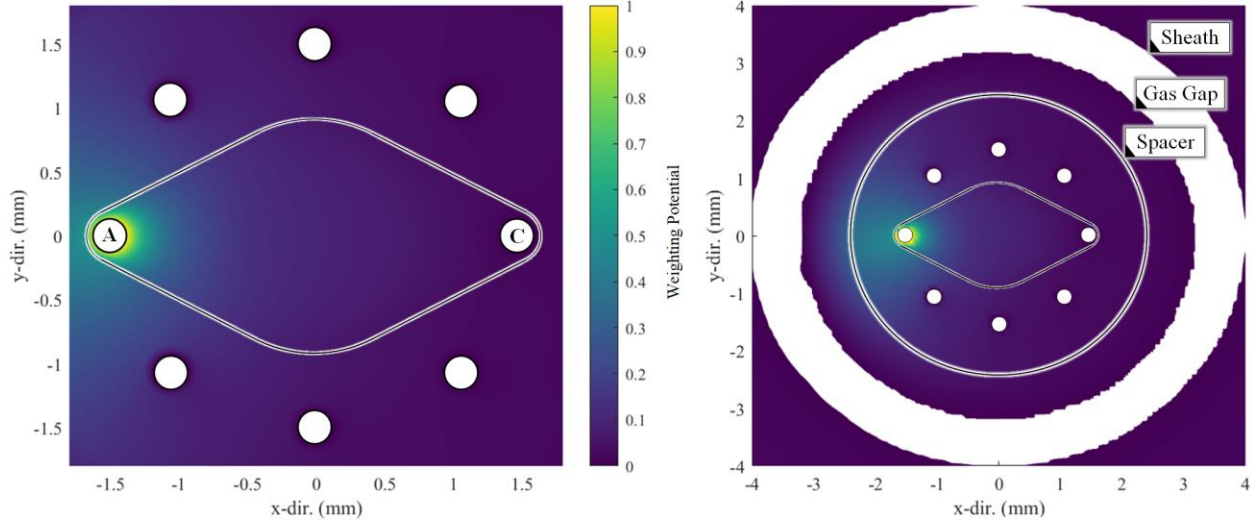


Fig. 3-34. Weighting potential field inside the diamond shaped chamber. A detailed view of the chamber (left) as well a full detector cross section (right) with labels for the spacer, gas gap, and sheath.

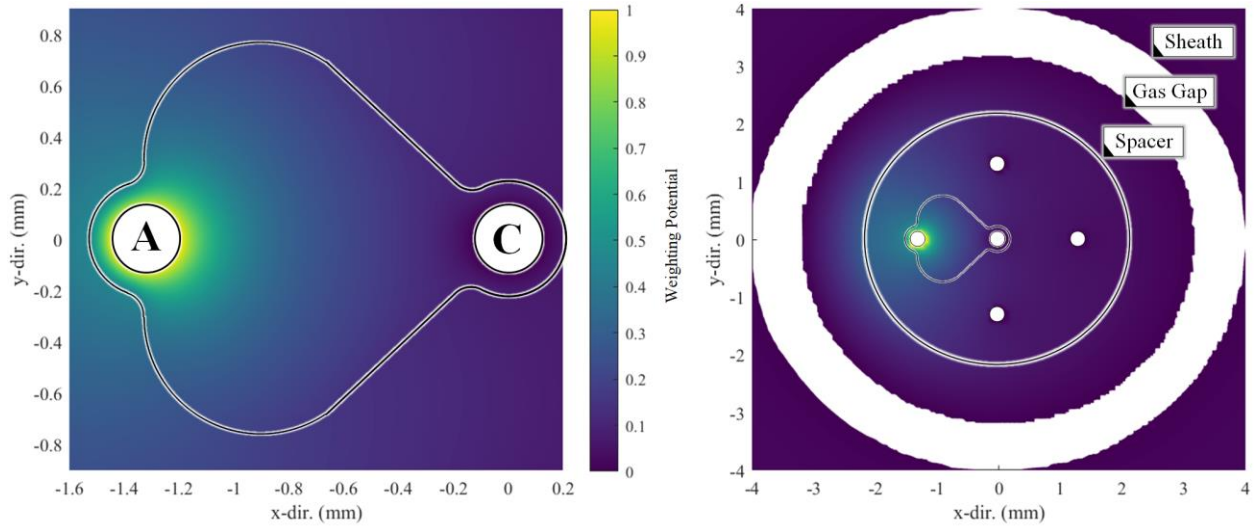


Fig. 3-35. Weighting potential field inside the clover shaped chamber. A detailed view of the chamber (left) as well a full detector cross section (right) with labels for the spacer, gas gap, and sheath.

### 3.3.2 Simulated Energy Deposition Spectra

In addition to electrostatic modeling, both the diamond and clover design was simulated with a fission fragment source in MCNP6.2 to estimate energy deposition spectra. Both geometries were modeled using the dimensions shown in Fig. 3-26 and Fig. 3-27 respectively and the fissile layer was modeled as uranyl nitrate,  $\text{UO}_2(\text{NO}_3)_2$ , at a density of  $2.81 \text{ g cm}^{-3}$  and the ionization gas was simulated at 60 psig. The clover design provides a lower average energy deposition of 15.99 MeV in comparison to the diamond design average energy deposition of 20.85 MeV as shown in Fig. 3-36.

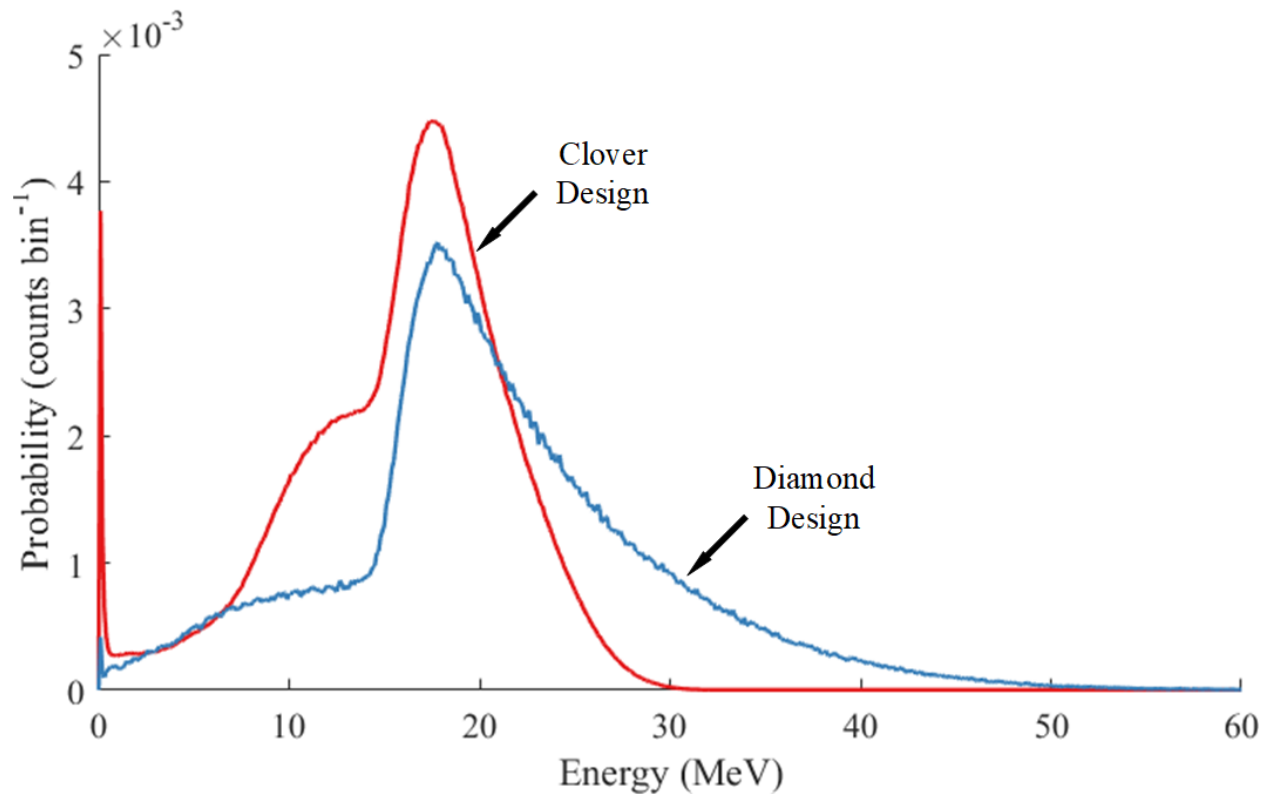


Fig. 3-36. Energy deposition spectra comparison between diamond and clover design with energy bins widths of 100 keV.

### 3.4 Fissile Coating Selection

Various fissile coatings were considered to study and balance the neutron sensitivity and detector lifetime. Due to the neutron environment in the reactor core, fissile coatings must be selected to have the necessary isotopic composition and mass to properly respond to the incident neutron flux magnitude and energy spectrum. There is a delicate balance between enrichment,

mass, and thickness. The enrichment of the fissile layer will skew the neutron energy response of the detector. For example, a depleted uranium ( $< 0.2\%$   $^{235}\text{U}$  by weight) fissile layer will have a neutron flux response dominated by fast neutrons, whereas a HEU ( $>20\%$   $^{235}\text{U}$  by weight) layer will have a response dominated by thermal neutrons. Too much mass and the detector will be too sensitive at high powers and may lead to signal saturation. Conversely, too little mass will lead to insensitivity in lower neutron flux environments. There is a practical limit for the thickness of the fissile layer, beyond which self-shielding will limit the number of fission fragments that can escape the fissile layer and enter the detector gas volume. Due to the dimensional limitations of the MPFD construction, mass and thickness of the fissile layer is coupled and will be described in further detail in Section 3.4.2.

### **3.4.1 Isotope Selection**

Fission chambers can utilize a large list of various fissile, fissionable, and fertile isotopes to balance the fission rate longevity and sensitivity of the detector. A list of common isotopes which can be used is provided in. Other isotopes, such as those of americium, curium, berkelium, and californium can be used as the reactive material in fission chambers but are rare with limited availability. These heavy elements and their isotopes are usually generated in nuclear reactor fuel, chemically separated, and refined. Due to the impracticality of procuring these rare isotopes, MPFD construction was limited mainly to natural Th, and varying enrichments of  $^{235}\text{U}$  (i.e., depleted, natural, 19.65%, and 93%). To illustrate the neutron-energy sensitivity of the material, effective microscopic cross sections are given for the various isotopes in Fig. 2-13. As demonstrated in previous work, detector response longevity can be stabilized by creating mixtures of fissile and fertile isotopes. As the detector operates and burns fissile material, the fertile isotopes will absorb neutrons, transmute into fissile material, which maintains the neutron sensitivity of the detector [70], [87].

Table 3-8. Table of fissile, fertile, and fissionable isotopes.

| Isotope*                | Natural Isotopic Abundance | $\sigma_{\text{Thermal}}$ (barns)<br>[1E-5 $\rightarrow$ 10 eV] | Res. Int. (barns)<br>[0.5 $\rightarrow$ 1E+5 eV] | $\sigma_{\text{Fast}}$ (barns)<br>[1 $\rightarrow$ 2E+4 keV] | Wescott<br>g-factor |
|-------------------------|----------------------------|-----------------------------------------------------------------|--------------------------------------------------|--------------------------------------------------------------|---------------------|
| <b><sup>232</sup>Th</b> | 100.00%                    | $2.5 \times 10^{-6}$                                            | $5.47 \times 10^{-6}$                            | 0.075                                                        | -                   |
| <sup>233</sup> Th       | -                          | 15.02                                                           | 114.6                                            | 0.101                                                        | 1.00062             |
| <sup>234</sup> Th       | -                          | $5.01 \times 10^{-3}$                                           | $1.91 \times 10^{-3}$                            | 0.034                                                        | 1.0011              |
| <b><sup>233</sup>Pa</b> | -                          | N/A                                                             | $2.93 \times 10^{-6}$                            | 0.233                                                        | -                   |
| <sup>233</sup> U        | -                          | 532.3                                                           | 766.9                                            | 1.91                                                         | 0.9966              |
| <b><sup>234</sup>U</b>  | 0.01%                      | 0.066                                                           | 0.703                                            | 1.181                                                        | 0.9906              |
| <sup>235</sup> U        | 0.70%                      | 574.9                                                           | 273                                              | 1.227                                                        | 0.9799              |
| <sup>236</sup> U        | 0.00%                      | 0.047                                                           | 4.462                                            | 0.576                                                        | 1.0044              |
| <sup>237</sup> U        | -                          | 1.674                                                           | 43.36                                            | 0.639                                                        | 0.9833              |
| <b><sup>238</sup>U</b>  | 99.29%                     | $1.85 \times 10^{-5}$                                           | $2.02 \times 10^{-3}$                            | 0.306                                                        | 1.0016              |
| <sup>239</sup> U        | -                          | 14.47                                                           | 464.1                                            | 0.382                                                        | 1.0169              |
| <b><sup>237</sup>Np</b> | -                          | 0.02                                                            | 0.635                                            | 1.333                                                        | 0.9762              |
| <sup>238</sup> Np       | -                          | 222.6                                                           | 1090                                             | 1.46                                                         | 1.0111              |
| <sup>239</sup> Np       | -                          | 0.028                                                           | 0.013                                            | 0.582                                                        | 1.0018              |
| <b><sup>238</sup>Pu</b> | -                          | 16.67                                                           | 19.92                                            | 1.934                                                        | 0.9381              |
| <sup>239</sup> Pu       | -                          | 786.4                                                           | 298.8                                            | 1.792                                                        | 1.0522              |
| <b><sup>240</sup>Pu</b> | -                          | 0.058                                                           | 3.391                                            | 1.342                                                        | 1.0323              |
| <sup>241</sup> Pu       | -                          | 1060                                                            | 560.6                                            | 1.601                                                        | 1.047               |
| <sup>242</sup> Pu       | -                          | 0.014                                                           | 0.232                                            | 1.139                                                        | 0.9977              |

\*Bolding indicates fertile isotopes

\*\* Cell color shading indicates primary fission energy range for isotope

### 3.4.2 Mass & Thickness Considerations

The MPFD sensitivity can be customized by selecting the total mass and the thickness of the neutron reactive layer. Based on the electrodeposition process, the most probable compound deposited is uranium dioxide (UO<sub>2</sub>) which has a nominal density of 10.97 g cm<sup>-3</sup>. With the assumption that the material deposited uniformly coats the platinum electrode, Eq. (10) can be used to determine the total mass as a function of electrode radius,

$$m = \rho h \pi r^2 \quad (10)$$

The mass of the fissile layer is limited in a practical sense by the thickness. As part of simulations described in Section 3.1.3, varying layer thicknesses were simulated to determine the effect on the detector performance. As demonstrated in Fig. 3-37, the rate at which fission fragments enter

the chamber gas is proportional to fissile-layer mass up to approximately 0.45  $\mu\text{g}$  of  $^{235}\text{U}$ , corresponding to 1.0  $\mu\text{m}$  of  $\text{UO}_2$ , at which point self-shielding begins to increase appreciably. Therefore, layer thicknesses greater than 1.1  $\text{mg cm}^{-2}$  mass-thickness of  $\text{UO}_2$  should be avoided, as the neutron sensitivity is no longer proportional to mass, and a larger fraction of fission fragment energy is lost in the fissile layer in the form of heat.

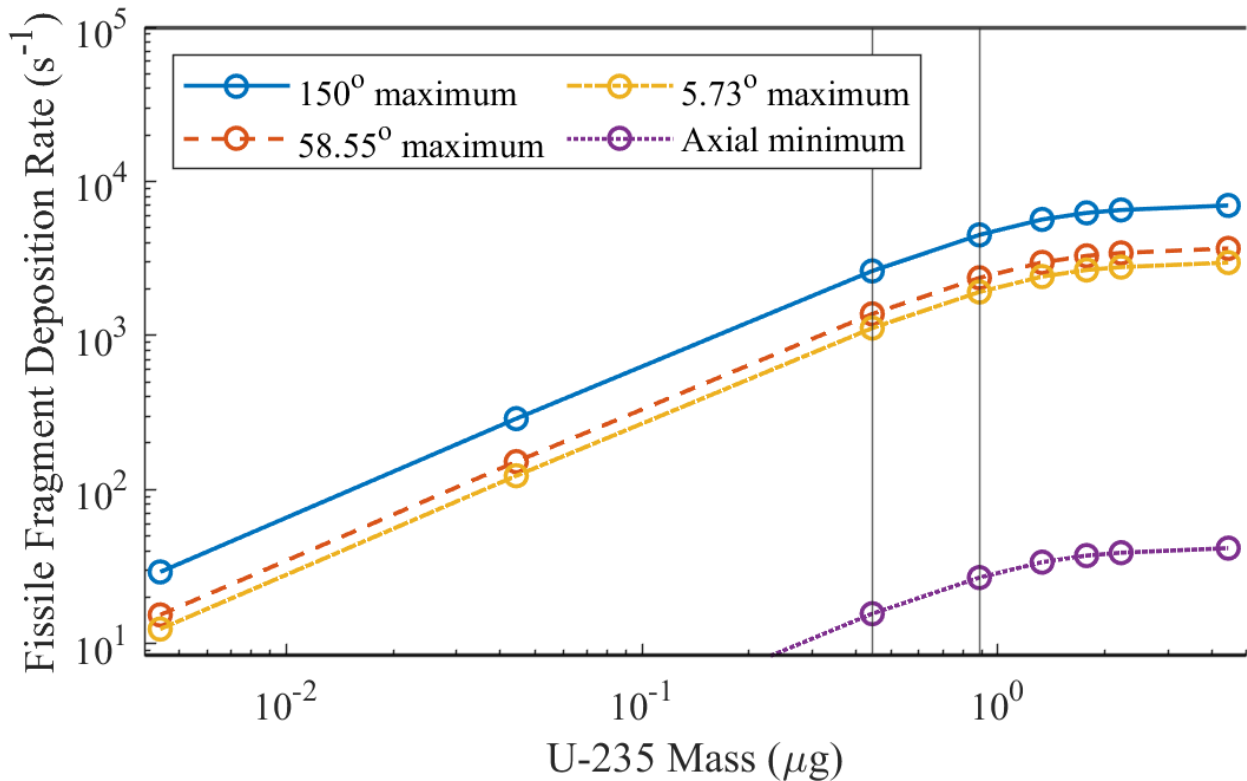


Fig. 3-37. Fission rate as a function of fissile-layer mass. Included for comparison are the electronics limit (top boundary of graph) and the fission-rate limit for 1% error within 20-minute measurement (bottom boundary of graph). Vertical lines indicate the fissile material masses equivalent to thicknesses of 1 micron (left) and 2 microns (right) [79].

Included for reference are limiting factors, such as the MPFD detector electronics count rate limit of 100 kHz, and the minimum count rate necessary to achieve a 1% counting uncertainty for a 20-minute measurement.

This chapter focuses on what materials are used to manufacture the MPFD. Starting with a description of the electrode manufacturing process, various techniques are discussed that improved manufacturability of the Ti/Pt electrode. The process of electrodepositing the fissile material is described, along with discussion about various methods to confirm layer uniformity and mass. Finally, the chapter concludes with discussion about the assembly process of the MPFD array, mention of post-assembly quality tests, and practical considerations for experimental deployments into a reactor.

### 4.1 Electrode Manufacturing

Over the course of the research presented herein, several attempts were made to develop an accurate and repeatable method of manufacturing the platinum plating electrode needed to deposit the fissile layer upon. Based on previous MPFD work, the legacy production method included a 50 Å thick evaporation of titanium onto an alumina substrate followed by 500 Å of platinum. This layer of titanium and platinum is referred to as the Ti/Pt plating electrode.

#### 4.1.1 Electron-Beam Evaporation

The current method of manufacturing the Ti/Pt plating electrode utilizes electron beam evaporation located in a class-100 clean room which is part of the Semiconductor Materials and Radiological Technologies Laboratory (SMARTlab). During this process the MPFD disk substrate is loaded into the electron-beam evaporator and coated with the titanium and platinum layers. This process operates as a line-of-sight deposition method that will coat the entire substrate surface unless a method of selective deposition is used (such as shadow masks or photoresist patterning). Both masking methods have advantages and drawbacks, and both are described in further detail in the following sections

##### Shadow-mask Method

Shadow masks are sacrificial substrates that are used to mask a substrate to allow for preferential patterning of evaporated material. Analogous to how an object into front of a light source will cast a shadow onto a surface, a shadow mask operates under the same principle. Areas that are shadowed by the shadow mask will not have direct line-of-sight to the evaporation source

and will therefore not be coated. The shadow mask will protect the substrate at the expense of intercepting the coating material instead. This coating can build up around features and can eventually distort the pattern for which it was designed to produce.

Shadow masks of multiple materials and patterns were employed during this research. Among these variations, stainless steel, aluminum, and 3D printed plastic were used for manufacturing the shadow mask. Masks were designed and implemented for full wafer or separated disks, as shown in Fig. 4-1. Early versions of 3D-printed shadow masks were made from high temperature plastic, RGD525, but later Acrylonitrile Styrene Acrylate (ASA) was used due to its higher heat deflection temperature. As noticed in previous work, these plastics can warp from heat and cause distortions during the evaporation [85]. Additionally, small details in the plastic shadow masks commonly did not produce the original designs either due to the printing process or handling thereafter. Machined-metal shadow masks were later adopted, due to higher production reliability. Key advantages of the 3D printed shadow masks include rapid turnaround in manufacturing, and no-need for additional post process machining.

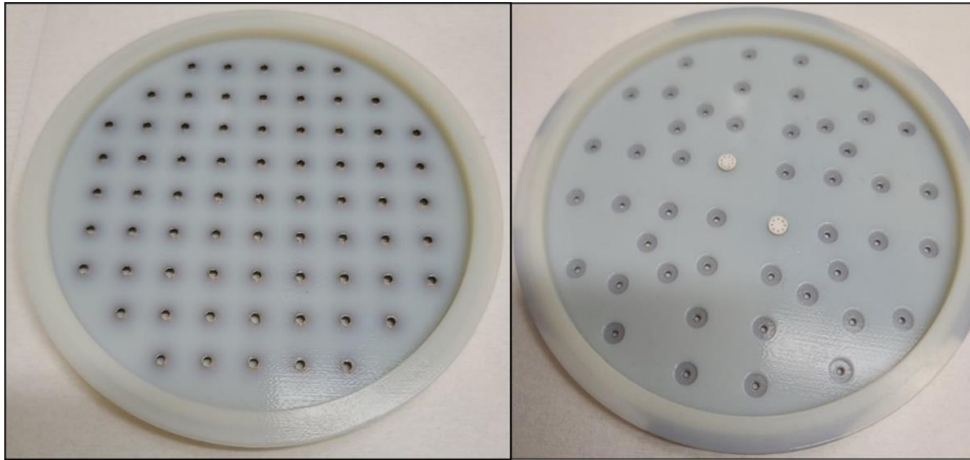


*Fig. 4-1. Images of two versions of the 3D-printed shadows masks. A full wafer version (left) and an individual disk (right) version.*

For metal shadow masks, initial experiments were conducted with stainless steel shim stock machined with a micro mill. A resulting mask example is shown in Fig. 4-2. Aluminum shadow masks were also tested and had similar performance as stainless steel. Both metal shadow mask materials had much better temperature tolerances, with no observed warping during the evaporation process. The main disadvantage of metal shadow mask is that they require precise machining and are limited to the accuracy of available drill bit sizes and reamers.

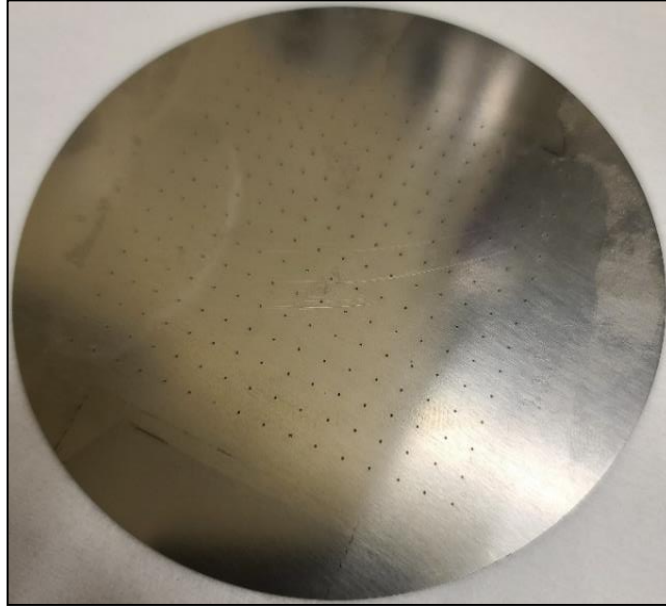


*Fig. 4-2. Full wafer metal shadow mask. Commonly, metal shadow mask holes would have edge burs due to the drilling process.*

During initial manufacturing, it was observed that features cut with the micro mill created edge effects such as shape distortion, tearing or burs due to the small thickness of the shim stock (0.001” – 0.01”). To resolve this issue, sanding was required which could result in additional damage to the pattern shape. Both plastic and metal shadow masks proved to be quick method of prototyping shadow mask geometries, with little specialized manufacturing experience required, but it was determined that both methods would only be suitable for pattern geometries significantly larger than those required for the MPFD project. As a result, photolithography processes were developed and later adopted due to higher achievable accuracy.

### **Photoresist Method**

Photoresist is a light-sensitive emulsified polymer that will either cross-link and strengthen (positive resist) or deteriorate (negative resist) in the presence of ultra-violet light (UV). Polymer that is not cross-linked will have a high solubility. This light-sensitive polymer is commonly used for Very Large-Scale Integration (VLSI) processing and can achieve high-dimensional accuracy down to single-nanometer resolution depending on the light source used. The Ti/Pt electrode diameter commonly ranges from 0.25—2 mm, therefore, the photolithography process is well suited for these dimensions. Photoresist can either be applied as a liquid solution or as a dry-film. The entire photolithography process with metallization is illustrated in Fig. 4-3.

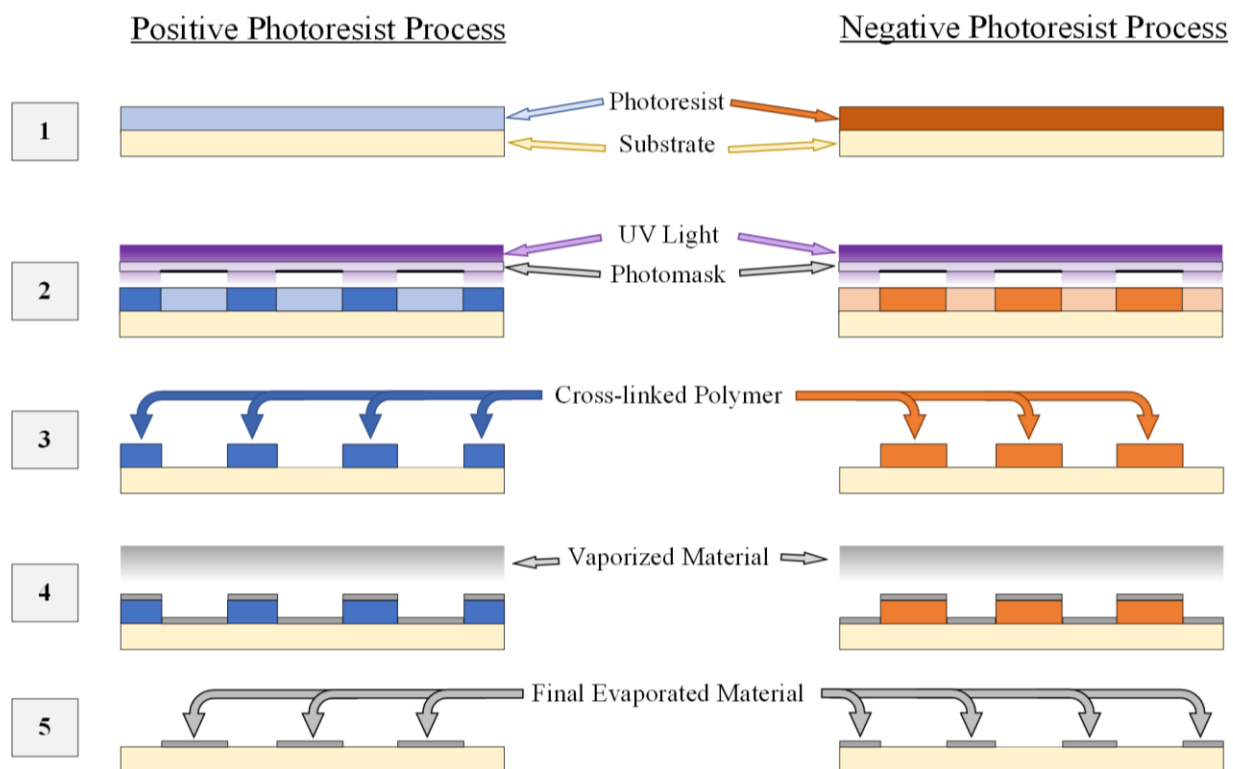


Fig. 4-3. Metallization process for both positive (left) and negative (right) photoresist. (1) Photoresist is applied to substrate, (2) A photomask is used to preferentially expose photoresist to UV light, (3) material is submerged in detergent which will preferentially remove soluble polymer, (4) material is applied to surface via metal vaporization, (5) remaining photoresist is stripped off surface, leaving only metal attached to substrate.

A negative photoresist (AZ 2070) was utilized, and a recipe was developed (see Appendix B) that allows full wafer patterning of Ti/Pt electrode features. Results of the process show a high degree of accuracy, both in the diameter and circularity of the feature and as shown in Fig. 4-4.

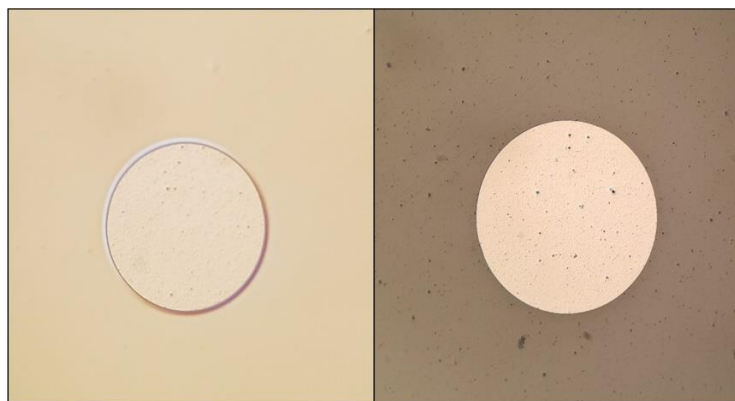


Fig. 4-4. Microscope images of 0.762 mm diameter Ti/Pt electrode feature after photoresist development (left), and after metallization and liftoff (right).

Two wafers were used to test the reliability of the Ti/Pt evaporation with the photolithographic process. Four Ti/Pt electrodes from each wafer in the four quadrants were measured to determine uniformity using a SEM. The Ti/Pt electrodes were determined to have an average diameter of  $0.756 \pm 0.005$  mm on the first wafer, and  $0.759 \pm 0.005$  mm on the second wafer. The first wafer is shown in Fig. 4-5 with crosshairs located in the center and periphery of the wafer. These crosshairs are used for alignment during the laser waterjet cutting process. The laser waterjet cutting process utilizes a high-pressure column of laminar flow water as a lightguide for a laser to cut substrates. The process shares similarities with laser cutting but has higher precision and when utilized, a single wafer can yield a high volume of MPFD pieces. For example, MPFD disks with an outer diameter of 3.175 mm can be patterned onto a 100 mm diameter alumina wafer yielding 600 disks. For comparison, previous shadow masks, such as those depicted in Fig. 4-1 and Fig. 4-2 were limited to between 48 and 256 disks per evaporation with relatively low consistency of the Ti/Pt electrode dimensions.

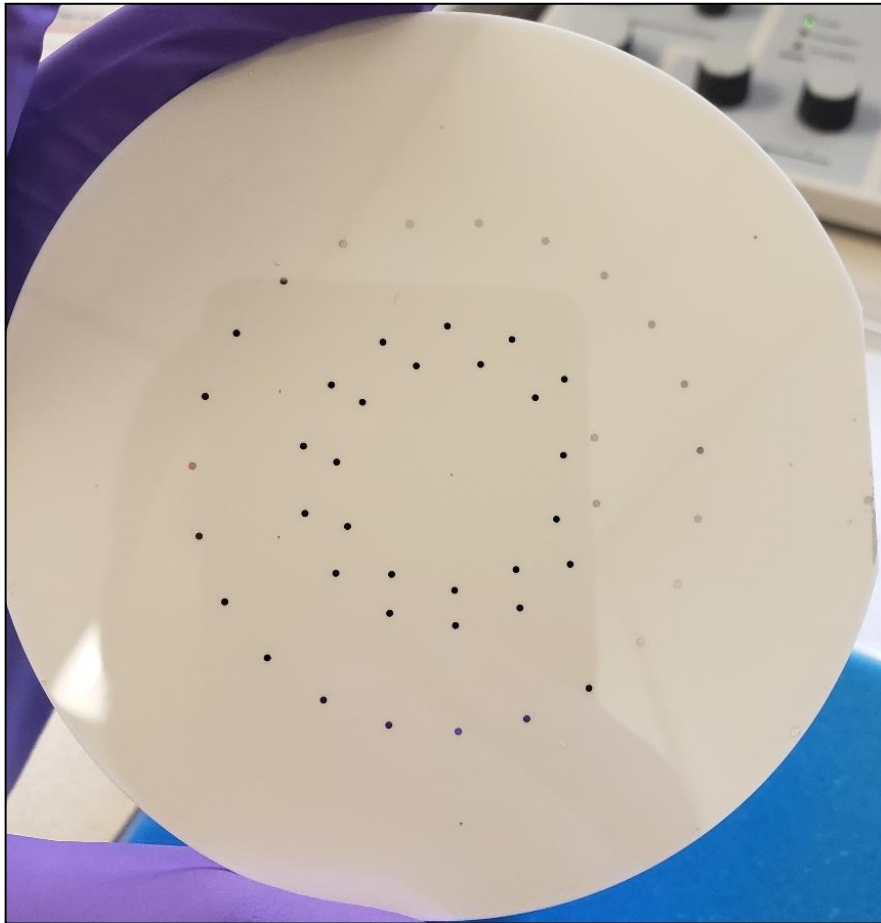


*Fig. 4-5. Ti/Pt evaporation on alumina wafer using the photolithography process.*

#### **4.1.2 Electrode Recycling (Aqua Regia)**

An aqua regia etch process was developed and used to recycle samples which had either sub-optimal electrode geometries or had suboptimal uranium coatings after the electrodeposition. The aqua regia process included 1:3 ratio of nitric acid to hydrochloric acid by molarity with 60°C of heat applied with a hot plate. Aqua regia was utilized for two reasons. First, the nitric acid dissolves any uranium, if present, on the electrode. Second, aqua regia is one of a small set of acidic solutions that can efficiently dissolve platinum group elements (also known as noble metals). The process originally included the use of hydrofluoric acid after the aqua regia bath to dissolve the titanium adhesion layer, but after initial testing, it was found that the titanium layer was sufficiently dissolved by the aqua regia bath.

Samples were submerged within the solution for 24 hours or until the electrode coating was visibly removed. Aqua regia produces chlorine ( $\text{Cl}_2$ ), nitrogen dioxide ( $\text{NO}_2$ ), and nitrogen oxide ( $\text{NO}$ ) fumes during the process and therefore all sample dissolving baths were conducted inside a fume hood and with a loose fit beaker cover. After the process was completed, samples were rinsed with deionized water, to ensure all residual acid was removed, and continuity checks were performed on the substrate surface to ensure the electrode was sufficiently dissolved and removed. It is important to note that this process was performed only on alumina substrate samples for two reasons. First, due the costlier nature of alumina in comparison to silicon, it was imperative to recover and reuse as many samples as possible. Second, the chemical resistance of alumina was sufficient to not merit concern when exposed to nitric and hydrochloric acid. After this process was completed, samples were designated as refurbished and were reused in the manufacturing process.

### **4.2 Electrodeposition Process**

Based on previous MPFD research, a cyclic-voltammetric electrodeposition process was chosen to apply the fissile layer on the Ti/Pt electrode. This process requires a uranium ion rich solution and the use of three electrodes (counter, reference, and working) to drift and adhere uranium compounds to the surface of the platinum electrodes.

### 4.2.1 Solution Preparation and Sample Deposition

Previous work has been published which described the process through which the electrodeposition was performed [42], [85], [88], [89]. Uranium or thorium electrolytic solutions are composed of various components listed in Table 4-1. A solution preparation recipe is provided in Appendix C.

*Table 4-1. Constituent parts of the uranium or thorium electrodeposition solutions.*

| Chemical                   | Vendor                                        | Formula                                                                            | Mol wt<br>(g cm <sup>-3</sup> ) | Density<br>(g cm <sup>-3</sup> ) | Qty     | units |
|----------------------------|-----------------------------------------------|------------------------------------------------------------------------------------|---------------------------------|----------------------------------|---------|-------|
| Thorium Nitrate Hydrate    | Strem Chemicals, Inc.                         | Th(NO <sub>3</sub> ) <sub>4</sub><br>(H <sub>2</sub> O) <sub>x</sub>               | 480.06                          | N/A                              | 51.25   | mg    |
| ----- or -----             |                                               |                                                                                    |                                 |                                  |         |       |
| Uranyl Nitrate Hexahydrate | International Bio-Analytical Industries, Inc. | UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub><br>(H <sub>2</sub> O) <sub>6</sub> | 502.13                          | 2.807                            | 51.25   | mg    |
| ----- and -----            |                                               |                                                                                    |                                 |                                  |         |       |
| Ammonium Nitrate           | Fisher Scientific                             | NH <sub>4</sub> NO <sub>3</sub>                                                    | 80.43                           | 1.725                            | 1801.25 | mg    |
| Deionized Water            | -                                             | H <sub>2</sub> O                                                                   | 18.02                           | 0.998                            | 12.5    | mL    |
| Ethanol                    | Fisher Scientific                             | CH <sub>3</sub> CH <sub>2</sub> OH                                                 | 46.07                           | 0.805                            | 1.25    | mL    |
| Final Volume               |                                               |                                                                                    |                                 |                                  | 18.75   | mL    |

The MPFD electrodeposition process, like industry standard processes, deposits thin films of various actinides, with a few caveats. Common electrodeposition methods utilize a conductive planchette such as stainless steel or platinum as the cathode surface to which actinides will adhere. Electrodeposition of these planchettes is relatively straightforward and simple cell geometries have been developed where the disc is located at the bottom of a sealed cylinder [90], [91]. Due to the MPFD Ti/Pt electrode's small size and on the alumina substrate, a modified cell geometry has been required. Most recent MPFD electrodepositions have been conducted in a cell consisting of a beaker or petri dish, with the reference, counter and working electrode entering the cell via the top of the beaker as illustrated in Fig. 4-6. While this setup has produced usable fissile layers on MPFD substrates, it has several key drawbacks.

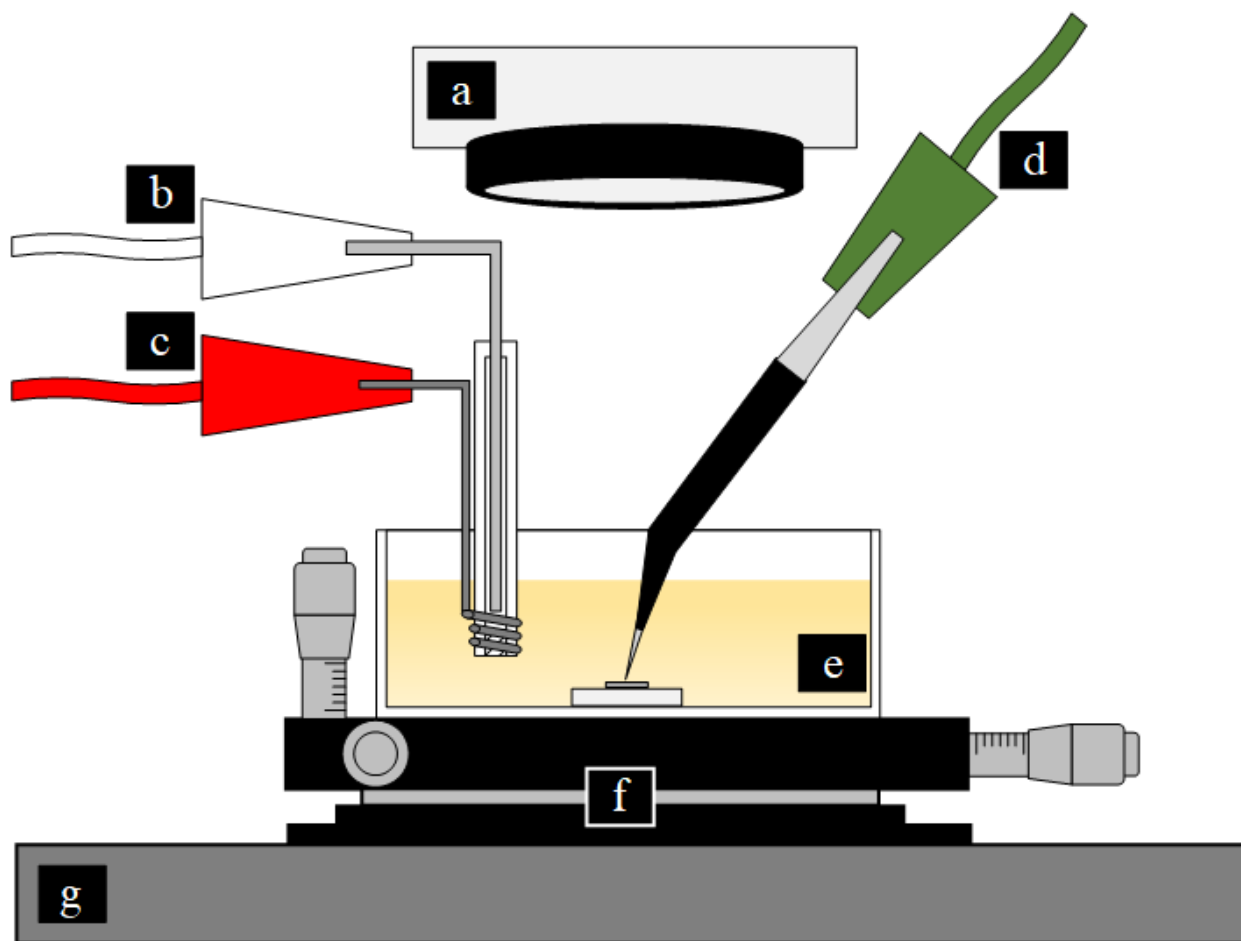


*Fig. 4-6. Illustration depicting the electrodeposition setup with various components. (a) digital microscope, (b) clip to the Ag/Ag-Cl reference electrode, (c) clip to platinum counter electrode coiled around reference electrode, (d) clip to tungsten working electrode which is in contact with Ti/Pt electrode, (e) uranium solution, (f) xyz manipulation stage, and (g) granite stability block.*

First, a 5  $\mu\text{m}$  tungsten micro-tip probe serves as the working electrode to contact the Ti/Pt electrode. This probe tip must be entirely coated with a chemically resistant material with only the tip bare as to avoid uranium ion accumulation on the probe rather than the Ti/Pt electrode. The tungsten probe must be delicately lowered to contact the Ti/Pt electrode to avoid scratching the surface, but while also having enough downward pressure as to make sufficient electrical contact. This process was accomplished by using of a fine adjustment xyz manipulation stage under the deposition beaker, as shown in Fig. 4-6, to raise the beaker until the Ti/Pt layer contacts the probe. During electrodeposition, a digital microscope was used to confirm contact between the probe and the Ti/Pt layer. It has been observed that confirming electrical contact between the probe tip and Ti/Pt layer is difficult and results in inconsistent contact pressure for every sample.

Second, due to the beaker setup, samples would need to be inserted into the solution individually and positioned at the bottom of the beaker using carbon-tipped tweezers. Because of the various instruments deployed into the beaker simultaneously, great care was needed to avoid hitting the various components and altering the cell geometry. Of greatest concern was the distance between the working electrode and counter electrode as this determines the current density during the deposition process. Because of these tedious steps, consistent electrode spacing and therefore deposition conditions were difficult to maintain.

To quantify the effectiveness of the current electrodeposition setup, a set of six natural enrichment thorium and ten natural enrichment uranium samples were deposited and characterized. Each sample was deposited using the cyclic voltammetric process with 250 sweep cycles varying between -0.62 and -0.64 V. Fig. 4-7 and Fig. 4-8 show images of pre- and post-deposition for a uranium and thorium sample set, respectively.

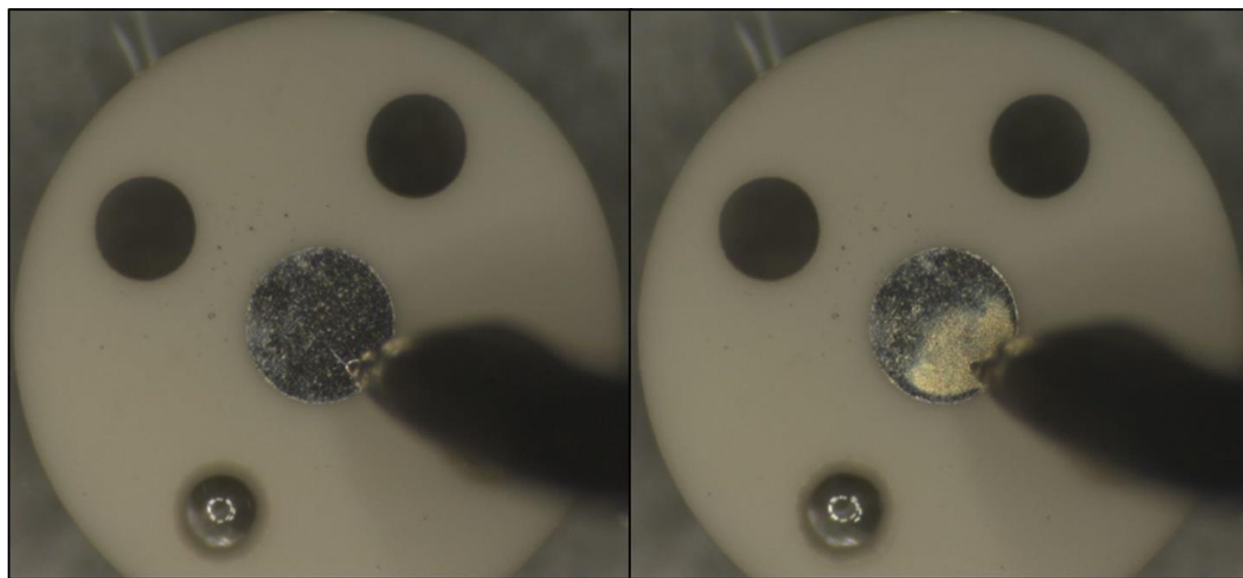


*Fig. 4-7. Images of pre-deposition (left) and post-deposition (right) of sample (MPFD.U.9).*

The actinide deposition is readily apparent in both sets of images, with the uranium sample having a slight gold hue and the thorium sample having a silver hue. Analysis of the sample masses was conducted with alpha spectrometry (discussed further in Section 4.2.2) to verify mass deposition. Analysis results of these samples are listed in Table 4-2, which yields an average mass of  $0.396 \pm 0.249 \mu\text{g}$  and  $0.228 \pm 0.149 \mu\text{g}$  for thorium and uranium, respectively.

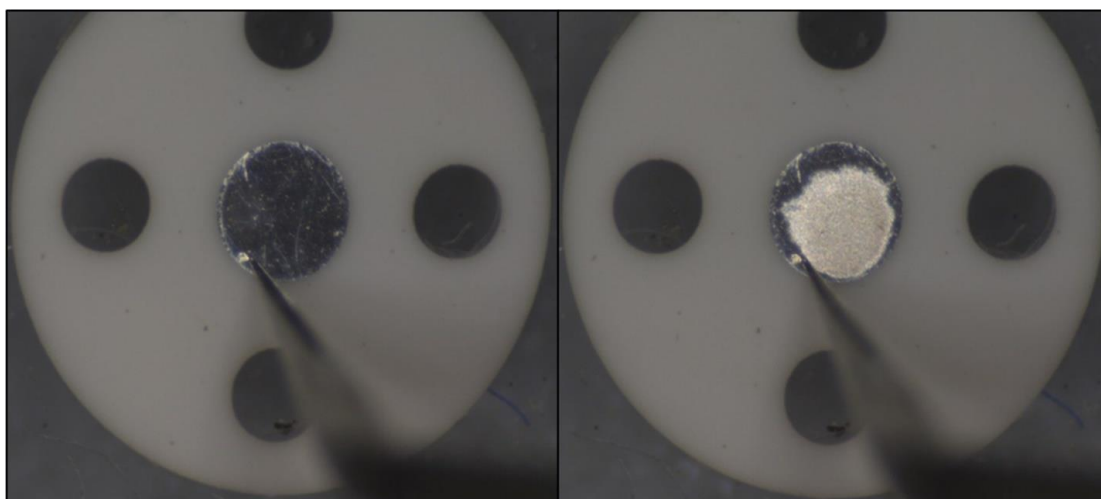


Fig. 4-8. Images of pre-deposition (left) and post-deposition (right) of sample (MPFD.Th.6).

Table 4-2. Summary of measured masses for six thorium and ten uranium samples. Note that there is one significant outlier for the thorium sample set (MPFD.Th.3) and two significant outliers for the uranium sample set (MPFD.U.3 and MPFD.U.5).

| Sample    | Chamber | Mass ( $\mu\text{g}$ ) | Error ( $\mu\text{g}$ ) | Abs Error |
|-----------|---------|------------------------|-------------------------|-----------|
| MPFD.Th.1 | AS_01   | 0.326                  | 0.014                   | 4.4%      |
| MPFD.Th.2 | AS_02   | 0.278                  | 0.013                   | 4.6%      |
| MPFD.Th.3 | AS_03   | 0.042                  | 0.007                   | 15.9%     |
| MPFD.Th.4 | AS_04   | 0.474                  | 0.016                   | 3.3%      |
| MPFD.Th.5 | AS_05   | 0.468                  | 0.015                   | 3.3%      |
| MPFD.Th.6 | AS_06   | 0.790                  | 0.020                   | 2.5%      |
| MPFD.U.1  | AS_07   | 0.187                  | 0.006                   | 3.0%      |
| MPFD.U.2  | AS_08   | 0.166                  | 0.005                   | 3.2%      |
| MPFD.U.3  | AS_09   | 0.029                  | 0.003                   | 9.3%      |
| MPFD.U.4  | AS_10   | 0.189                  | 0.007                   | 3.6%      |
| MPFD.U.5  | AS_11   | 0.061                  | 0.003                   | 5.5%      |
| MPFD.U.6  | AS_12   | 0.160                  | 0.005                   | 3.3%      |
| MPFD.U.7  | AS_13   | 0.248                  | 0.006                   | 2.5%      |
| MPFD.U.8  | AS_14   | 0.315                  | 0.007                   | 2.2%      |
| MPFD.U.9  | AS_15   | 0.472                  | 0.010                   | 2.1%      |
| MPFD.U.10 | AS_16   | 0.457                  | 0.009                   | 1.9%      |

The variability of the electrodeposition process is readily apparent from these results. Masses deposited have a high standard deviation which is a large concern when considering mass-production of MPFDs. Deposited sample mass can be affected by many variables that need to be controlled during the electrodeposition process. As mentioned previously the distance between the

counter and working electrode determines the current density on the working electrode. The conductivity of the electrolytic solution determines the resistance of the ED cell, and therefore the electrical current for a given applied voltage. Finally, the pH of the solution has a large impact on the actinide compound species present in solution for deposition. The solution pH also affects the electrolysis rate of the solution which can cause hydrogen bubble nucleation on the surface of the Ti/Pt layer, thereby reducing electrodeposition [90]–[94].

#### 4.2.2 Fissile Mass Confirmation Techniques

An important step in the MPFD fabrication process is to confirm the amount of mass electrodeposited on the Ti/Pt electrode. Historically, alpha-spectrometry methods, complimented with X-Ray Fluorescence (XRF) spectroscopy, were used to confirm the amount of mass deposited but additional methods were considered and investigated.

##### Alpha-Spectrometry Method

To accomplish alpha spectrometry an Ortec Alpha Suite™ with 16 individual alpha-spectroscopy chambers was used. These chambers utilize ion-implanted silicon detector heads (Ortec ENS8U300) for radiation detection and additional information can be found in the product data sheet [95]. Prior to usage, an energy/efficiency calibration procedure was performed for each chamber and is detailed in Appendix D. After calibration, samples were loaded into each chamber with the tray position nearest to the detector head as illustrated in Fig. 4-9.

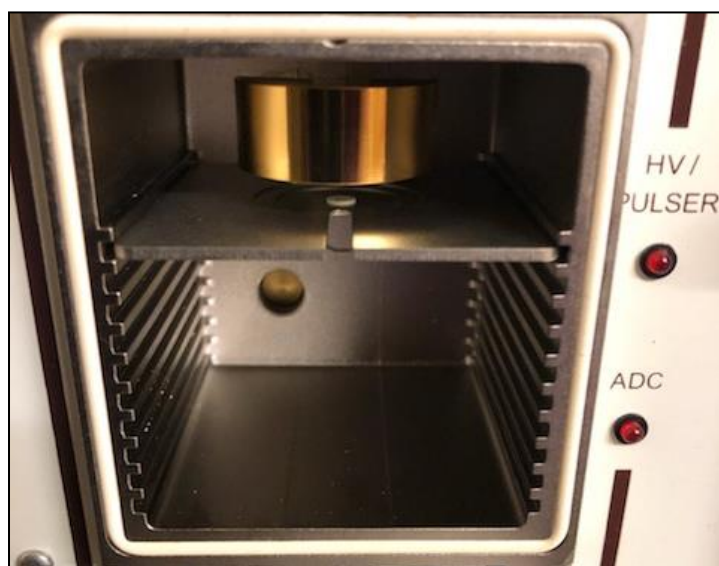


*Fig. 4-9. Image of MPFD sample positioning inside Alpha Suite™ chamber. Note the sample is placed as close as possible without contacting the detector head to avoid contamination.*

Sample measurement duration was tailored to achieve relative measurement error of  $< 1\%$  typically, based on the enrichment and mass deposited. Eq. (11) & (12) are used to determine mass, and mass uncertainty respectively,

$$m = \frac{\left[ \frac{G}{t_G} - \frac{B}{t_B} \right]_{ROI}}{\eta \Omega (SA)_x} \quad (11)$$

$$\sigma_m = \frac{\sqrt{\left[ \frac{G}{t_G^2} + \frac{B}{t_B^2} \right]_{ROI}}}{\eta \Omega (SA)_x} \quad (12)$$

where  $G$  is the gross counts,  $t_G$  is the gross measurement duration,  $B$  is the background counts,  $t_B$  is the background measurement duration,  $\eta$  is the intrinsic detector efficiency,  $\Omega$  is the fractional solid angle, and  $(SA)_x$  is the specific activity of the actinide enrichment. A summary of alpha-particle and gamma-ray energies and emission intensities for the given isotopes of uranium and thorium can be found in Table 4-3.

*Table 4-3. Summary of material/isotope emissions. Only the three most prominent alpha particles and gamma rays are shown for each isotope [34].*

| Material          | Nat. Iso. Abund.     | Spec. Activity (Ci g <sup>-1</sup> ) | $\alpha$ (keV) | Int (%) | $\gamma$ (keV) | Int (%)              |
|-------------------|----------------------|--------------------------------------|----------------|---------|----------------|----------------------|
| Nat. Th           | -                    | $2.2 \times 10^{-7}$                 | -              | -       | -              | -                    |
| <sup>230</sup> Th | 0.0002               | $2.1 \times 10^{-2}$                 | 4479.8         | 0.12    | 143.9          | 0.05                 |
|                   |                      |                                      | 4620.5         | 23.4    | 186.1          | 0.009                |
|                   |                      |                                      | 4687           | 76.3    | 253.7          | 0.01                 |
| <sup>232</sup> Th | 0.9998               | $1.1 \times 10^{-7}$                 | 3811.1         | 0.07    | 63.8           | 0.263                |
|                   |                      |                                      | 3947.2         | 21.7    | 140.9          | 0.021                |
|                   |                      |                                      | 4012.3         | 78.2    | -              | -                    |
| <sup>234</sup> U  | $5.0 \times 10^{-6}$ | $6.2 \times 10^{-3}$                 | 4603.5         | 0.2     | 53.2           | 0.123                |
|                   |                      |                                      | 4722.4         | 28.4    | 120.9          | 0.035                |
|                   |                      |                                      | 4774.6         | 71.4    | 455.0          | $2.5 \times 10^{-5}$ |
| <sup>235</sup> U  | 0.007                | $2.2 \times 10^{-6}$                 | 4215.8         | 6.01    | 19.6           | 63                   |
|                   |                      |                                      | 4364.3         | 18.9    | 143.8          | 10.96                |
|                   |                      |                                      | 4395.4         | 57.7    | 185.7          | 57                   |
| <sup>236</sup> U  | $1.0 \times 10^{-5}$ | $6.5 \times 10^{-5}$                 | 4332           | 0.15    | 49.5           | 0.078                |
|                   |                      |                                      | 4445           | 26      | 112.8          | 0.019                |
|                   |                      |                                      | 4494           | 74      | 171.2          | $6.2 \times 10^{-5}$ |
| <sup>238</sup> U  | 0.9929               | $3.4 \times 10^{-7}$                 | 4038           | 0.08    | 49.6           | 0.064                |
|                   |                      |                                      | 4151           | 21      | 113.5          | 0.0102               |
|                   |                      |                                      | 4198           | 79      | -              | -                    |

For both uranium and thorium deposited samples, an assumption is made that the electrodeposition solution only contains the starting elements with no decay daughter elements present. For uranium samples, the dominant alpha-particle signal comes from the  $^{234}\text{U}$ , regardless of enrichment.

### **X-ray Fluorescence Analysis**

As a qualitative confirmation of both the relative mass content, and the mass distribution, a Hitachi S-3400N Scanning Electron Microscope (SEM) coupled with an XRF analysis system was used. This system, as depicted in Fig. 4-10, allows for both electron microscopy imaging and elemental composition analysis. It is important to note that isotope composition analysis cannot be determined using this system. An example of SEM imagery can be observed in Fig. 3-24.



*Fig. 4-10. Image of the Hitachi S-3400N SEM with coupled XRF analysis system.*

Several scanning features are available when using the XRF system. Analysis can be conducted to determine the x-ray spectrum of the sample including analysis of estimated elemental weight percentages as shown in Fig. 4-11. Additionally, a two-dimensional map of elements can be produced, showing the spatial distribution of the sample. As evident in Fig. 4-12, the uranium distribution can vary considerably across the Ti/Pt electrode. A halo-effect of highly concentrated deposition can be seen surrounding the periphery of the Ti/Pt electrode. This effect is most likely a result of the sharp edge of Ti/Pt layer near the edges of the electrode which causes preferential accumulation of uranium ions during electrodeposition. The sharp edge of the Ti/Pt layer is an artifact of the Ti/Pt electrode manufacturing process.

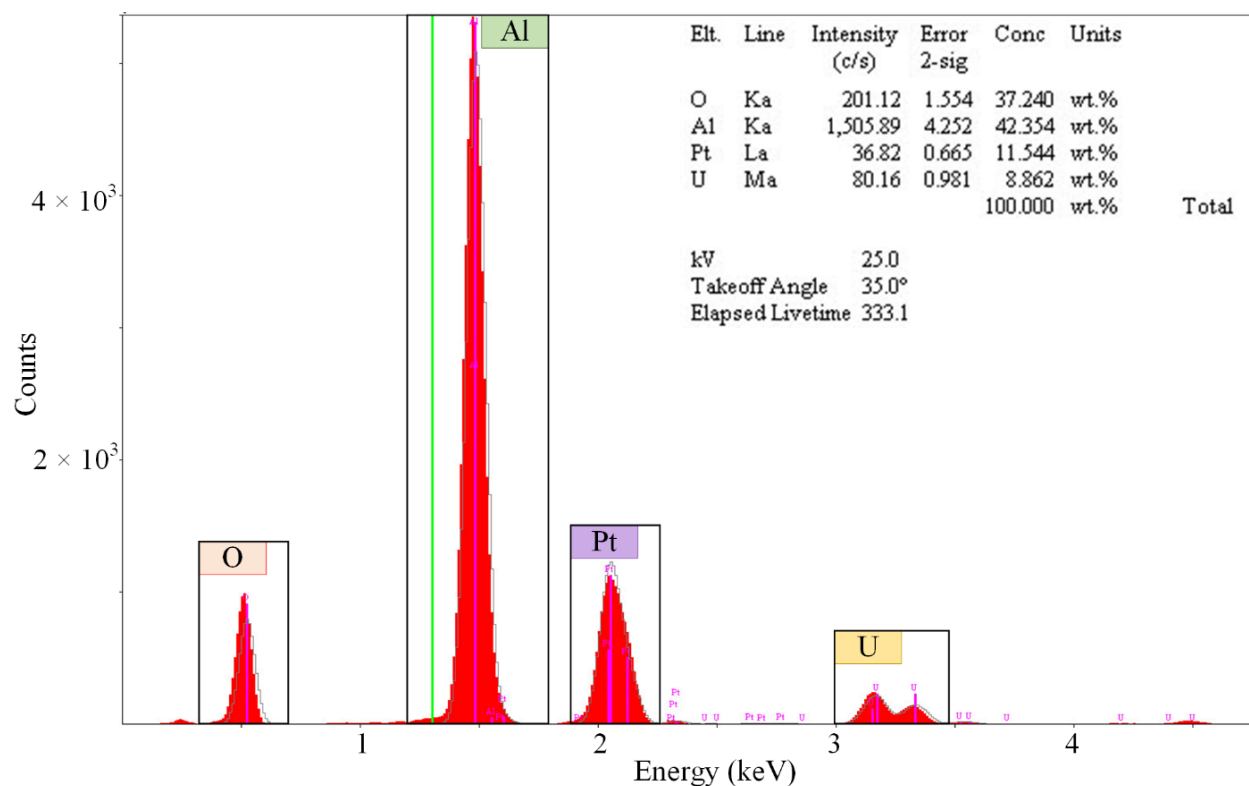


Fig. 4-11. X-ray spectrum collected from uranium-coated MPFD sample. Weight percentages indicated in upper right corner.

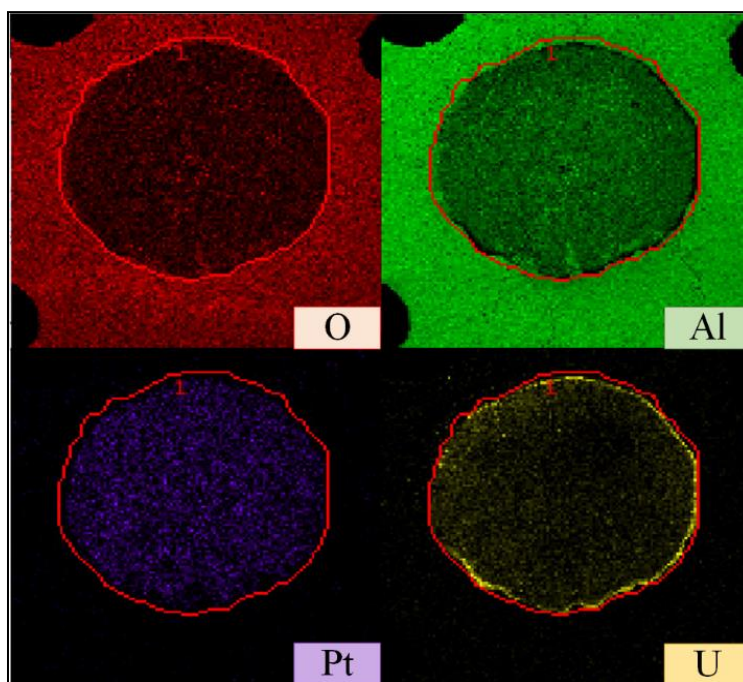


Fig. 4-12. Elemental map of MPFD sample showing distribution of oxygen, aluminum, platinum, and uranium.

### **Proposed Gamma-ray Spectrometry Method**

Peer-review publications have suggested the ability to determine mass and enrichment of uranium samples with gamma-ray spectrometry [96], [97]. The process shares similarities with alpha-particle spectrometry but relies on the gamma-ray emissions of the various actinide isotopes. A concern with this method is two-fold; firstly, the most prominent gamma rays emitted by uranium or thorium isotopes are lower energies (between 10-300 keV) and secondly, because characteristic x rays from these materials can blend with gamma-ray peaks in the collected spectra. A list of emitted gamma rays is provided in Table 4-3. Due to the low intensities of gamma rays emitted by uranium and thorium isotopes, gamma-ray spectrometry requires a significant amount of mass to achieve minimum detectable activity. For example, a 1 mg natural enrichment uranium sample will have a gamma-ray emission rate of  $3 \text{ s}^{-1}$  in comparison to an alpha-particle emission rate of  $25 \text{ s}^{-1}$ . Likewise, a 1 mg natural enrichment thorium sample will have a gamma-ray emission rate of  $0.01 \text{ s}^{-1}$  in comparison to an alpha-particle emission rate of  $8 \text{ s}^{-1}$ . These low emission rates coupled with the typically low mass of the MPFD samples makes gamma-ray spectrometry unsuitable for mass confirmation.

### **Laser and Optical Microscopy Analysis**

Laser and optical microscopy were also used to determine fissile layer deposition morphology. Electrodeposited MPFD samples were sent to the High Temperature Test Laboratory (HTTL) at INL for measurement. A Keyence VKX-250 was used to scan two samples to determine fissile layer thickness to quantify mass deposited. Scans of sample MPFD0006 and MPFD0010, corresponding to samples MPFD.U.6 and MPFD.U.10 respectively, are shown in Fig. 4-13. Several measurements were taken to determine the estimated fissile layer thickness. First, scans of the surrounding bare substrate were taken to determine surface roughness. Second, scans of the Ti/Pt electrode area with fissile deposits were taken.

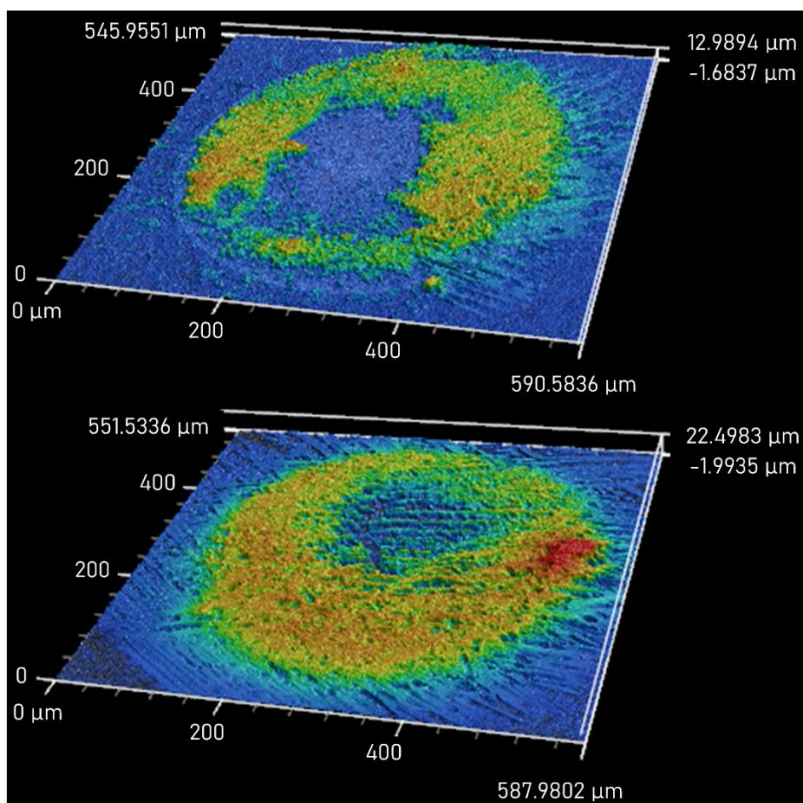


*Fig. 4-13. Topographic images of an MPFD fissile layer for MPFD0010 (top) and MPFD0006 (bottom). Images provided by Kevin Tsai.*

Finally, scans were taken of just the areas with visible fissile deposition. Thickness thresholds were then estimated to use as a delineation between the Ti/Pt layers and the fissile deposit layer. Based on the scan results it was concluded that the fissile layer deposit had an average thickness of 2.11  $\mu\text{m}$  and 2.52  $\mu\text{m}$ , and an estimated total coverage area of  $1.25 \times 10^5 \mu\text{m}^2$  (63.6%) and  $1.35 \times 10^5 \mu\text{m}^2$  (68.8%) for samples MPFD0010 and MPFD0006, respectively. Based on the alpha spectrometry of these samples, mass was determined to be  $811 \pm 9.92 \text{ ng}$  and  $810 \pm 9.80 \text{ ng}$  respectively, which yielded an estimated effective density of the fissile layer to be 3.06 and 2.38  $\text{g cm}^{-3}$ . This density has a few possible explanations. Electrodeposited materials on the Ti/Pt electrode are higher density compounds such as  $\text{U}_3\text{O}_8$  (8.3  $\text{g cm}^{-3}$ ) or  $\text{UO}_2$  (10.97  $\text{g cm}^{-3}$ ), but are organized in a porous structure, thus causing a lower effective density. Alternatively, this result may suggest that the compounds deposited may have been lower density materials such as uranyl nitrate (2.81  $\text{g cm}^{-3}$ ) or uranyl hydroxide (5.73  $\text{g cm}^{-3}$ ). Further investigation into the compound species is required to determine the fissile layer composition.

## 4.3 Assembly Process

An MPFD array can be configured to accommodate between 1- 4 nodes. The most common configuration presently used is the 4-node common cathode design, although other designs with a single node can be assembled using the diamond spacers. An overview of the assembly process will be discussed in the following sections along with the post-assembly purge/gas-fill process, and the post-assembly electrical tests.

### 4.3.1 Standard Configuration Procedure

The MPFD assembly is composed of seven components: the electrode wires, alumina chamber pieces, silica insulation, sheath, upper assembly, electrical feedthrough, and upper wire insulation. Prior to assembly, the crushable silica pieces are kept inside a desiccator to avoid adsorption of moisture. Additionally, the MPFD sheath interior is rinsed using isopropanol to remove any residual oil or contaminants leftover from the manufacturing process. The assembly of an MPFD array is as follows:

1. Electrode wires are cut and straightened to length of the sheath + length of upper assembly + 1-inch excess.
2. Wires are cleaned with a 3-step process of acetone, isopropanol, and deionized water.
3. Crushable silica capillary tube is threaded onto the electrode wires to the desired length.
4. Alumina pieces (blank disc, spacer, fissile disk) are threaded onto the electrode wires.
5. Alumina pieces are rinsed with isopropanol and dried with compressed N<sub>2</sub> gas to remove wire shavings.
6. Steps 3-5 are repeated until all nodes are threaded onto electrode wires (each node using a unique peripheral electrode wire to form the chamber).
7. Excess electrode wire at the top of the array is insulated using quartz fiberglass sleeving.
8. Electrode wires are threaded through upper assembly and soldered to electrical feedthrough.
9. Sheath is slipped over MPFD array internals and connected to the upper assembly.
10. Swagelok joints are tightened, and epoxy is added to the Lemo connector joints to create hermetic seal.

Various stages of the assembly process are shown in Fig. 4-14.

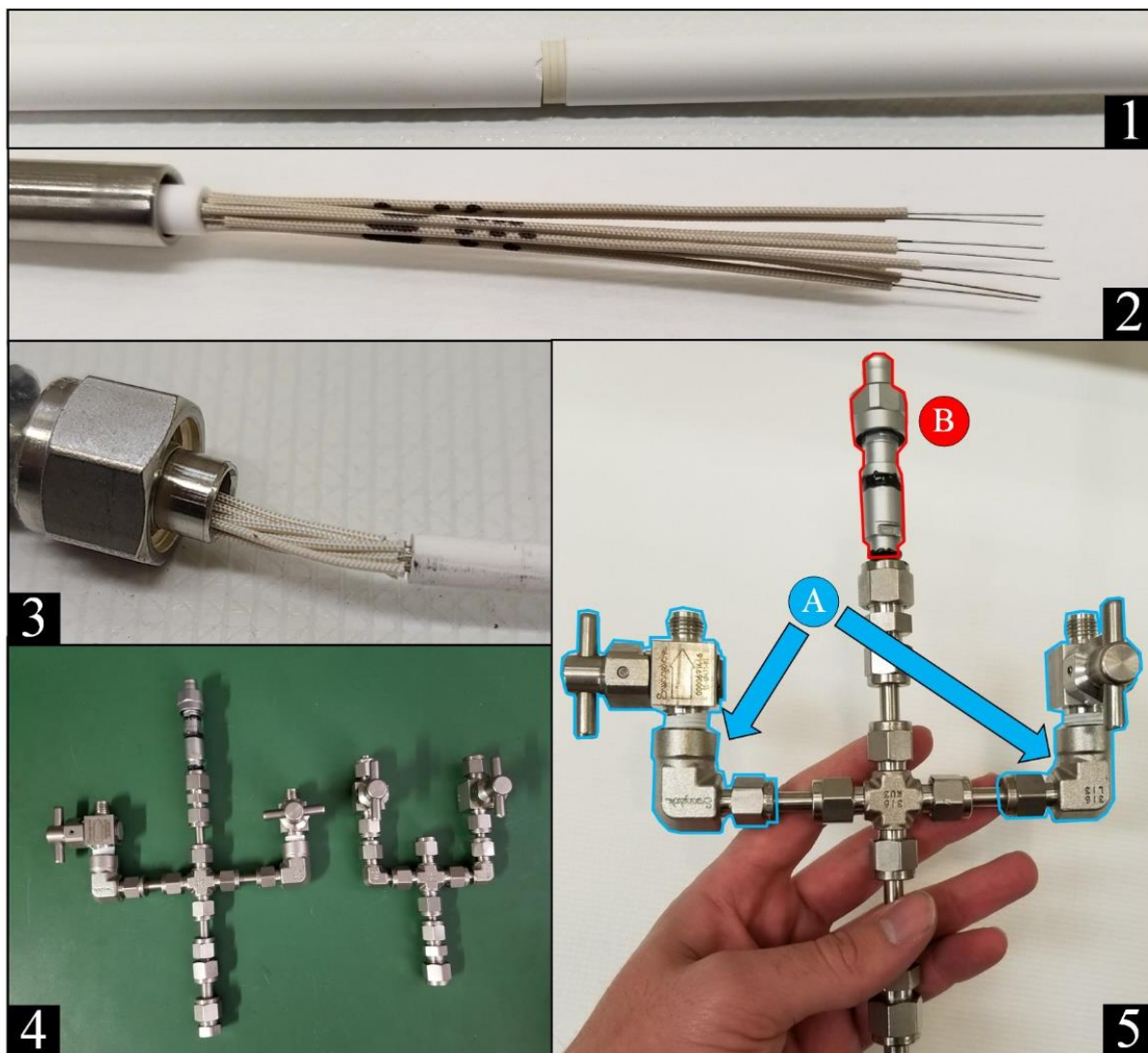


Fig. 4-14. Image (1) shows the crushable silica and alumina pieces threaded onto the electrode wires, (2) shows fiberglass insulation sleeves applied to the top of the electrode wires. (3) shows insulated wires threading through the upper assembly. (4) compares previous (left) and current (right) generations of the upper assembly. (5) shows the MPFD upper assembly with the purge/backfill valves (A) and the Lemo electrical feedthrough (B) with epoxy sealant.

The fiberglass quartz sleeving used for electrode wire insulation was Omega FBGS-N-30 non-fraying fiberglass sleeving. A two-part epoxy, Henkel Loctite™ Stycast 2651 was used to seal the electrical feedthrough joints.

#### 4.3.2 Evacuation and Gas-fill Process

After an MPFD is fully assembled, the internals must be purged of all contaminant gases and moisture. A process has been developed to aide in the removal of contaminants using external

heating and vacuum pumps. Completed MPFD arrays were inserted into a custom-fabricated heating tube and set to 170°C to aide in vaporizing residue moisture and to promote desorption of gases for the porous crushable silica insulation. The entire heated purge/backfill setup is shown in Fig. 4-15, with the various components labeled. This process included three cycles of 2-hour heated purge using a standard Edwards roughing pump. Between each heated purge, high-purity argon gas would be introduced into the detector assembly to a pressure of 60 psig to flush contaminant gases. After 3 cycles of rough-vacuum heating were completed, a 12-hour minimum heated cycle was performed using an Alcatel ATP 80/100 single stage turbo pump to achieve high vacuum ( $1 \times 10^{-6}$  Torr). After the 12-hour duration, the heating tape was turned off and the detector assembly was allowed to cool to room temperature while still under high vacuum. Finally, a valve connecting the turbo pump to the detector assembly would be closed, the turbopump switched off, and the array would be filled with the final gas backfill at desired pressure. Matheson Purity argon gas (99.9995%) was used for the final backfill due to its low oxygen and water content (< 1 ppm for both).

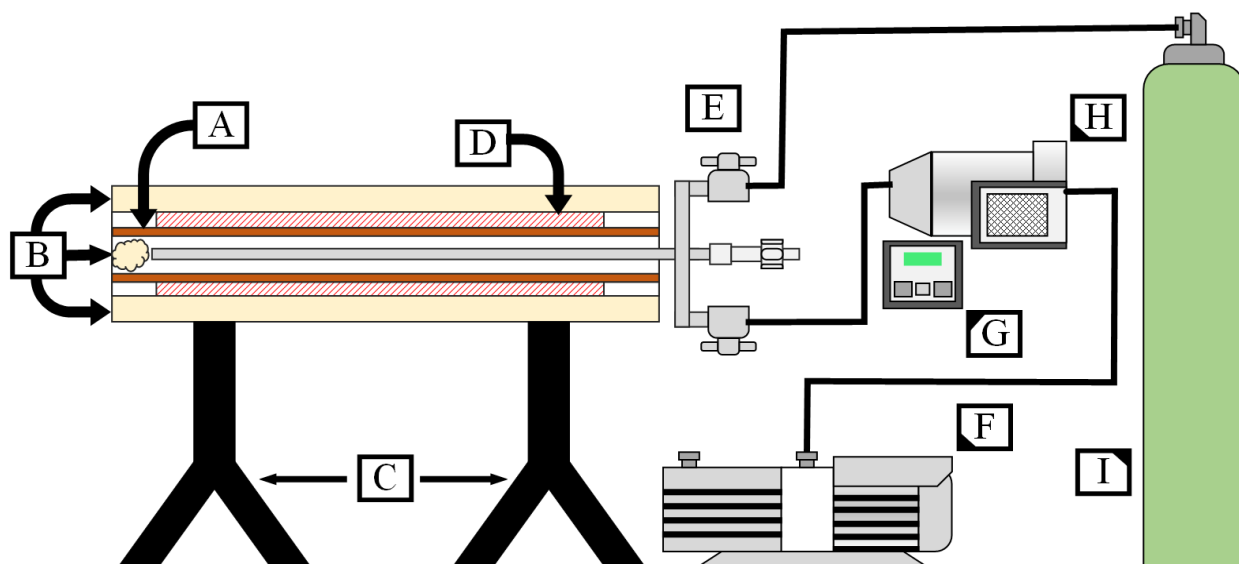


Fig. 4-15. Diagram of the heated purge and backfill setup. (A) copper tube, (B) quartz fiberglass insulation, (C) tube stands, (D) heating tape, (E) IFD upper assembly, (F) roughing pump, (G) turbo pump controller, (H) single stage turbo pump, (I) high-purity argon gas cylinder.

#### 4.3.3 Post-Assembly Fabrication Confirmation Methods

After assembly and backfill of the detector is completed, it is important to confirm the integrity of the internals prior to usage. Confirmation checks such as electrical continuity and

pressure integrity testing reduce the probability of spurious and erroneous measurements and provide for higher confidence that the detector will perform properly. These tests are essential and a procedure for testing each component was developed and used as a method of quality assurance (QA) for each manufactured assembly. Additionally, each assembly must be confirmed to have a hermetic seal prior to usage.

### **Electrical Testing**

To confirm detector electrical integrity, capacitance and resistance between electrode wires and the sheath is measured. Checks were performed using three different instruments: a Fluke 87V multimeter, an Agilent U1731C LCR meter, and an AEMC model 1026 megohmmeter. If an electrode shorts to either the sheath or another electrode, continuity checks using the multimeter will result in a low resistance reading ( $< 10$  Ohms). Using the LCR meter, capacitance between electrodes is measured (typically between 100 – 400 pF) and if a value varies beyond one standard deviation of the average of the other electrodes, it may indicate that a wire was kinked or damaged during assembly. Finally, resistance between wires can be confirmed using the megohmmeter to estimate leakage current during operation.

### **Pressure Testing**

Various methods of confirming hermetic sealing of the detector array were explored. Among these methods are soap-water bubbling, submersion bubbling and pressure longevity testing. When assembling the MPFD upper assembly, large gas leaks were quickly ascertained by using a mixture of Dawn dish soap and water and adding it on the various Swagelok joints. As gas leaked from the detector joints, bubbles would rapidly form and would be used to locate leaking joints which were then tightened to stop leakage. For minor leaks this method was not effective and therefore alternative methods such as submersion testing were required. Submersion testing required a large container into which the detector assembly was fully submerged. It was important for this test to be performed with positive internal pressure to avoid water leaking into the assembly, therefore an argon gas line was attached during the test. This test was typically conducted only on the upper assembly prior to attaching the sheath or electrical wires to avoid water damage to the internal components. Once all major and minor leaks are addressed, the MPFD assembly would be pressurized set to the desired pressure, isolated, and left for a 24-hour period.

If noticeable pressure reduction had occurred, additional examination of the array would be performed to determine the gas leak location.

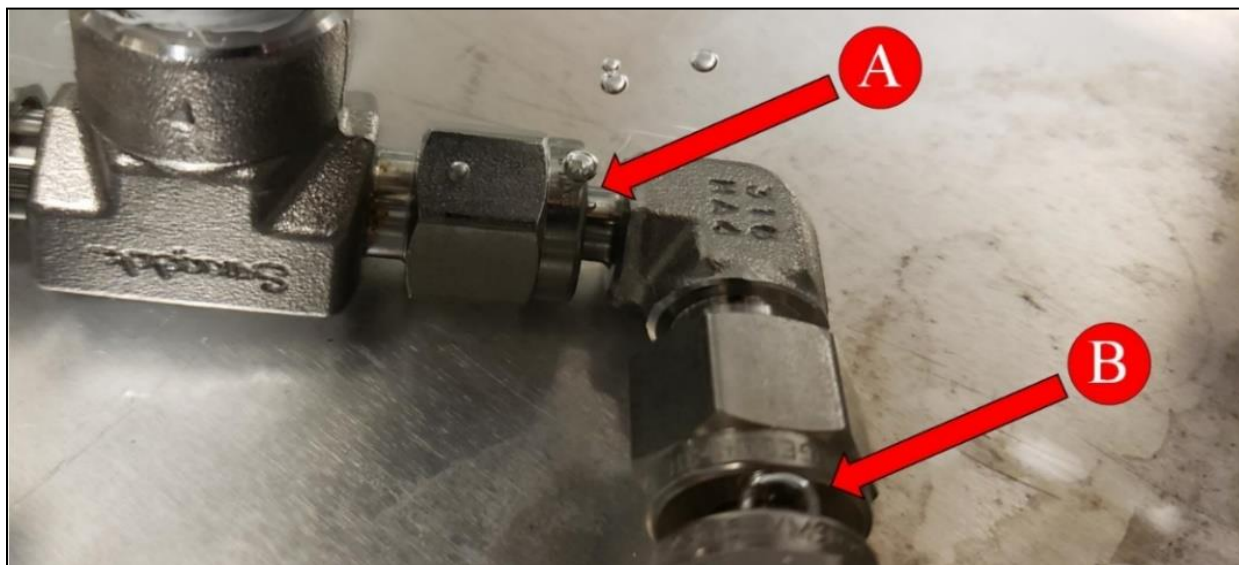


*Fig. 4-16. Image showing a submerge upper assembly with minor leaks at two different joints (A & B) which required additional tightening.*

## **4.4 Electrical Grounding Concerns**

During past experimental deployments, the MPFD sheath has been in direct contact with the metallic structure of the reactor core or structure. A significant amount of electromagnetic radiation and charged particles are produced as part of the fission process as described in Section 2.3 which results in current flow in the reactor structure that can have deleterious effects on MPFD performance. Additionally, during past experimentation, it has been observed that electromechanical reactor systems such as pumps, control rod drives, and high-voltage supply cables induced noise on the MPFD sensor and processing electronics [85]. Various techniques such as electronics grounding, the use of Faraday cages, and even shutting down reactor cooling have been employed to reduce these effects. Recently, electrically isolating the MPFD is being considered for future deployments that should also be employed to reduce these effects. To accomplish this task, various methods of isolating the MPFD such as applied permanent and temporary electrical insulation to the sheath are being considered.

#### **4.4.1 Permanent Coating Methods**

A first attempt was made to apply a ceramic compound coating to the detector exterior to reduce conductivity. This compound, Aremco Glass-Coat™, is a ceramic mixture that is first coated onto the metal sheath and then baked at high temperatures to induce hardening. This material is quoted as providing a dielectric breakdown of 1000 VDC/mil. A bake procedure requires air drying for 10-15 minutes, then heating to (200°F, 400°F, 1200°F) for 10-15 minutes. As this method employs a high-temperature furnace to bake and set the compound, this process must be completed prior to assembly.

Another method under consideration is to induce oxide growth on the sheath exterior. Known as anodizing, an oxide layer can be promoted to grow on the surface of the metal, causing an increase in resistivity. Anodizing also has the added benefit of increasing the durability of the metal sheath by creating a scratch-resistant surface. To perform anodizing, the MPFD sheath prior to assembly, would be submerged into an electrolytic solution such as KOH or NaOH with an anode material. A DC current would be passed into the cell causing oxide growth on the MPFD sheath [98]. As MPFD sheaths have varied in length from 1 – 6 feet in length, the solution volume would need to be large to accommodate the full length of the sheath.

#### **4.4.2 Sleeve and Standoff Component Methods**

Alternative materials for electrical insulation are commercially available. Insulating overbraid can be manufactured using materials such as 3M™ Nextel™ ceramic fibers which are primarily composed of  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$  with a small fraction of  $\text{B}_2\text{O}_3$ . For reactor deployments, lower boron content is preferable to limit negative reactivity due to the high thermal neutron absorption cross section of  $^{10}\text{B}$ . Nextel™ 440, and 610 are good candidates, as they possess a low fraction of  $\text{B}_2\text{O}_3$  (2% for 440, and trace for 610). While these braided sleeve options are intended for high temperature applications, they are also suitable as flexible electrically-resistive covers for the MPFD sheath to electrically isolate it from the reactor structure.

Plastics have also been considered to provide electrical isolation from the reactor. Various plastics have been considered to suit this purpose and are listed in Table 4-4.

Table 4-4. Common plastics considered for use in the reactor environment.

| Material      | Coefficient of Friction | Dielectric Constant | Dielectric Strength (kV/mm) | Tensile Strength (MPa) | Density (g cm <sup>-3</sup> ) | Max Temp (°C) | Gamma-ray Tolerance |
|---------------|-------------------------|---------------------|-----------------------------|------------------------|-------------------------------|---------------|---------------------|
| Nylon         | 0.4                     | 3                   | 20                          | 45                     | 1.13                          | 80            | Fair                |
| Polycarbonate | 0.31                    | 2.8                 | 17                          | 61                     | 1.175                         | 90            | Good                |
| Polyethylene  | 0.2                     | 2.3                 | 17                          | 25                     | 0.955                         | 100           | Fair                |
| PEEK          | 0.25                    | 3.2                 | 24                          | 90                     | 1.275                         | 154           | Excellent           |
| PMMA          | 0.4-0.5                 | 2                   | 15                          | 38                     | 1.185                         | 70            | Good                |
| Polystyrene   | 0.3-0.35                | 2.4                 | 16                          | 35                     | 1.04                          | 65            | Good                |
| PTFE          | 0.05-0.2                | 2.1                 | 17                          | 9                      | 2.15                          | 260           | Good                |

Due to the radiation environment of a nuclear reactor, plastics will degrade in the high gamma-ray and neutron flux. Because of this, plastics are only considered for low/medium fluence operations. However, a benefit of these materials is their low cost and ease of manufacturing. An insulating cover for the MPFD sheath can be easily extruded by manufacturers, or in the case of the upcoming MPFD testing at ATRC (see Section 6.3.2 for further details), plastic standoffs can be machined to provide electrical isolation (shown in Fig. 4-17).

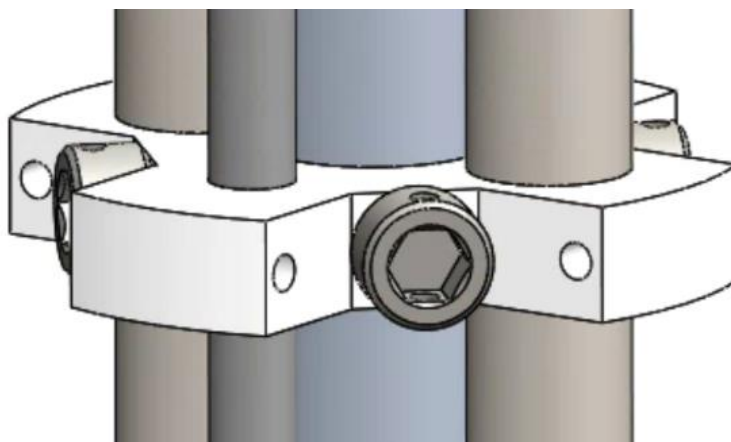


Fig. 4-17. CAD rendering of a plastic standoff used in the upcoming ATRC experiment. This “scaffold” (pictured as white) will be manufactured from PEEK and will isolate the various detectors in the assembly from the reactor structure.

Described in the following chapter are experimental methods used to calibrate MPFDs with activation foils. Both pulse- and current-mode calibration methods are described. Additional information is given on current MPFD processing electronics hardware along with commentary on the updated features.

## 5.1 Calibration Efforts

Over the course of the MPFD research, several attempts have been made to correlate detector response to flux magnitude and the neutron energy spectrum. As previously discussed in Section 3.1.3, detector response has been estimated by simulations of detector deployment in the ATRC core. In contrast, as part of the MPFD deployment at the University of Wisconsin-Madison Nuclear Reactor (UWNR), an experimental process was tested to correlate MPFD response to neutron flux conditions. Prior to MPFD array assembly, a set of enriched uranium (19.6%  $^{235}\text{U}$ ) samples were manufactured, and deposited mass was characterized using alpha spectrometry, with results listed in Table 5-1.

*Table 5-1. Measured uranium mass and mass error for each node of the 7 MPFD arrays.*

|         | Node | mass<br>( $\mu\text{g}$ ) | error<br>( $\mu\text{g}$ ) |         | Node | mass<br>( $\mu\text{g}$ ) | error<br>( $\mu\text{g}$ ) |
|---------|------|---------------------------|----------------------------|---------|------|---------------------------|----------------------------|
| Array 1 | 4    | 0.0555                    | 0.00068                    | Array 5 | 4    | 0.0368                    | 0.00059                    |
|         | 3    | 0.0551                    | 0.00055                    |         | 3    | 0.0360                    | 0.00057                    |
|         | 2    | 0.0544                    | 0.00084                    |         | 2    | 0.0358                    | 0.00055                    |
|         | 1    | 0.0656                    | 0.00057                    |         | 1    | 0.0442                    | 0.00050                    |
| Array 2 | 4    | 0.0856                    | 0.00066                    | Array 6 | 4    | 0.0498                    | 0.00047                    |
|         | 3    | 0.0498                    | 0.00047                    |         | 3    | 0.0418                    | 0.00063                    |
|         | 2    | 0.0558                    | 0.00055                    |         | 2    | 0.0432                    | 0.00074                    |
|         | 1    | 0.0819                    | 0.00102                    |         | 1    | 0.0578                    | 0.00073                    |
| Array 3 | 4    | 0.0597                    | 0.00055                    | Array 7 | 4    | 0.0520                    | 0.00054                    |
|         | 3    | 0.0424                    | 0.00074                    |         | 3    | 0.0285                    | 0.00039                    |
|         | 2    | 0.0433                    | 0.00071                    |         | 2    | 0.0379                    | 0.00060                    |
|         | 1    | 0.0552                    | 0.00084                    |         | 1    | 0.0485                    | 0.00047                    |
| Array 4 | 4    | 0.0449                    | 0.00087                    |         |      |                           |                            |
|         | 3    | 0.0295                    | 0.00044                    |         |      |                           |                            |
|         | 2    | 0.0327                    | 0.00047                    |         |      |                           |                            |
|         | 1    | 0.0446                    | 0.00048                    |         |      |                           |                            |

### 5.1.1 Foil Activation

To unfold the neutron energy spectrum and correlate it to MPFD node response, several foils were used to capture different portions of the neutron energy spectrum. Axial flux characteristics were evaluated as part of this procedure by deploying foils of each material at the approximate axial positions of the MPFD array nodes. The foils used along with general characteristics are given in Table 5-2, and the region of interest for each foils' neutron energy spectrum response is summarized in Fig. 5-1. Foils upon activation were measured using gamma-ray spectroscopy to determine neutron activation levels. Certain foils were paired with cadmium shields to reduce response to thermal neutron energies. All foils were irradiated in the UWNR D67 position at 100 kW<sub>th</sub> with Au, Au(Cd), In, In(Cd), irradiated for 5-min durations, and Mo, Mo(Cd), Al, and Rh irradiated for 30-min durations.

*Table 5-2. Table of various foils and their characteristics used for the UWNR experiment.*

| Isotope            | Nat. Iso. Abund. | Density (g cm <sup>-3</sup> ) | Reaction                  | T <sub>1/2</sub> (h) | Gamma-ray (keV) | Int. (%) | ROI Min (eV)            | ROI Max (eV)           |
|--------------------|------------------|-------------------------------|---------------------------|----------------------|-----------------|----------|-------------------------|------------------------|
| <sup>27</sup> Al   | 100%             | 2.7                           | (n,α) <sup>24</sup> Na    | 15                   | 1368.63         | 99.99    | 6.56 × 10 <sup>6</sup>  | 1.21 × 10 <sup>7</sup> |
| <sup>197</sup> Au  | 100%             | 19.3                          | (n,γ) <sup>198</sup> Au   | 64.8                 | 411.8           | 95.62    | 1.31 × 10 <sup>-2</sup> | 5.46                   |
| <sup>197</sup> Au* |                  |                               |                           |                      |                 |          | 3.99                    | 59.9                   |
| <sup>115</sup> In  | 95.71%           | 7.31                          | (n,γ) <sup>116</sup> In   | 0.9                  | 1293.56         | 84.8     | 1.41 × 10 <sup>-2</sup> | 1.63                   |
| <sup>115</sup> In* |                  |                               |                           |                      |                 |          | 1.22                    | 1.99                   |
| <sup>98</sup> Mo   | 24.13%           | 10.22                         | (n,γ) <sup>99</sup> Mo    | 67.2                 | 739.5           | 12.2     | 2.96 × 10 <sup>-2</sup> | 2.26 × 10 <sup>5</sup> |
| <sup>98</sup> Mo*  |                  |                               |                           |                      |                 |          | 12                      | 3.48 × 10 <sup>5</sup> |
| <sup>103</sup> Rh  | 100%             | 12.4                          | (n,n') <sup>103m</sup> Rh | 0.935                | 39.78           | 0.068    | 4.91 × 10 <sup>5</sup>  | 5.21 × 10 <sup>6</sup> |

\* Indicates use of cadmium filters

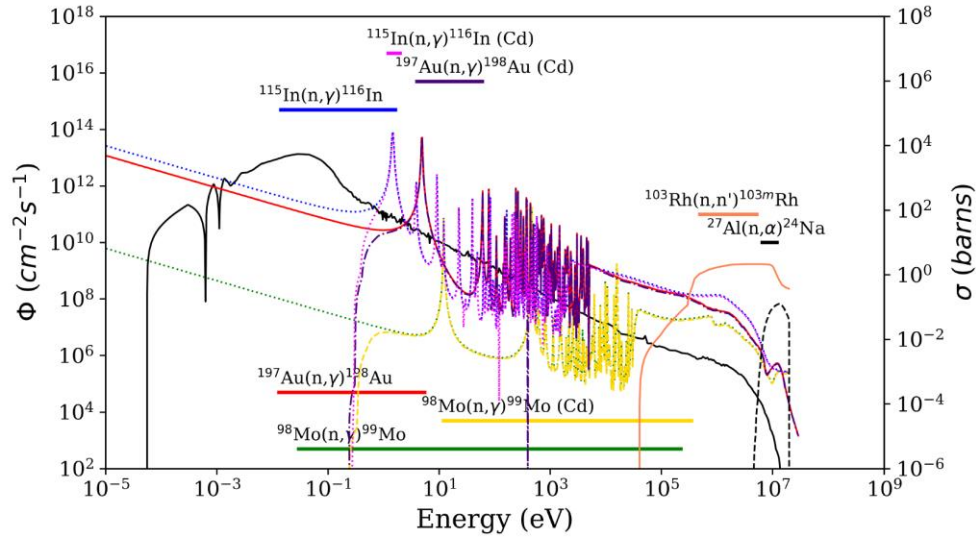


Fig. 5-1. Plot showing various neutron cross sections for the selected activation foils in comparison to the expected neutron energy spectrum for UWNR. Image courtesy of John Boyington.

### Measurement Efforts

Using masses and irradiation durations, the saturation activity was determined for each foil at locations relative to the bottom the standard MPFD array geometry, with saturation activities normalized per atom shown in Fig. 5-2. These activities were then used to unfold the neutron energy spectrum and compared to simulation values shown in Fig. 5-3.

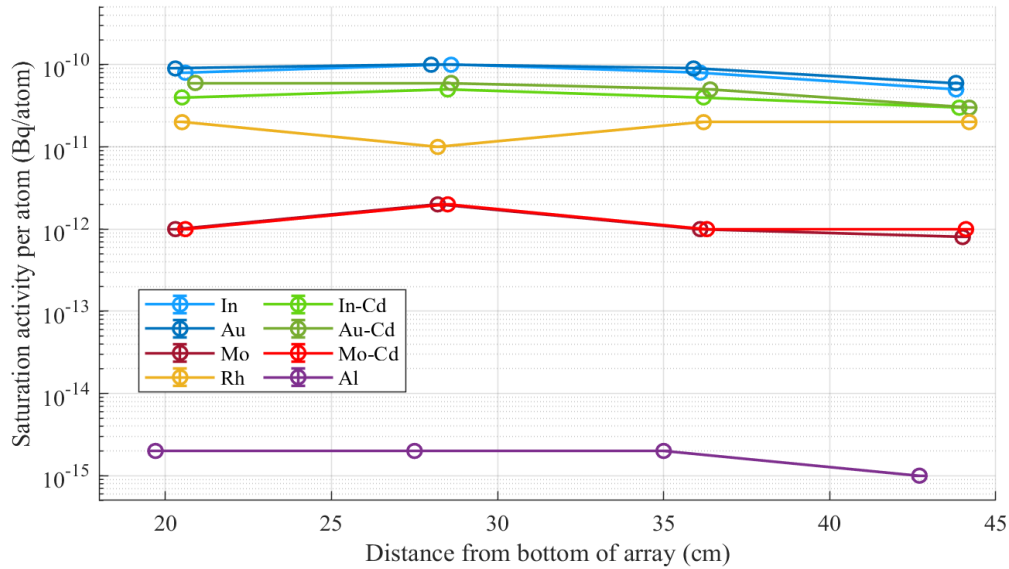


Fig. 5-2. Saturation activities per atom of each foil with indicated location. Note the slight deviation in foil location is due to variability in the assembly process.

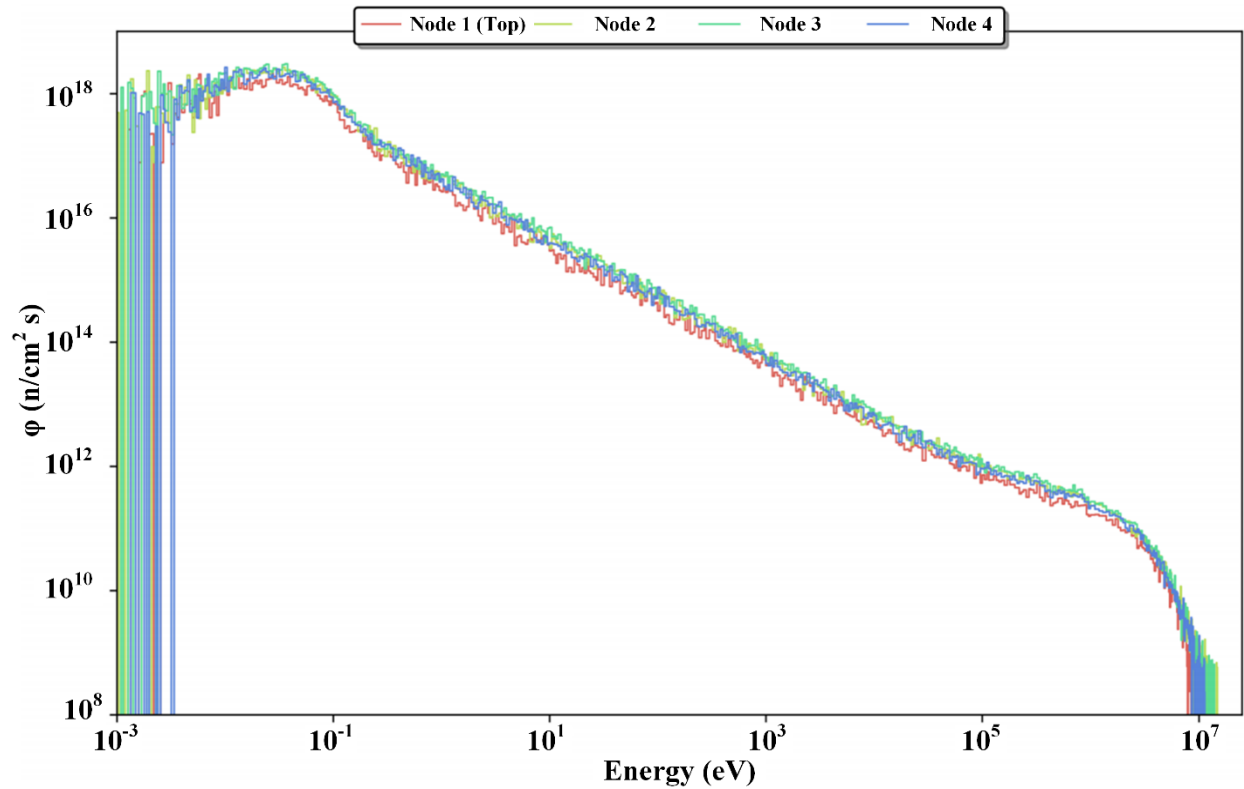


Fig. 5-3. MCNP-simulated differential neutron energy spectrum. Image courtesy of John Boyington.

Comparisons between simulated and measured saturation activity values show varying nodal ratios (shown in Table 5-3). The results show the neutron energy simulations underestimated the high energy neutron flux magnitude and overestimated the low energy neutron flux magnitude.

Table 5-3. Ratio of simulated versus measured saturation activities for each foil and node location (top) and the corresponding neutron energy spans to which each foil is sensitive (bottom).

| Node                                  | Rh                | Al                | Mo(Cd)            | Mo                   | Au                   | Au(Cd)            | In                   | In(Cd)            |
|---------------------------------------|-------------------|-------------------|-------------------|----------------------|----------------------|-------------------|----------------------|-------------------|
| 1                                     | 0.039             | 0.215             | 0.269             | 0.382                | 0.84                 | 1.079             | 2.152                | 3.07              |
| 2                                     | 0.045             | 0.279             | 0.297             | 0.296                | 0.974                | 1.393             | 2.405                | 4.058             |
| 3                                     | 0.098             | 0.227             | 0.251             | 0.278                | 0.963                | 1.259             | 2.12                 | 3.128             |
| 4                                     | 0.052             | 0.201             | 0.27              | 0.274                | 0.915                | 1.218             | 2.183                | 3.282             |
| Neutron Energy Sensitivity Range (eV) |                   |                   |                   |                      |                      |                   |                      |                   |
| Min                                   | $4.9 \times 10^5$ | $6.6 \times 10^6$ | $1.2 \times 10^1$ | $3.0 \times 10^{-2}$ | $1.3 \times 10^{-2}$ | $4.0 \times 10^0$ | $1.4 \times 10^{-2}$ | $1.2 \times 10^0$ |
| Max                                   | $5.2 \times 10^6$ | $1.2 \times 10^7$ | $3.5 \times 10^5$ | $2.3 \times 10^5$    | $5.5 \times 10^0$    | $6.0 \times 10^1$ | $1.6 \times 10^0$    | $2.0 \times 10^0$ |

## 5.2 Pulse-Mode and Current-Mode Calibration

Each of the 7 MPFD arrays were irradiated successively in the D67 position of the UWNR core shown in Fig. 5-4 to perform a dynamic range test (DRT) in both pulse- and current-mode operations and determine the linearity of MPFD node response.

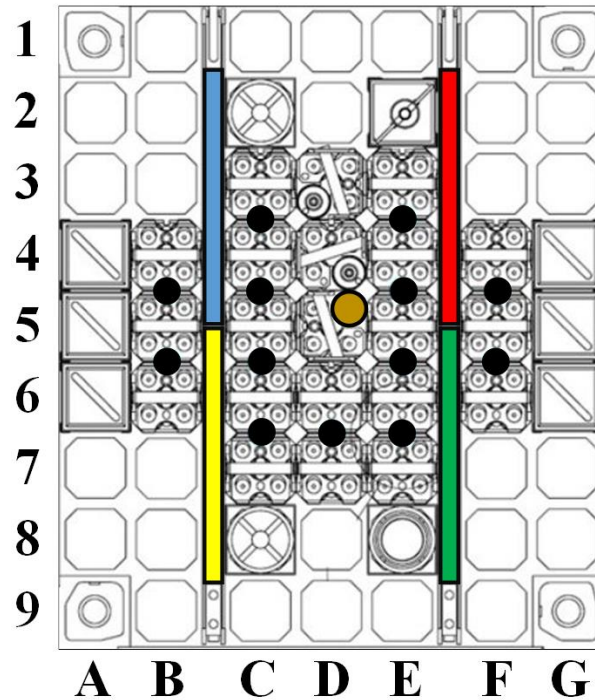


*Fig. 5-4. Map of UWNR core with core locations designated for lateral values (1-9) and longitudinal values (A-G). Various in-core locations are highlighted; MPFD array deployment locations (black dots), control blade 1 (yellow), control blade 2 (blue), control blade 3 (orange), the regulating blade (green), and the transient rod (brown).*

Due to the limitations of the MPFD electronics, a global lower-level discriminator (LLD) was set for all nodes in the array. To determine this LLD, each detector node was examined for the necessary LLD to remove spurious count rates at zero reactor power. The global LLD was then set to a level corresponding to 110% of the maximum LLD of the array nodes. This global LLD method has the unfortunate side-effect of setting high LLDs based on the noisiest node of an array thus penalizing lower noise nodes.

### 5.2.1 Linear Detector Response Confirmation

To quantify the linear response to flux of each MPFD node, a DRT was performed in both pulse and current mode. A DRT consists of operating in steady state at various power levels in

sequence (100, 300, 500, 400, 200 kW<sub>th</sub>) to capture the linear response of each node. At each power level, the count rate in both pulse mode and current mode was recorded to determine linearity as shown in Fig. 5-5 and Fig. 5-6.

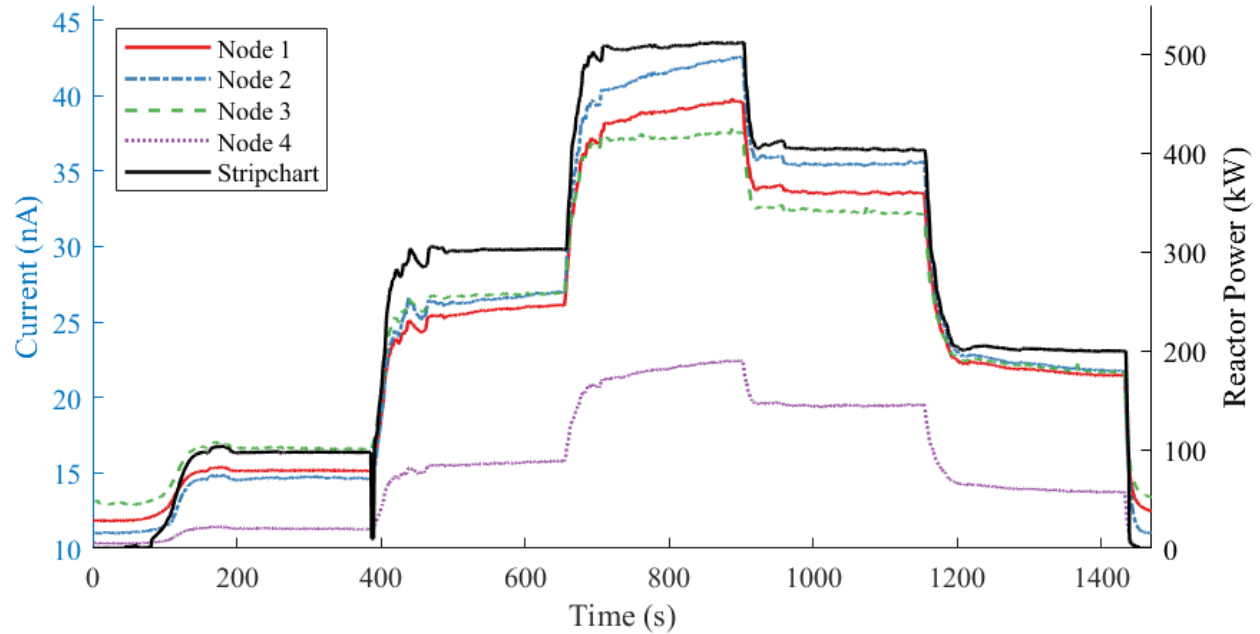


Fig. 5-5. Plot of DRT for MPFD array 3 performed in current mode. Note that raw count rate is presented for MPFD response.

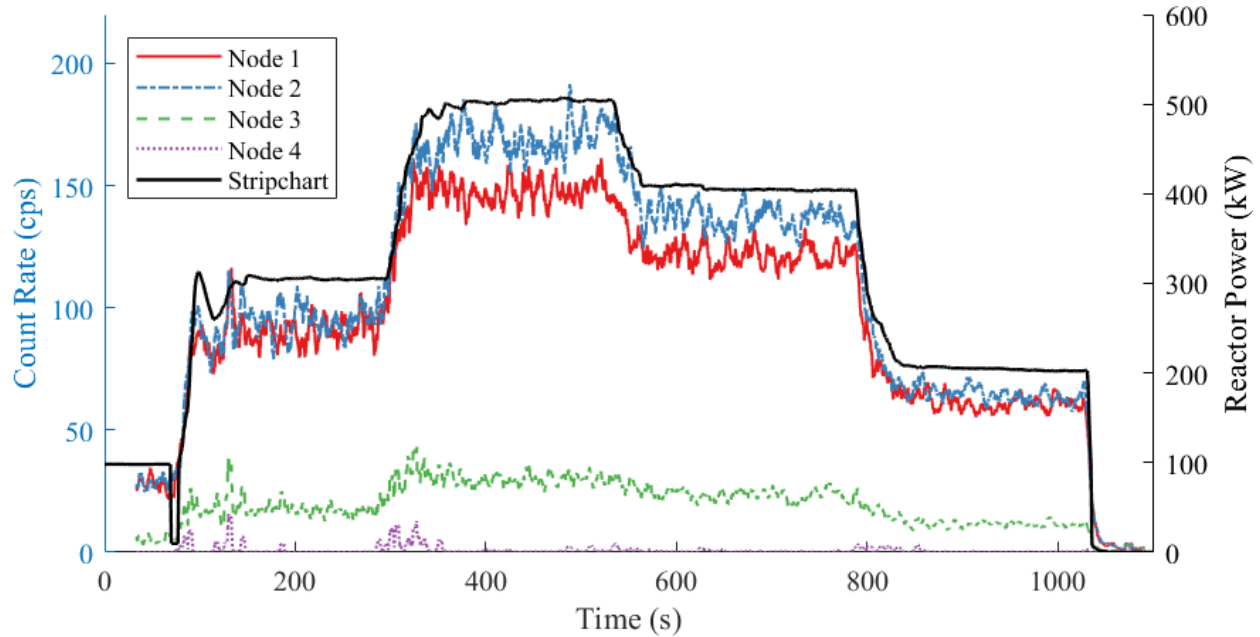


Fig. 5-6. Plot of DRT for MPFD array 3 performed in pulse mode. Note that MPFD data presented is smoothed with a moving average window of 200 data points.

Comparing the results shown in Fig. 5-5 and Fig. 5-6, it was observed that pulse-mode operation suffers from significant noise intrusion. It was also observed that in pulse-mode operations, the global LLD was significant enough to discriminate most pulses registered in node 4, highlighting the key drawback discussed previously. In current mode operations, as seen in Fig. 5-5, the detector current gradually increases when transitioning to a higher reactor power level. The inverse effect is observed as reactor power decreases and the registered count rate fades to a low level as time continues. This phenomenon has several explanations that will be elaborated upon in Section 6.2.2.

Due to the higher noise of pulse-mode operation, array 3 was set to current-mode operations for the experiment and will be discussed further in Section 6.2.1. Linearity of array 3 was determined by using count rates obtained during current-mode operation and is shown in Fig. 5-7. From the trendline equations listed, it was observed that each node had similar zero-power current between 10 – 13 nA. For comparison, pulse mode linearity is shown in Fig. 5-8. This process was performed for each of the MPFD arrays are provided in Appendix E.

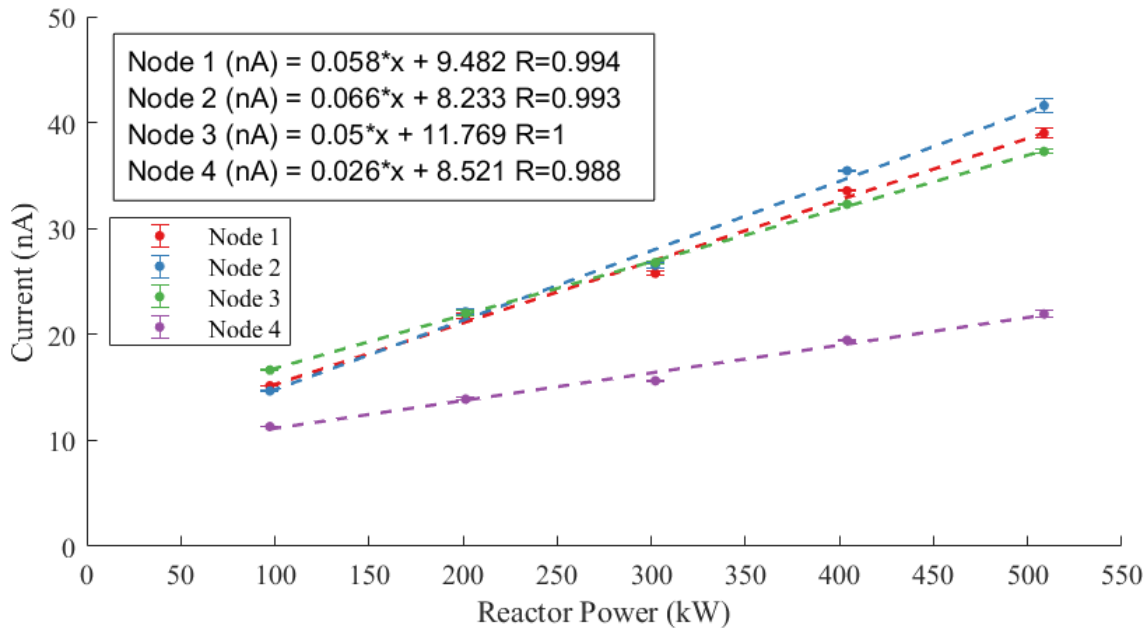


Fig. 5-7. Current mode linearity response of MPFD array 3 based on data presented in Fig. 5-5. Linear least-square equations are shown for each node.

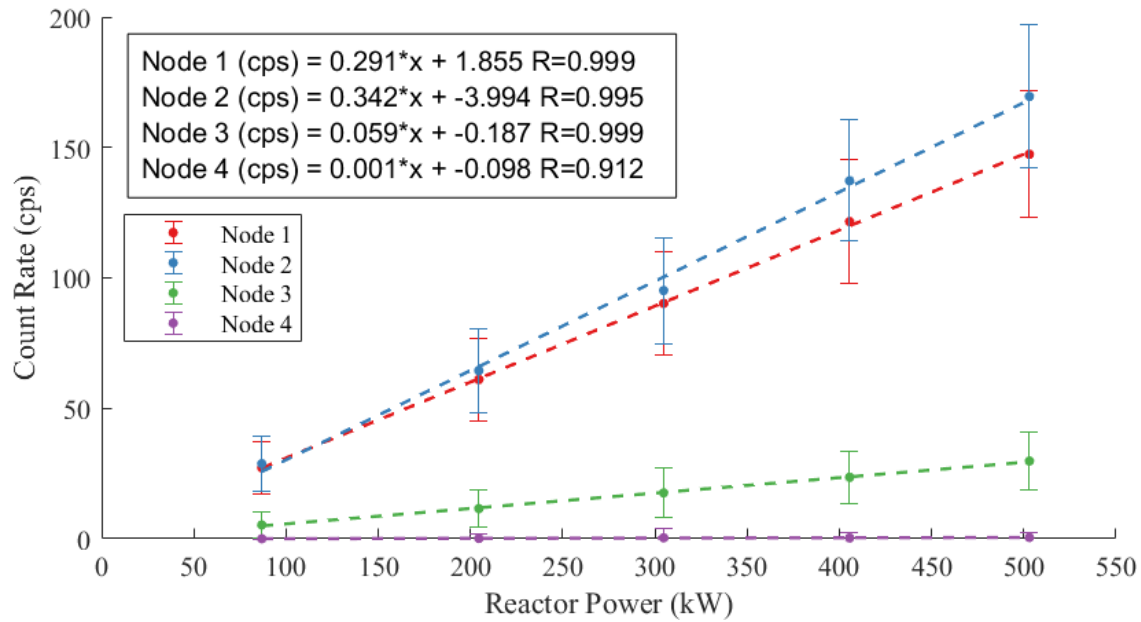


Fig. 5-8. Pulse mode linearity response of MPFD array 3 based on data presented in Fig. 5-6. Linear least-square equations are shown for each node.

### 5.2.2 Estimated MPFD sensitivity

As part of the effort to calibrate MPFD response to flux, the specific saturation activities of the gold foils, shown in Fig. 5-2 were used to estimate axial neutron flux magnitudes and are shown in Table 5-4. Array 3 was chosen to determine the detector sensitivity and efficiency due to large signal to noise ratio in current mode, and moderate response in pulse mode.

Table 5-4. Gold foil masses and axial position of gold foils with calculated specific saturation activities and the estimated thermal flux in the D67 calibration position.

| Nodes      | Distance from center plane (cm) | Foil Mass (mg) | Specific Saturation Activity (Bq atom <sup>-1</sup> ) | Thermal Flux* (n cm <sup>-2</sup> s <sup>-1</sup> ) |
|------------|---------------------------------|----------------|-------------------------------------------------------|-----------------------------------------------------|
| (top) 4    | 11.6                            | 4.2            | $(6 \pm 0.2) \times 10^{-11}$                         | $(5.0 \pm 0.17) \times 10^{11}$                     |
| 3          | 3.7                             | 4.3            | $(9 \pm 0.3) \times 10^{-11}$                         | $(7.5 \pm 0.25) \times 10^{11}$                     |
| 2          | -4.2                            | 4.1            | $(10 \pm 0.4) \times 10^{-11}$                        | $(8.3 \pm 0.33) \times 10^{11}$                     |
| (bottom) 1 | -11.9                           | 4.1            | $(9 \pm 0.3) \times 10^{-11}$                         | $(7.5 \pm 0.25) \times 10^{11}$                     |

\* D67 position at 100 kW<sub>th</sub>

Using the estimated thermal neutron flux coupled with the mass of each node shown in Table 5-1, theoretical fission rates were determined for each of the array 3 nodes to be  $2.09 \times 10^{-8}$ ,  $1.49 \times 10^{-8}$ ,  $1.52 \times 10^{-8}$ , and  $1.94 \times 10^{-8}$  cps/nv for nodes 1, 2, 3, and 4 respectively. The

theoretical current generated in the MPFD detector was also determined using the average energy deposition for fission fragments (14.25 MeV) found in Table 3-3, which produces approximately  $5.48 \times 10^5$  electron-ion, (0.175 pC), pairs per fission event. Assuming each fission event will result in a fission fragment entering the chamber and depositing 0.175 pC of charge and factoring in the fission rate for each node of array 3, the theoretical current per unit flux was determined to be  $3.66 \times 10^{-20}$ ,  $2.61 \times 10^{-20}$ ,  $2.66 \times 10^{-20}$ , and  $3.40 \times 10^{-20}$  A/nv for nodes 1, 2, 3, and 4 respectively. The theoretical MPFD sensitivities for both pulse mode and current mode were then compared with the experimental calibration results of MPFD array 3 shown in Fig. 5-5 and Fig. 5-6. Using the slope of the linear least square regression fit to the experimental data, an estimate of detector efficiency was determined for each node and is presented in Table 5-5. It observed that in current mode operation, all MPFD nodes measured less than 30% of the expected current. This may be the result of charge carriers, mainly electrons, laterally diffusing into the chamber wall reducing the overall observed current, like the effect described in Section 3.2.2. Additionally, as mentioned in Section 3.1.2, the detection efficiency is reduced due to gamma-ray energy deposition accounting for approx. 16.7% of the total energy deposited. For pulse mode operations, it is observed that the detector efficiency of all nodes was less than 0.276% for all the nodes. This may be primarily a result of the global LLD setting used to filter out noise. The global LLD was set to such a high level, that most pulses produced in node 4 were discriminated from counting as evidenced in Fig. 5-6.

*Table 5-5. Slope of linear least square regression fit to the experimental data shown in Fig. 5-5 and Fig. 5-6. Detector efficiency was then determined by comparing experimental values against theoretical estimations.*

| Node                                | 1                                 | 2                                 | 3                                 | 4                                  |
|-------------------------------------|-----------------------------------|-----------------------------------|-----------------------------------|------------------------------------|
| PM slope<br>(cps/kW <sub>th</sub> ) | 0.2912                            | 0.3424                            | 0.059                             | 0.0012                             |
| PM sens.<br>(cps/nv)                | $(38.8 \pm 1.3) \times 10^{-12}$  | $(41.3 \pm 1.64) \times 10^{-12}$ | $(7.87 \pm 0.26) \times 10^{-12}$ | $(0.24 \pm 0.001) \times 10^{-12}$ |
| Det. Eff.                           | 0.185%                            | 0.276%                            | 0.052%                            | 0.001%                             |
| CM slope<br>(nA/kW <sub>th</sub> )  | 0.058                             | 0.066                             | 0.05                              | 0.026                              |
| CM sens.<br>(A/nv)                  | $(7.73 \pm 0.26) \times 10^{-21}$ | $(7.95 \pm 0.32) \times 10^{-21}$ | $(6.67 \pm 0.22) \times 10^{-21}$ | $(5.20 \pm 0.18) \times 10^{-21}$  |
| Det. Eff.*                          | 17.7%                             | 25.5%                             | 21.1%                             | 12.9%                              |

\* Value is compensated for gamma-ray energy deposition

## 5.3 MPFD Electronics Hardware

During each MPFD experimental test, lessons were learned pertaining to the operation of the detectors coupled with the support electronics. Various generations of MPFD electronics have been employed over the course of research which have facilitated detector operation and data collection. Most notably among the previous tests, the MPFD deployment of the UWNR facility highlighted concerns about cross-mode use (switching between pulse- and current-mode) as well as signal diagnostics and application of proper electrical grounding. As mentioned previously, concern about setting operation parameters for each node individually arose. A new generation of physical hardware (MPFD-4 v2.0) was proposed and is presently being introduced into the MPFD platform. All research presented herein was conducted using the MPFD-4 v1.0 electronics which is presented alongside the MPFD-4 v2.0 electronics for comparison.

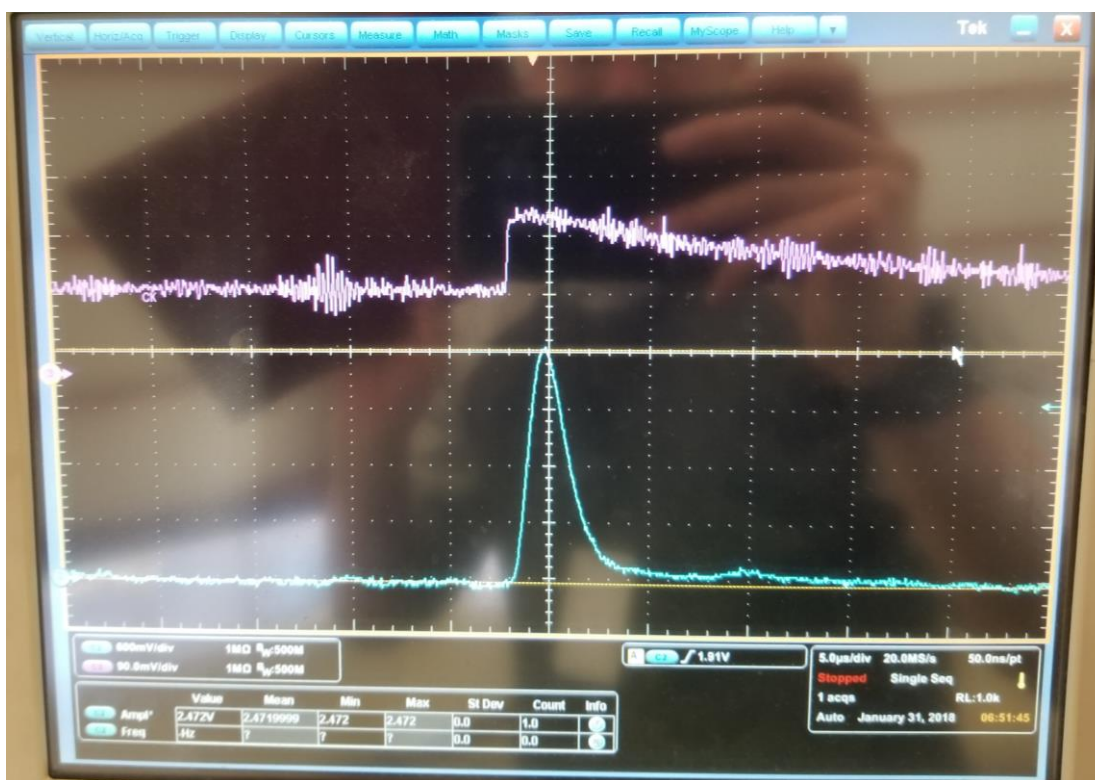


*Fig. 5-9. Picture of pulse signatures captured on the oscilloscope during in-core testing showing a common neutron pulse signal from the preamplifier (top) and shaping amplifier (bottom). The signals were collected with MPFD-4 v1.0 electronics.*

### 5.3.1 Physical Hardware and Capabilities

Both the previous and current generation of MPFD electronics can accommodate a minimum of 4 MPFD nodes. The standard MPFD-4 v1.0 setup can handle 4 nodes using the standard preamplifier as shown in Fig. 5-10, but can accommodate 8 nodes using a specialized MPFD-8 preamplifier, but requires two NIM bin module to amplify, shape and discriminate the signals. The MPFD-4 v2.0 electronics were designed to handle up to 8 nodes by utilizing two frontend modules coupled with a single receiver box, shown in Fig. 5-10. Electronics diagrams for each generation are shown in Fig. 5-11 and Fig. 5-12 depicting the internal signal processing chain and component coupling. For comparison, the operating characteristics of each of the electronics generations is provided in Table 5-6. It is important to note, that the v1.0 electronics required separate pulse- and current-mode preamplifiers for the two operating modes, whereas v2.0 has dual-mode capability integrated into a single module. The v1.0 electronics utilizes a single D-sub9 connector through which each node must be connected, whereas the v2.0 electronics utilize 4 independent Lemo 0B.302 connectors allowing coupling of a combination of single node detectors, or multimode detectors without the need for a single connector integration, allowing for more flexibility. Both generations can be controlled using the NIM bin module (v1.0) or the receiver box (v2.0), or through a software user interface via USB-A connection.

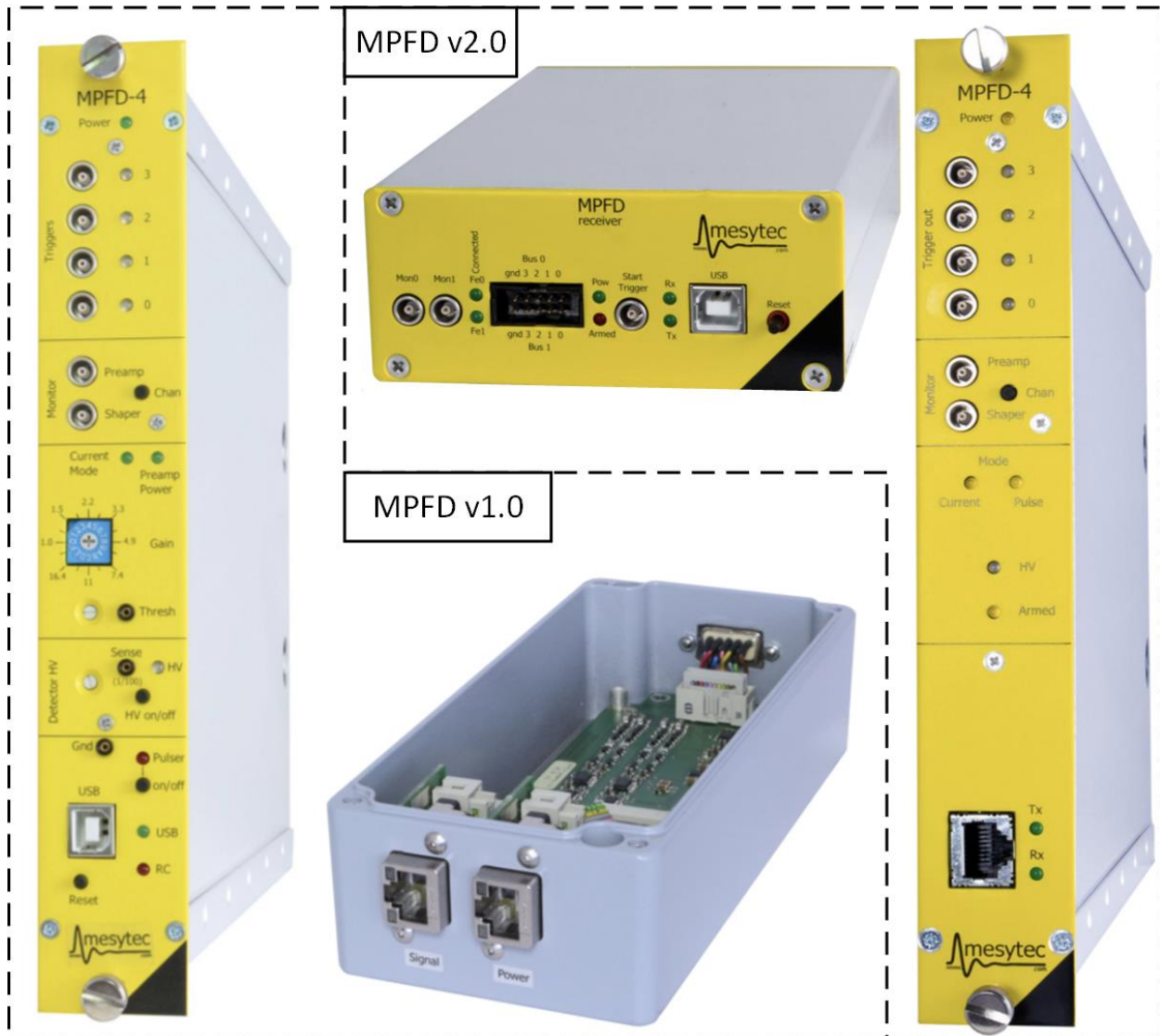


Fig. 5-10. Picture of the MPFD v1.0 electronics (left) which includes the NIM bin module (left), and the water-tight preamplifier box compared to the MPFD v2.0 electronics (right) which includes the frontend NIM bin module (left) and the receiver box (right).

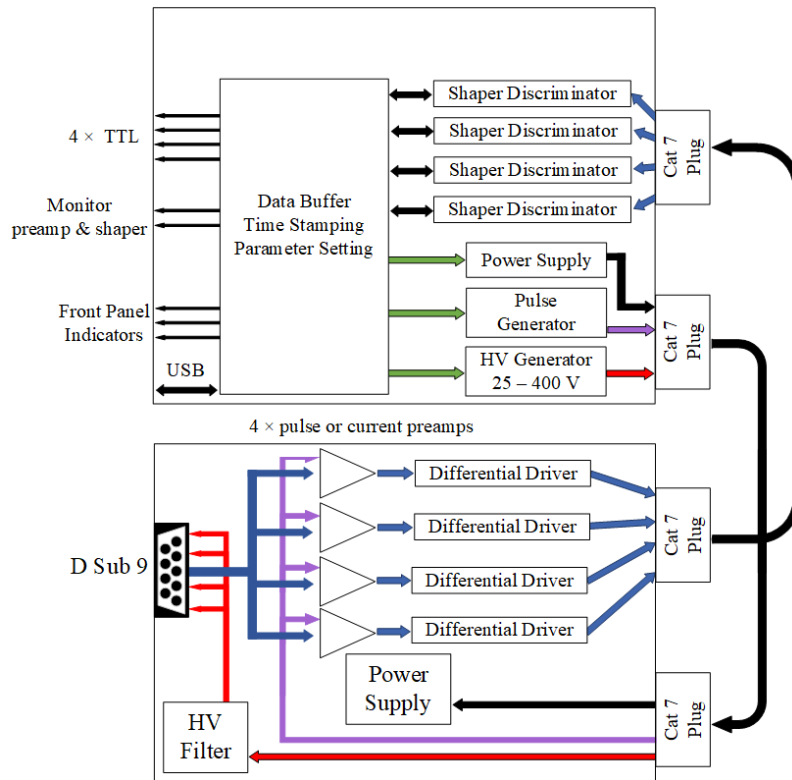


Fig. 5-11. MPFD-4 v1.0 electronics diagram showing preamplifier box (bottom), and NIM bin module (top) with internal circuit components.

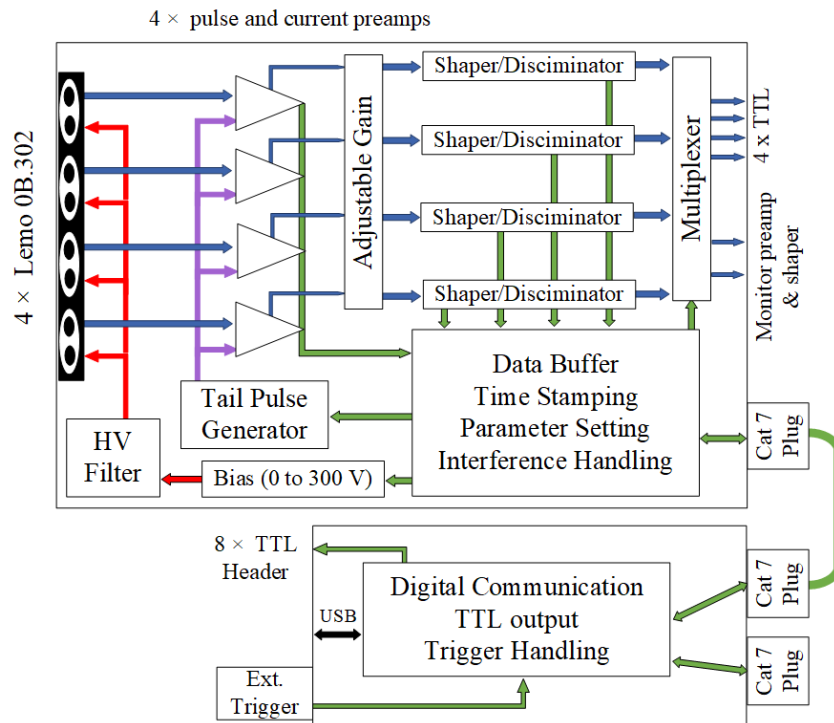


Fig. 5-12. MPFD-4 v2.0 electronics diagram showing the frontend NIM bin compatible electronics (top) and the receiver box (bottom) which can accommodate two frontend modules.

Table 5-6. Operating characteristics of the previous (MPFD v1.0) and current generation (MPFD v2.0) electronics

|                         |                          | MPFD v1.0                           | MPFD v2.0         |
|-------------------------|--------------------------|-------------------------------------|-------------------|
| General Characteristics | Max detector capacitance | 1 nF                                |                   |
|                         | Voltage bias             | Common for all channels             |                   |
| Pulse mode              | Max pulse rate           | N/A                                 | 100 cps at 500 ns |
|                         | Voltage range            | 25 – 400 V                          | 25 – 300 V        |
|                         | Max current              | 100 $\mu$ A per channel             |                   |
|                         | Shaping times            | 0.25, 0.5, 1.0 or 2 $\mu$ s         |                   |
|                         | Discriminator threshold  | Common                              | Independent       |
|                         | Discrimination type      | Leading Edge                        |                   |
| Current mode            | Current conversion       | 100 cps $\text{nA}^{-1}$            |                   |
|                         | Max current              | 100 $\mu$ A                         |                   |
|                         | Voltage range            | 12.5 – 200 V                        | 25 – 300 V        |
| Capture Mode            | Sampling interval        | 0.25, 0.5, 1.0 or 2.0 ms            |                   |
|                         | Buffer length            | 1024 pre-trigger, 3072 post-trigger |                   |

This chapter focuses on MPFD experimentation campaigns conducted at various research reactors. Among the reactors discussed are tests at the KSU TRIGA Mark II Nuclear Research Reactor involving reactor pulsing. Testing at UWNR included calibration efforts, flux tilting, steady state, and transient operations (square wave, ramp, and reactor pulsing). Finally, tests conducted at INL in the TREAT facility and an upcoming deployment at ATRC will be discussed.

## 6.1 Kansas State University Testing

The MPFD research has been primarily conducted at KSU TRIGA Mark II Nuclear Research Reactor, with detector fabrication conducted in the Semiconductor Materials and Radiological Technologies (SMART) Laboratory. In preparation for the large scale MPFD deployment at UWNR, several testing regiments were conducted at KSU to confirm MPFD design parameters and electronics operations.

### 6.1.1 Transient Testing

The KSU TRIGA Mark II Nuclear Research Reactor has the unique ability to perform a rapid power excursion, increasing in power level by a factor of  $10^8$  within 20 ms, known as a reactor pulse. To achieve this result, the reactor achieves a supercritical state by ejecting the transient control rod from the core. As the reactor quickly increases in power, the fuel begins to rapidly increase in temperature. The fuel heating causes hydrogen in the TRIGA fuel matrix to begin oscillating, giving the fuel a large negative temperature feedback coefficient of reactivity. The reactor power level will then rapidly reduce due to the lowered fission rate in the fuel. This process is a result of the unique composition of TRIGA fuel which utilizes a uranium zirconium hydride fuel. Reactor pulse transients can be accurately modeled using the Fuchs-Nordheim model as described in Eq. (13) with Eq. (14) to determine the FWHM of the pulse:

$$P(t) = \frac{\beta^2 C_p}{2\alpha l'} \left[ \frac{\rho_0}{\beta} - 1 \right]^2 \text{sech}^2 \left( \frac{t(\rho_0 - \beta)}{2l'} \right), \quad (13)$$

$$t = \frac{\cosh^{-1}(\sqrt{2})}{\left[ \frac{\rho_0 - \beta}{2l'} \right]}, \quad (14)$$

$C_p$  = TRIGA fuel heat capacity ( $6.648 \times 10^{-4} \text{ J K}^{-1}$ )       $\beta$  = TRIGA delayed neutron fraction (0.007)  
 $\alpha$  = TRIGA fuel temperature coefficient ( $8 \times 10^{-5} \Delta k \text{ K}^{-1}$ )       $l'$  = prompt neutron lifetime ( $4.3 \times 10^{-5} \text{ s}$ )

The Fuchs-Nordheim model relies on key assumptions to predict the reactor transient. First, the reactor is assumed to have adiabatic heat transfer during the duration of the pulse. Second, it is assumed that no delayed neutrons will contribute to the transient due to the short time duration (<100 ms). Third, the fuel elements are assumed to have constant heat capacity over the duration of the transient. Fourth, the reactivity insertion is assumed to be immediate, and imitate a step insertion. In practicality, due to the method of transient rod ejection, the reactivity insertion is non-linear, with a finite but changing slope.

Because of the short duration and rapid change in neutron population, reactor pulses represent the most extreme transients that a neutron monitoring sensor can experience. Because of these characteristics, reactor pulsing is used as a benchmark of the detector's performance and capabilities. While MPFDs have been deployed during reactor pulsing experiments previously, results were inconclusive as to whether or not the MPFDs accurately tracked the pulse [70], [85]. To characterize the MPFD accuracy, a 4-node array was built to represent the design for use in the upcoming UWNR tests. The array was fabricated with node masses and displacements listed in Table 6-1 and located in the Intra-Reflector Irradiation System (IRIS) shown in Fig. 6-1. As part of the IRIS, there are two fuel element storage positions, with the MPFD array being placed in the eastern storage location.

*Table 6-1. Summary of node masses and displacement from core centerline.*

| Node #     | Mass (ng)      | Axially displacement from fuel center plane(cm) |
|------------|----------------|-------------------------------------------------|
| 3 (top)    | $66.3 \pm 1.6$ | 21.3                                            |
| 2          | $65.2 \pm 1.5$ | 29.0                                            |
| 1          | $65.9 \pm 1.5$ | 36.6                                            |
| 0 (bottom) | $62.2 \pm 1.5$ | 44.2                                            |

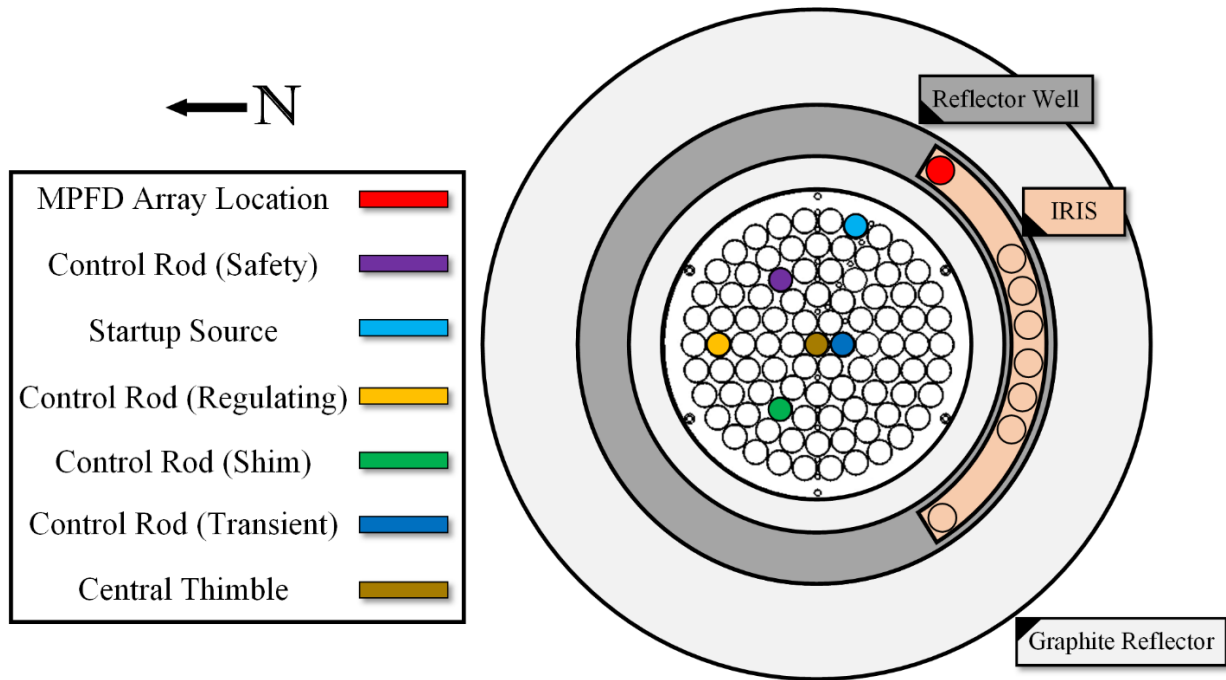


*Fig. 6-1. Map of the KSU TRIGA Mark II Nuclear Research Reactor core.*

These pulse tests were exploratory for the new MPFD design and concerns arose that the mass of each chamber would not be sufficient to trigger the MPFD electronics. Therefore, the summed signal of all four chambers was used to track the transient. The detectors were operated in current-mode and two pulses were conducted with a \$2.77 and \$2.50 reactivity insertion. Data collected from the MPFD was compiled and compared against the Fuchs-Nordheim model for both reactor pulses. As shown in Fig. 6-2 and Fig. 6-3, the MPFD array was able to track these two power excursions of \$2.50 and \$2.77 with measured FWHMs deviating 11.46% and 8.61% from the theoretical values. The theoretical and experimental results are listed in Table 6-2.

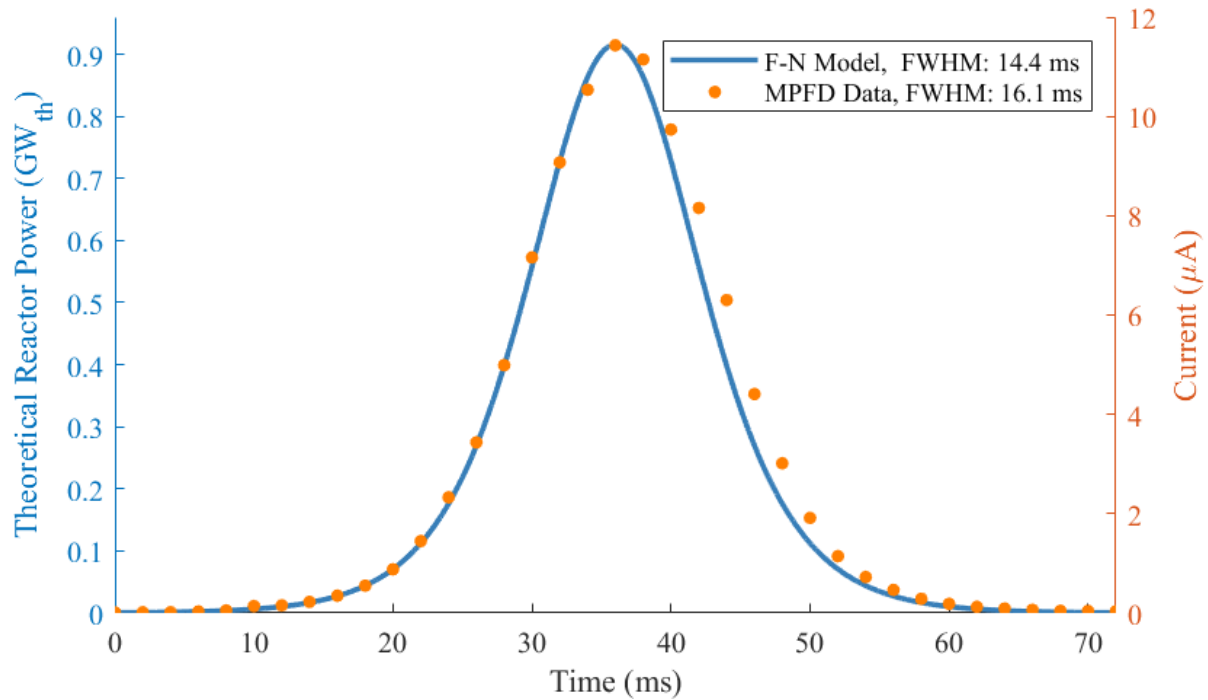


Fig. 6-2. MPFD pulse data compared to scaled Fuchs-Nordheim Model for \$2.50 reactivity pulse. MPFD data sampling rate was set to 2 ms.

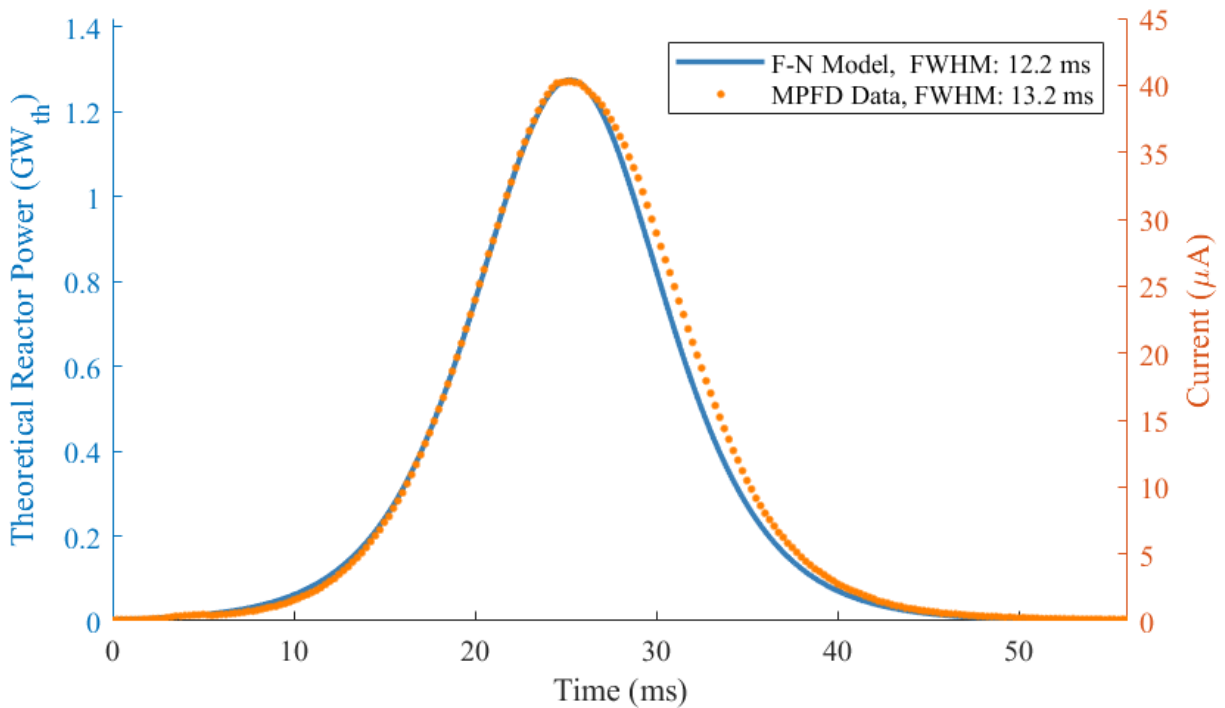


Fig. 6-3. MPFD pulse data compared to scaled Fuchs-Nordheim Model for \$2.77 reactivity pulse. MPFD data sampling rate was set to 0.25 ms.

Table 6-2. Model results vs experimental data and detector operation parameters.

| Fuchs-Nordheim Model |                               |           | MPFD Operation   |          |                  |      |
|----------------------|-------------------------------|-----------|------------------|----------|------------------|------|
| Reactivity           | Max Power (GW <sub>th</sub> ) | FWHM (ms) | Max current (μA) | Bias (V) | Sample Rate (ms) |      |
| \$2.50               | 0.916                         | 14.4      | 16.05            | 11.43    | 100              | 2.00 |
| \$2.77               | 1.27                          | 12.2      | 13.25            | 40.28    | 150              | 0.25 |

During these reactor pulse tests, an interesting anomaly was observed in both datasets. Preceding the main detector response, a low amplitude peak was observed during the initial rise in detector response. Seen in Fig. 6-4, this small elevation in counts occurs in both datasets, but is most prominent during the \$2.77 pulse due to the higher resolution sampling interval of 0.25 ms. This temporarily elevated response was calculated to precede the peak response by 26 ms and 21.25 ms for the \$2.50 and \$2.77 pulse, respectively. At present time, there is no explanation to this phenomena, and additional testing will be required in the future to better identify this anomalous result.

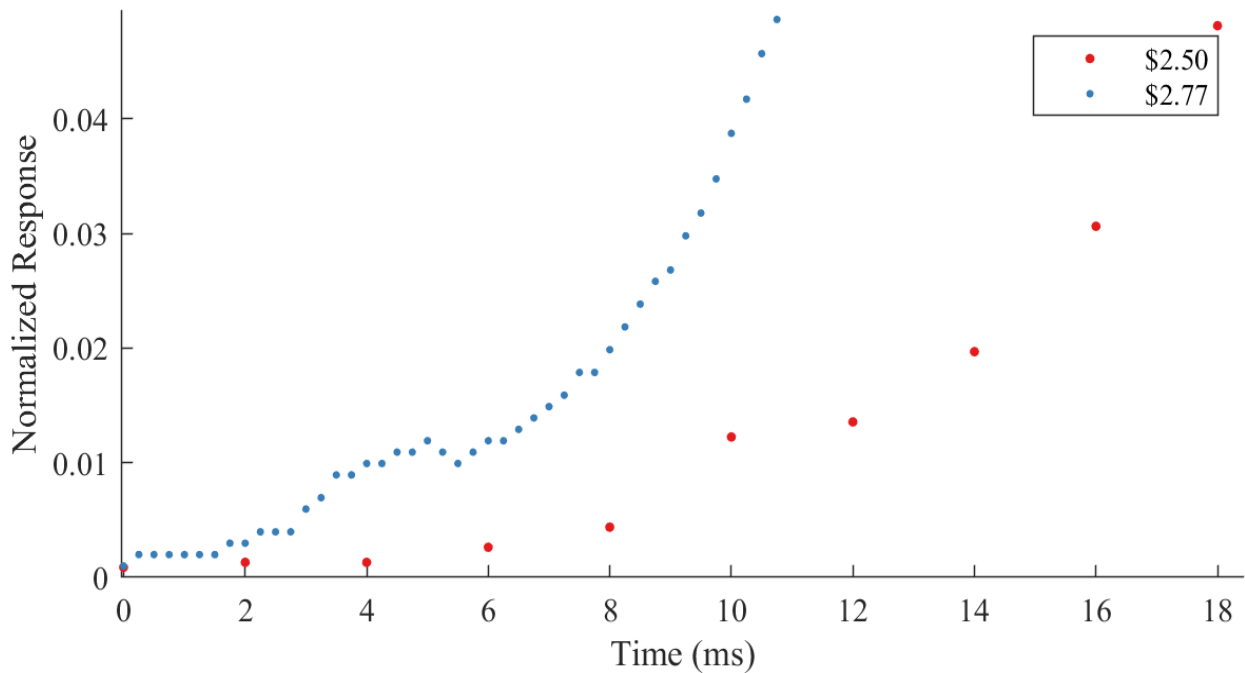


Fig. 6-4. Plot of normalize detector response for both reactor pulses showing a signal perturbation on the leading edge of the pulse.

### **6.1.2 Summary**

The data gathered at KSU provided valuable insight into the performance of the newly manufactured common-cathode design. These tests also constituted the first time MPFDs had been used to accurately track a reactor pulse and demonstrated the MPFD's potential for fast transient tracking. The MPFD response deviated from Fuchs-Nordheim model by a slight margin. This effect may be the result of a few causes. First, the Fuchs-Nordheim model makes several key assumptions that are approximations of practical conditions, most notably step reactivity insertion due to ejection of the transient control rod. Second, the MPFD nodes were placed inside the reflector well of the reactor, which may cause a modification in the thermal neutron flux due to the proximity of the graphite reflector. Finally, due to experimenter concerns, the response of all 4 nodes were interlinked to improve signal magnitude during the test. Because nodes spanned a total axial distance of 9 inches, it may be possible that the pulse width was broadened slightly by the node displacement due to the delay in neutron flux propagation to each node. From these tests, experimenters determined the proper procedure for operating MPFD during reactor pulsing or fast transient testing. Additionally, these tests allowed experimenters to better prepare for the deployment at UWNR.

## **6.2 University of Wisconsin Madison Testing**

Testing at UWNR was conducted as part of a Nuclear Energy University Program (NEUP) grant with the goal of three-dimensionally mapping the core for neutron flux and temperature during steady state and transient operations. The results from these tests were used to validate core modeling codes as part of the NEAMS program. A set of 7 MPFD arrays were manufactured, each with 4 nodes, used for neutron flux measurements and 3 RTD probes, each with 6 nodes, were used for temperature measurements. MPFDs were manufactured using the common-cathode geometry and were designed to have 4 nodes evenly spaced to span the fuel region of the core. The node spacing was designed to be a mirror configuration with respect to the fuel region center plane. For steady state operations, dynamic range testing (DRT) at reactor power levels in sequence consisting of 100, 300, 500, 400 and 200 kW<sub>th</sub> were conducted. Additionally, flux shaping measurements were conducted at 100 kW<sub>th</sub>, with 5 unique control blade configurations to induce flux tilting. For transient operations, reactivity insertion tests were conducted for power

ramping at 50, 30 and 20-seconds periods, as well as square wave tests up to 250, 500, and 1000 kW<sub>th</sub>, and finally three reactor pulses with \$1.43, \$1.71 and \$1.97 reactivity insertions were conducted. Among these tests, several were repeated with MPFDs and RTDs located at alternate deployment positions in the core as shown in Fig. 6-5.

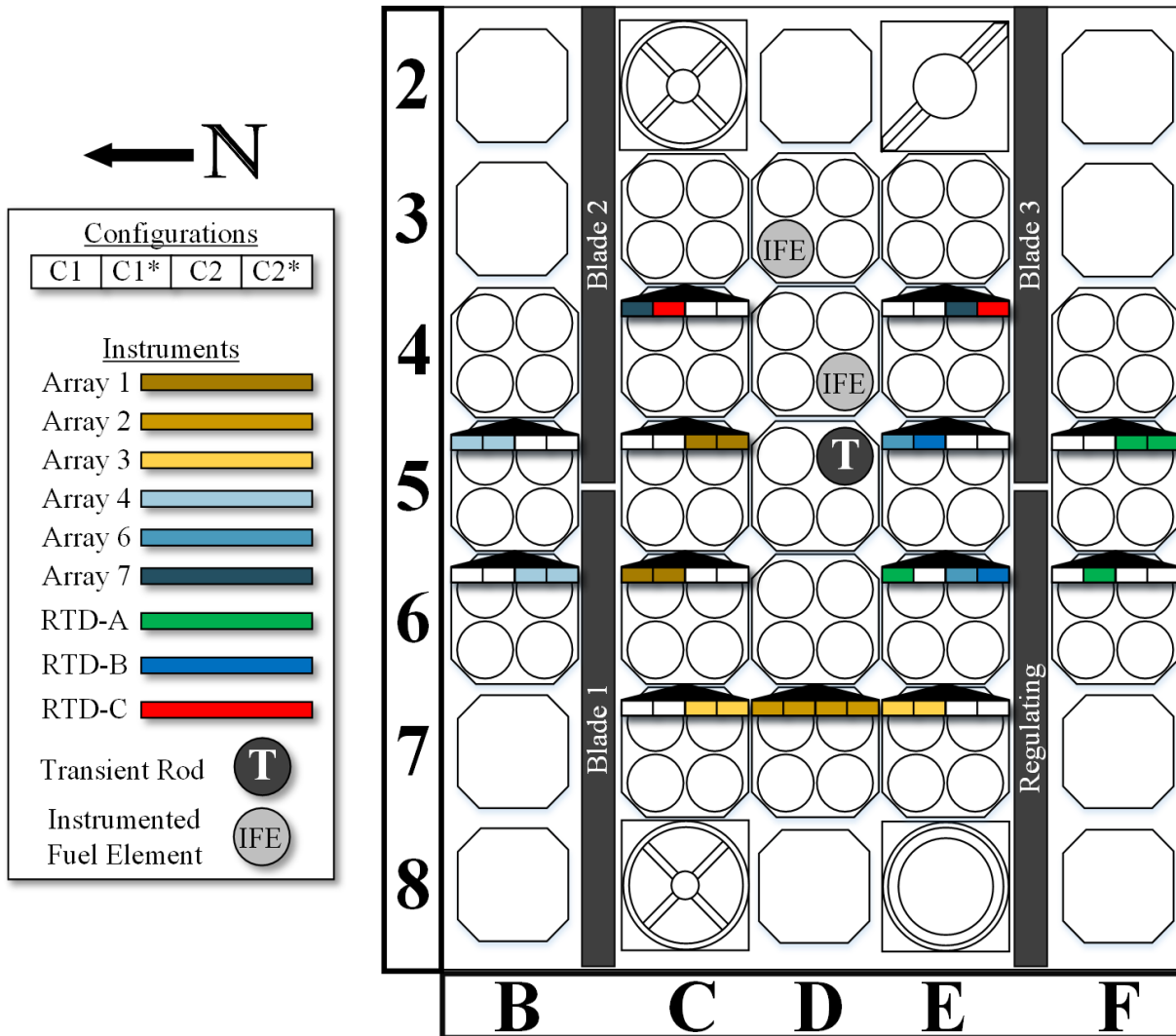


Fig. 6-5. Map of UWNR core with experiment instrument locations during four unique configurations and permanent reactor components such as control blades, instrumented fuel elements (IFE) and the transient rod.

The entire experiment campaign spanned 8 days of testing. The first 5 days were utilized to calibrate each of the MPFD arrays and RTD probes. Steady state and transient tests were conducted over the course of 3 days following calibration. A summary of the entire test campaign is given in Table 6-3.

Table 6-3. Summary of calibrations and experimental tests performed at UWNR.

| Day 1 (7/23)       |                      |                        |                      |
|--------------------|----------------------|------------------------|----------------------|
| Function Test      |                      |                        |                      |
| Day 2 (7/24)       |                      |                        |                      |
| W1 <sup>Δ</sup>    | Mo+Cd <sup>ΔΔ</sup>  | Mo <sup>ΔΔ</sup>       | Al <sup>ΔΔ</sup>     |
| Day 3 (7/25)       |                      |                        |                      |
| W2 <sup>Δ</sup>    | W3 <sup>Δ</sup>      | W4 <sup>Δ</sup>        |                      |
| Day 4 (7/26)       |                      |                        |                      |
| Au <sup>ΔΔΔ</sup>  | Au+Cd <sup>ΔΔΔ</sup> | In <sup>ΔΔΔ</sup>      | In+Cd <sup>ΔΔΔ</sup> |
| Rh <sup>ΔΔ</sup>   | W5 <sup>Δ</sup>      | W6 <sup>Δ</sup>        |                      |
| Day 5 (7/27)       |                      |                        |                      |
| W1 <sup>ΔΔΔΔ</sup> | W7 <sup>Δ</sup>      | RTD-A                  | RTD-B                |
| RTD-C              |                      |                        |                      |
| ↑ Calibrations ↑   |                      | ↓ Experimental Tests ↓ |                      |
| Day 6 (7/30)       |                      |                        |                      |
| A                  | C1 (DRT)             | B                      | C1 (all Blade FT)    |
| Day 7 (7/31)       |                      |                        |                      |
| C                  | C1 (50-s ramp)       | D                      | C1 (30-s ramp)       |
| E                  | C1 (20-s ramp)       | F                      | C1* (DRT)            |
| G                  | C1* (Blade 1 FT)     | H                      | C1* (50-s ramp)      |
| J                  | C2* (DRT)            | K                      | C2* (Blade 2 FT)     |
| L                  | C2* (30-s ramp)      | M                      | C2* (500kW SW)       |
| Day 8 (8/1)        |                      |                        |                      |
| N                  | C2 (DRT)             | O                      | C2 (Blade 3 FT)      |
| P                  | C2 (20-s ramp)       | Q                      | C2 (250kW SW)        |
| R                  | C2 (500kW SW)        | S                      | C2 (1MW SW)          |
| T                  | C2 \$1.43 Pulse      | U                      | C2 \$1.71 Pulse      |
| V                  | C2 \$1.97 Pulse      | W                      | C1 \$1.43 Pulse      |
| X                  | C1 \$1.71 Pulse      | Y                      | C1 \$1.97 Pulse      |

DRT = Dynamic Range Test  
(100,300,500,400,200 kW)

SW = Square Wave

FT = Flux Tilt

<sup>Δ</sup>(PHS→pm→cm)

<sup>ΔΔ</sup>(100 kW / 30 min)

<sup>ΔΔΔ</sup>(100 kW / 5 min)

<sup>ΔΔΔΔ</sup>(8-in lower 100 kW)

### 6.2.1 Calibration Efforts

Prior to the experiment, each MPFD array underwent activation foil calibration in the D67 core position using the process discussed in Section 5.1. Each MPFD array was subsequently tested in pulse and current mode in the D67 core location. During initial calibration of each array, each

node was examined to determine which operation mode (pulse or current) the node was to be utilized during experimentation and is shown in Table 6-4. Array 5 was observed to have significant noise on all nodes and was therefore precluded from the remainder of tests. In total, 24 of the original 28 fabricated MPFD nodes were used for experimental purposes. A more thorough description of the calibration process is provided in Sections 5.1 and 5.2.

*Table 6-4. Electronics operation mode for each of the MPFD arrays. Note that array 5 was not included due to high noise level and spurious response.*

| Array | 1                                           | 2  | 3 | 4  | 6 | 7  |
|-------|---------------------------------------------|----|---|----|---|----|
| Node  | P = Pulse-Mode, C = Current-Mode, * = Noisy |    |   |    |   |    |
| 0     | P                                           | P  | C | C  | C | C  |
| 1     | P                                           | P  | C | C* | C | C  |
| 2     | P                                           | P  | C | C  | C | C* |
| 3     | P*                                          | P* | C | C  | C | C  |

Detector response linearity was confirmed for each of the 24 nodes tested with resulting linear regressions produced for each node and in both current and pulse mode operation. Array 2 was used as a reference during experimentation and remained in the D67 to correlate MPFD response to the calibration procedure. Concerns arose during initial calibration that the MPFD arrays had been manufactured with nodes not mirrored about the fuel region center plane. To confirm this suspicion, MPFD array 1 was modified to be lowered 8-in further into the core. This test took place at the beginning of day 5, and yielded results that refuted manufacturing concerns. After this test, MPFD array 1 was reconfigured to its original design and testing proceeded as planned.

### 6.2.2 Steady State Results

A total of eight steady state tests were performed: four DRT and four flux tilting measurements. To illustrate how local neutron flux deviates at a set power level measured by ex-core sensors, MPFD array 6, operated in current mode, is compared between the D67 calibration position to the C2 position (E56) and C1 position (E45). The three deployment locations represent positions with incremental proximity to the transient control rod and core center. Position E45 is directly adjacent to the transient control rod. Based on the results shown in Fig. 6-6, MPFD array 6 has almost identical response in positions D67 and E56. In contrast, MPFD array 6 response in E45 at 500kW<sub>th</sub> has a significantly elevated response across all channels with an increase of 61.6%,

22.7%, 14.5%, and 64.6% for nodes 1 through 4 respectively. These results indicate a rise in local neutron flux magnitude near the center of the core.

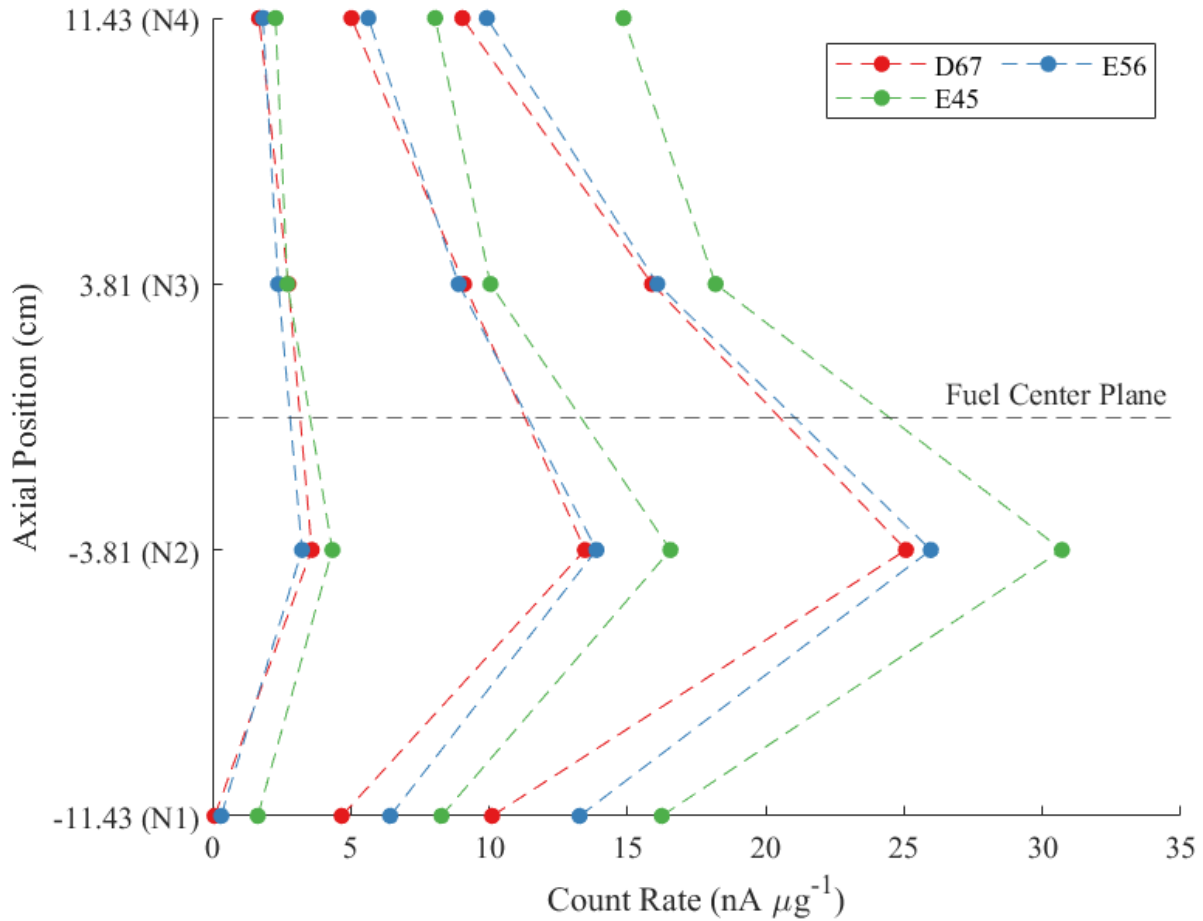


Fig. 6-6. Power and mass-normalized response for MPFD array 6 in three separate core locations. Two power levels are depicted, 100 kW<sub>th</sub> (left curves), 300 kW<sub>th</sub> (middle curves) and 500 kW<sub>th</sub> (right curves). Dotted lines included to emphasize linked data; it does not indicate interpolated information. Data extracted from DRTs performed on day 4 (D67), day 6 (E45) and day 8 (E56).

On day six of testing, flux tilting was performed for each of the 5 control elements. Detectors were arranged in the C1 configuration with array 6 placed directly congruent to the transient rod near the center of the core in the E45 position. Each control element flux tilt, as indicated in Fig. 6-7, was achieved by fully withdrawing the desired rod, and then the remaining control elements were positioned to achieve criticality at 100 kW<sub>th</sub>. The goal of this test was to observe the flux tilt effect from each control blade.

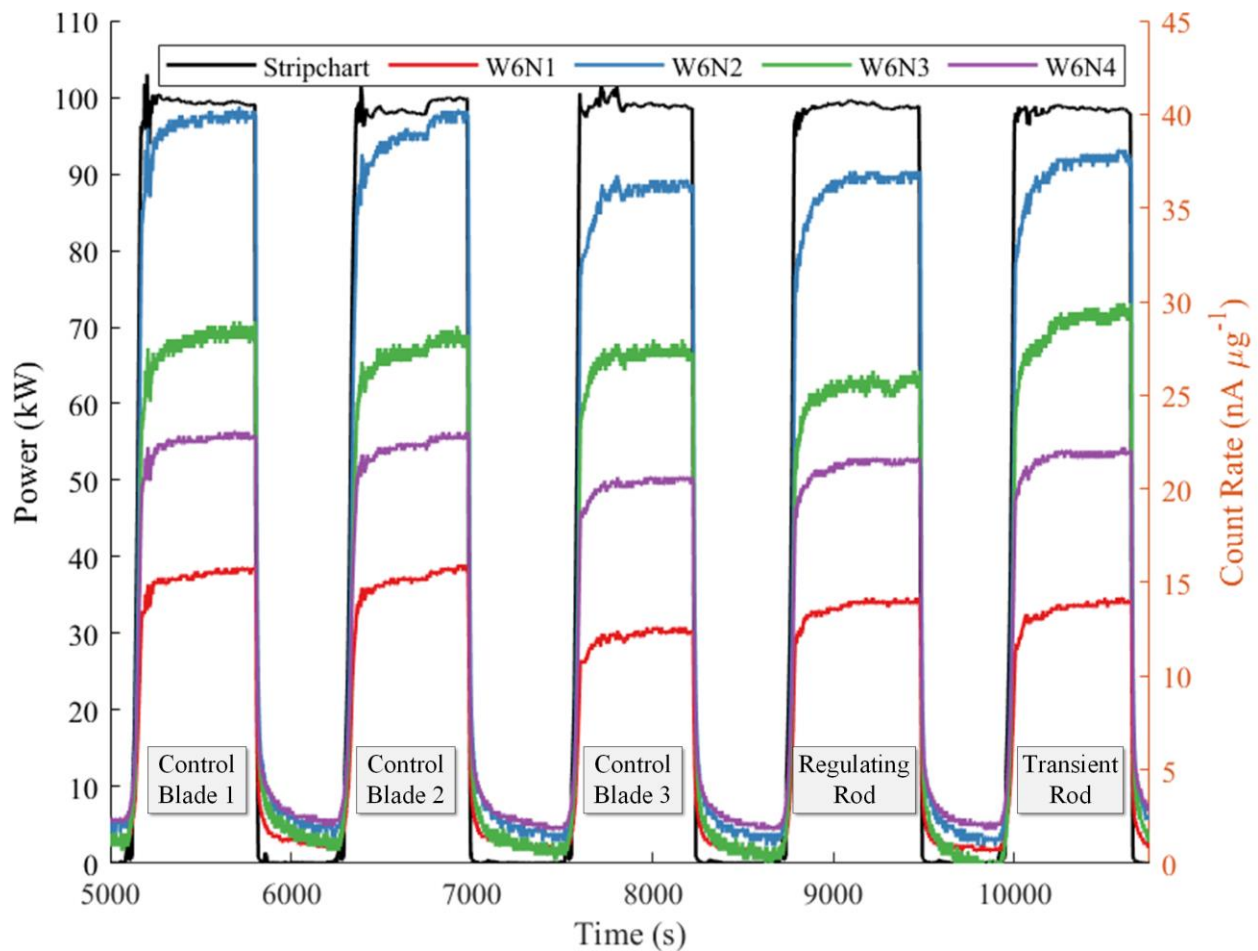


Fig. 6-7. Graph comparing the signal from all four nodes of MPFD array 6 to the reactor power level. This array was in the E45 position during this test, which occurred on day 6 during experiment E.

As mentioned in Section 5.2.1, signal drift is observed during current mode operation on all four detector nodes and is evident in Fig. 6-7. This effect was observed during other tests and was more pronounced during high power experiments. This effect is hypothesized to be the result of the delayed buildup of gamma-ray emitting fission products in the reactor fuel. During prompt changes in reactor power, current-mode operating gas detectors observe an asymptotic increase to equilibrium of signal contribution from gamma rays. At a step change in reactor power, detector current is composed of prompt gamma-ray interactions from direct fissioning of the reactor fuel, and from fission fragments produced from neutron absorption in the MPFD fissile layer. As time progresses, gamma-ray emitting daughter products increase in population in the fuel, raising the delayed-gamma flux which increases until equilibrium. Based on simulations described in Section

3.1.2, at equilibrium conditions for both neutrons and gamma rays, the average energy deposition rate of gamma rays would be 16.7% of the total energy deposition from all sources of ionization (fission fragments, gamma rays, alpha and beta particles). From the results shown in Fig. 6-7, count rates for each node and each flux tilt were compared for the beginning and the end of the tilt. After the reactor power was achieved, MPFD node count rates increased by  $15.4\% \pm 2.80\%$  over the duration that the reactor was at steady state power. Based on the signal responses shown, count rate had not yet achieved an equilibrium value, which may account for the discrepancy between theoretical and experimental values.

### **6.2.3 Transient Results**

Testing at UWNR also included a total of 16 individual transients (6 ramp reactivity insertions, 4 square wave power profiles, and 6 reactor pulses). Ramp reactivity insertion testing was performed with periods of 20, 30, and 50 seconds. Square wave power profiles were achieved for power levels of 250, 500, and 1000 kW<sub>th</sub> and were sustained for 4 minutes. Finally, reactor pulse operations, much like tests performed at KSU gave insight into the fast-transient performance of the MPFD. The UWNR tests provided a unique opportunity that allowed the meshing of the three-dimensional grid of MPFD nodes. This condition was used to observe the reactor pulse effect on a localized flux in real-time as the neutron flux wave propagated across the reactor core.

For each of the pulsed experiments detector performance was compiled by comparing measured FWHM for a single node compared to the FWHM determined by the ex-core sensor. Additionally, all arrays were operating in the same transient triggering mode, whereby the electronics determine count rate by measuring integrated charge per sample interval duration. According to MPDF electronics manual, the MPFD electronics conversion is 1 cps for every 10 pC of integrated charge for the sample intervals. Arrays 1, 2, and 7 have the most consistent response with respect to pulse shape, inter-node comparison, and with deviation to the ex-core sensor. An example comparison of MPFD nodes to reactor ex-core sensor is shown in Fig. 6-8, with comparisons for all six reactor pulses shown in Appendix E.

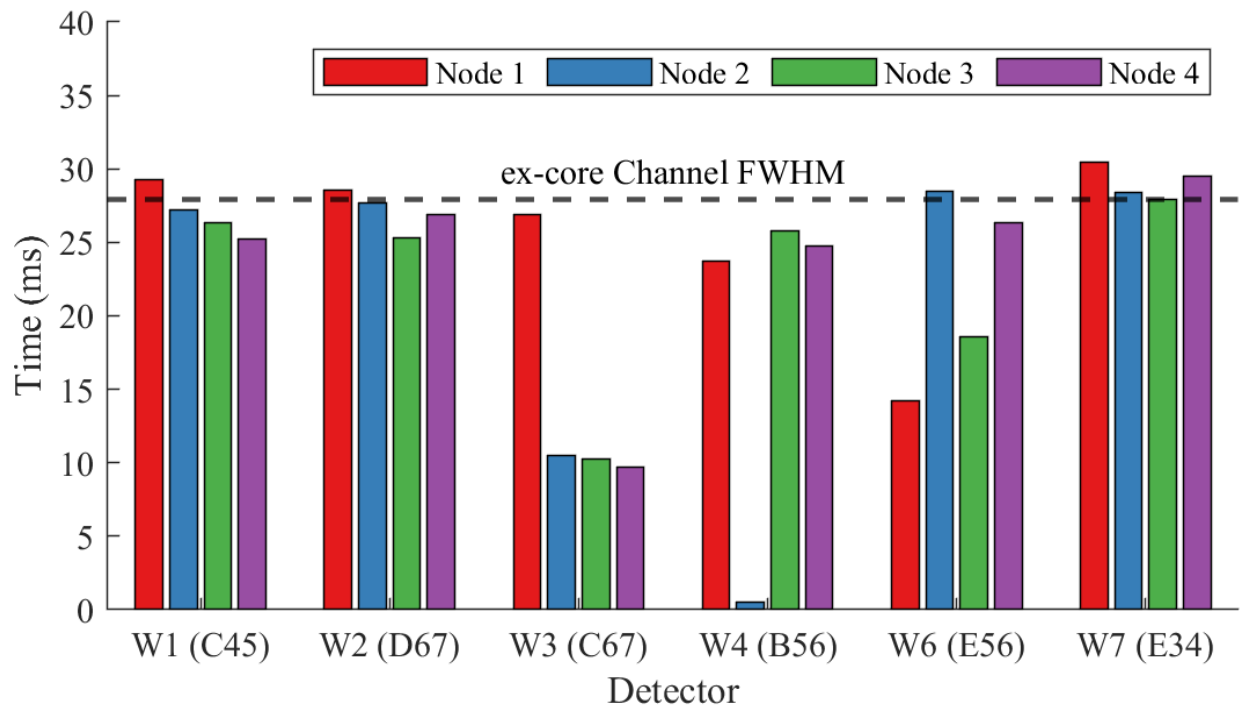


Fig. 6-8. \$1.43 pulse in C2 configuration with MPFD node FWHM comparison to ex-core sensors. Each array position is depicted for spatial reference. Note, that array 4 node 2 had a stagnant, low count rate response during this pulse leading to an erroneous short FWHM.

As part of the reactor pulse tests, it was observed at lower reactivity insertion (\$1.43 pulses), post transient power oscillations were observed both in the ex-core sensors and MPFD nodes. Reactor staff have concluded that these oscillations are due to the transient rod partially rebounding into the core after ejection from the core. This result was more prominent in the \$1.43, and \$1.71 reactivity insertion pulses but is most distinct in the \$1.43 pulses, as shown in Fig. 6-9.

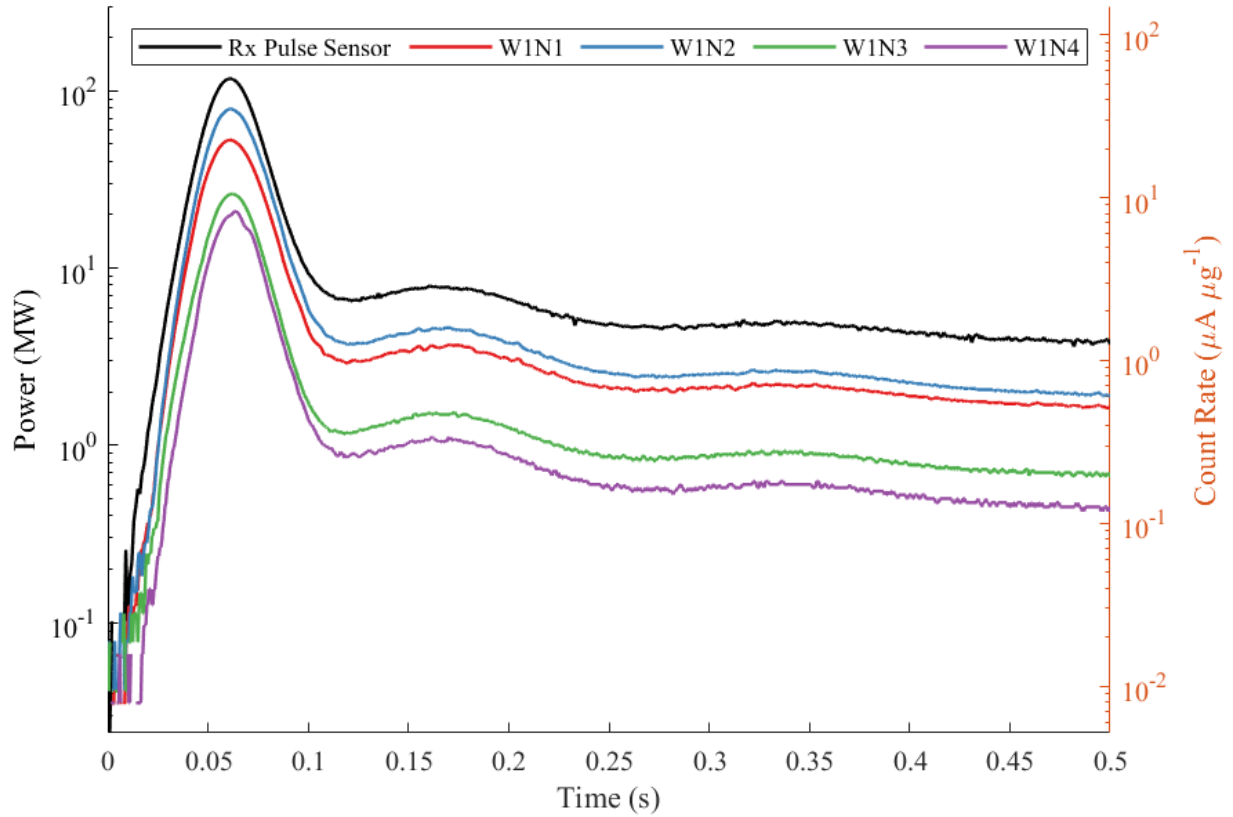


*Fig. 6-9. MPFD array 1 node response compared to reactor sensor response to \$1.43 pulse in C1 configuration showing power oscillations following primary pulse feature.*

### 6.2.4 Summary

These tests constituted the first real-time three-dimensional measurement of reactor core neutron conditions using MPFDs. Prior to testing at UWNR, MPFDs had only been successfully tested with axially dispersed nodes in a linear array. A rich dataset was produced for a variety of steady state and transient operations which has given insight into the reactor core dynamics during operations. The MPFD array set was also able to accurately measure a reactor pulse propagation in three dimensions. While some detectors did not perform as designed or intended, with significant noise, this experiment provided valuable insight into pragmatic practices for future deployments. The complexity of measuring 24 spatially discrete real time nodes was challenging and lessons learned will shape how future deployments are conducted.

## 6.3 Idaho National Laboratory Testing

MPFDs have been tested in the ATR facility at INL during previous research projects [85]. Most recently, with the restart of the Transient Reactor Test Facility (TREAT), efforts have been underway to provide neutron flux monitoring detectors for use during materials testing and as part of the accident tolerant fuels (ATF) campaign [86]. MPFDs are one of a set of neutron monitoring detection platforms that are presently being investigated as support instrumentation during ongoing reactor experiments. Additionally, an upcoming experiment at ATRC, will utilize two 4-node MPFD arrays to measure axial neutron flux magnitude to determine localized neutron flux effects from nearby hafnium Outer Shim Control Cylinders (OSCC). Both experiments will be discussed herein.

### 6.3.1 TREAT Testing

During summer 2018 a series of transient and steady state tests were performed at TREAT during which MPFDs in conjunction with SPNDs, SPGDs, and various other temperature sensors were deployed. The goal of these tests was to document reactor dynamics during various high-power excursions to better understand how the detectors perform with the eventual goal to support Accident Tolerant Fuel (ATF) testing. Experiment tests included loss of coolant accident (LOCA) DRT, temperature-limited reactor pulsing, and narrow pulse width testing. The primary focus of this section will be the temperature-limited pulsing and narrow pulse width testing, as these tests would be the best method of determining detector longevity after successive tests at high flux with short duration. Due to the high magnitude of neutron flux ( $10^{17}$  n cm<sup>-2</sup> s<sup>-1</sup>) and short duration (<300 ms), TREAT reactor pulsing, represents one of the worst-case scenarios for neutron flux monitoring devices. The extreme and rapid change in neutron flux over short periods of time can cause damage to detectors, by generating stagnant heat in the neutron reactive coating which can lead to degradation and potential failure as a result. Throughout the experiment campaign, all sensors, including the MPFDs, were deployed into the K-10 position as indicated in Fig. 6-10. A total of four individual MPFD nodes were deployed simultaneous, but only two were operational during testing: a natural enrichment uranium fissile-coated node, and a natural enrichment thorium fissile-coated node.

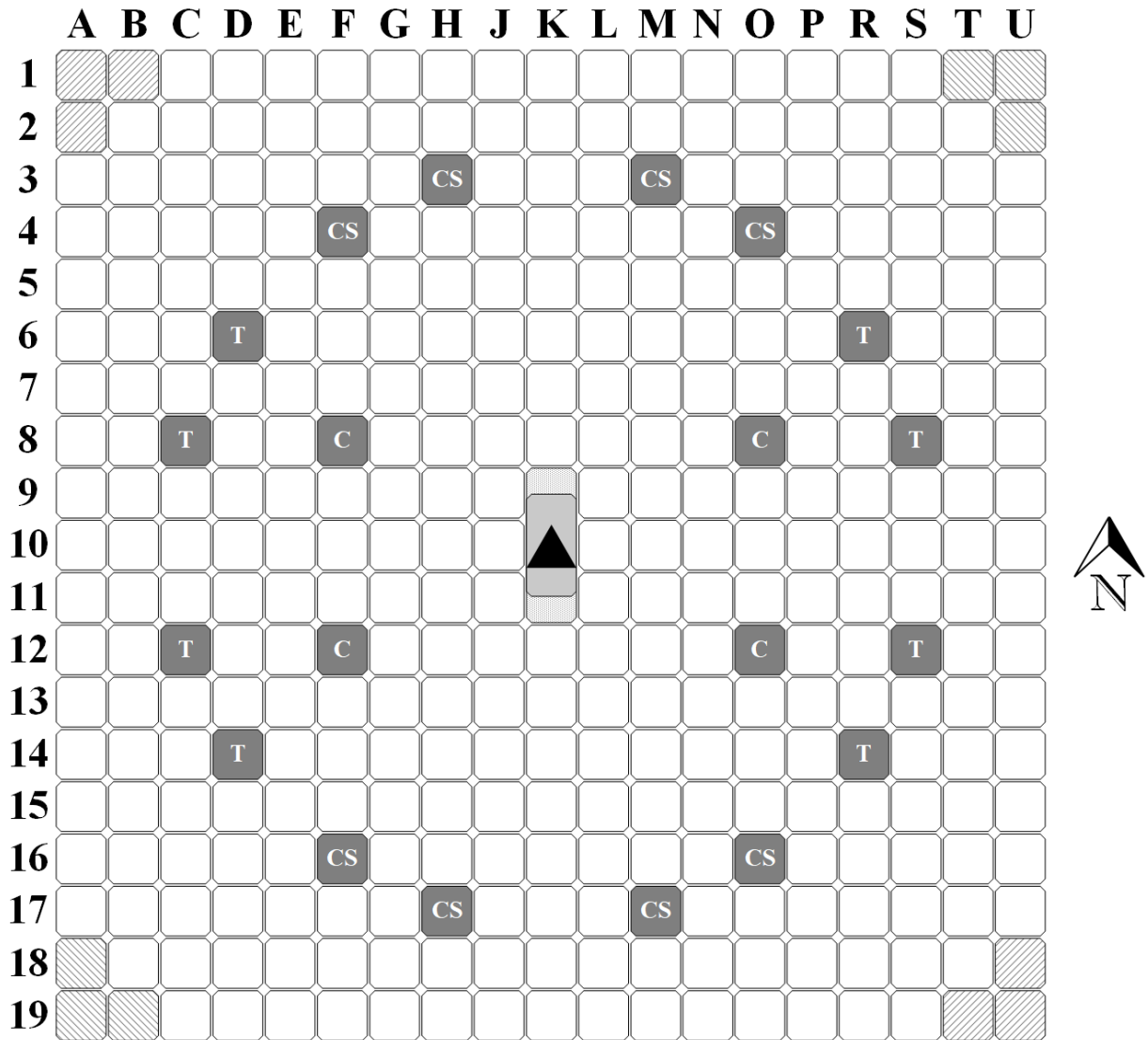


Fig. 6-10. Diagram of TREAT core with the instrument package (black triangle) including the MPFD shown. There are three banks of core rod dive assemblies shown, including the control/shutdown (CS), the compensation I, and the transient (T) rods.

### Reactor Pulsing Results

A total of 7 reactor pulses were conducted over the course of a month of testing, with each test's characteristics listed in Table 6-5. Two forms of reactor pulsing were conducted; natural pulse transients (NPT) during which the power excursion is temperature limited, and narrow-pulse-width clipped transients (NPW) during which a 4.2%  $\Delta k/k$  pulse is initiated and control rods are automatically reinserted after a pre-set delay to terminate the pulse and achieve a pre-defined

FWHM duration between 90 – 100 ms [99]. As mentioned previously, other experiments such as LOCA testing occurred during the test campaign. Transients 2902 and 2903 were both LOCA testing whereby reactor power was brought to 45 MW<sub>th</sub> for 30s and then reduced to 2 MW<sub>th</sub> and held for 400s. Comparing detector responses across the two transients, little to no detector degradation was observed, therefore, these tests will be omitted from discussion.

*Table 6-5. List of TREAT pulses with max power and FWHM listed for comparison. Note that NPW descriptions indicate delay between the start of the transient and control rod reinsertion. Power and FWHM data were captured using ex-core reactor detectors [99].*

| Date                      | Transient # | Experiment Name | Description                             | Peak Power (GW <sub>th</sub> ) | FWHM (ms) |
|---------------------------|-------------|-----------------|-----------------------------------------|--------------------------------|-----------|
| 6/20/2018                 | 2900        | MIT-D           | 1.6% $\Delta k/k$ *                     | 1.30                           | 299.7     |
| 6/25/2018                 | 2901        | MIT-E           | 3.85% $\Delta k/k$ *                    | 12.47                          | 114.1     |
| 7/3/2018                  | 2904        | MIT-G           | 475-ms **                               | 9.24                           | 90.5      |
| 7/11/2018                 | 2905        | MIT-H           | 500-ms **                               | 11.67                          | 90.1      |
| 7/12/2018                 | 2906        | MIT-I           | 513-ms **                               | 12.74                          | 92.2      |
| 7/17/2018                 | 2907        | MIT-J           | 525-ms **                               | 13.66                          | 91.5      |
| 7/19/2018                 | 2908        | MIT-L           | 450-ms **                               | 6.08                           | 88.7      |
| * Natural Pulse Transient |             |                 | ** Narrow-Pulse-Width Clipped Transient |                                |           |

The tests listed represent the highest power levels experience by MPFDs. The thermal neutron flux capabilities of the TREAT reactor extend to a neutron flux of  $10^{17}$  n cm<sup>-2</sup> s<sup>-1</sup>. Concerns prior to testing revolved around delamination of the fissile layer due to the short duration high energy deposition in the fissile layer, Ti/Pt layer, and alumina substrate. The tests provided insight into the fissile layer durability as illustrated by Fig. 6-11 showing count rate vs power level during consecutive pulses. Three outlier cases emerged which all occurred for power levels greater than 12 GW<sub>th</sub>. Initially, it was speculated that this decrease in node sensitivity was a result of detector count rate saturation due to the high neutron flux [99], [100]. An alternative theory was developed based on the rebounded response shown in tests 3, 4, and 7, coupled with no saturation effects for test 4 at 11.67 GW<sub>th</sub>, and a consistent count rate ratio between the thorium and uranium nodes. The current working theory is that the device operating parameters, such as applied bias and shaping time, were altered to reduce probability of saturating the support electronics response during these high-power pulses. Reducing the operating voltage will reduce the electric field in the detector chamber, producing a lower count rate vs. power result while maintaining the ratio of node response.

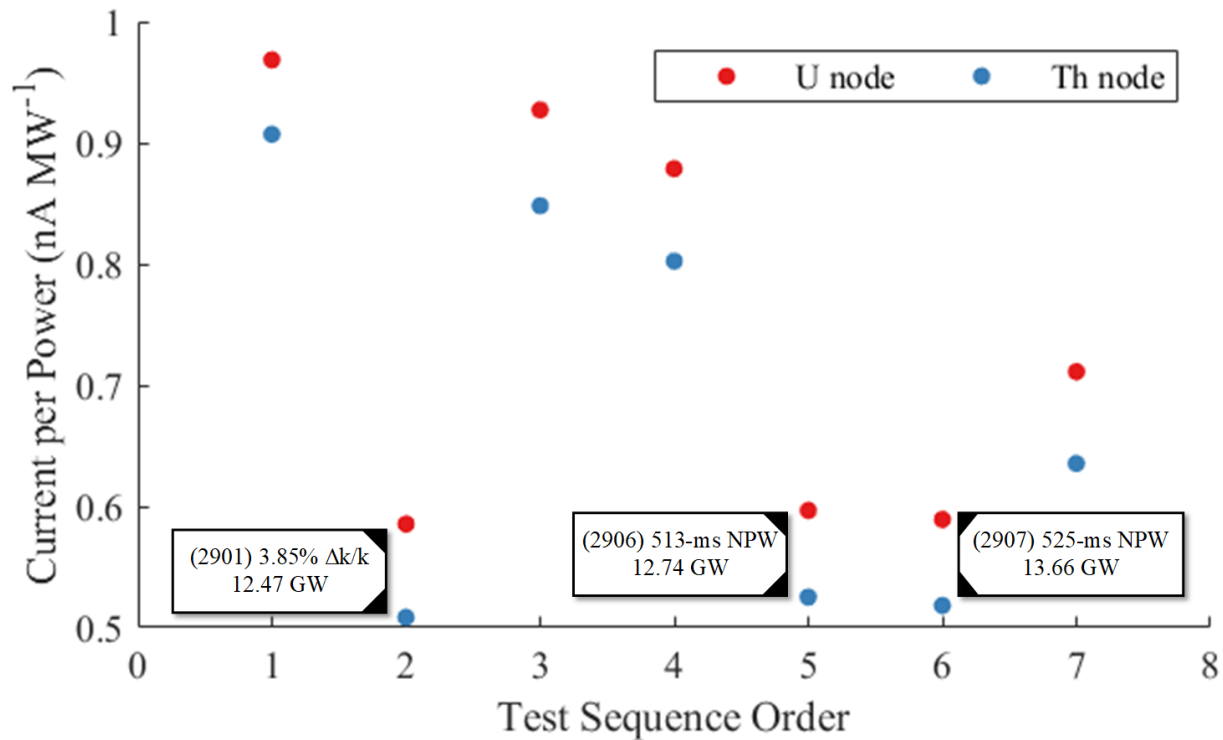


Fig. 6-11. Figure comparing consecutive reactor pulsing tests with peak current compared to reactor power. Note three outlier cases with data points labeled all with power levels greater than  $12 \text{ GW}_{th}$ .

If detector node saturation was the predominant factor in reducing detector response, the final pulse test, transient 2908 at  $6.08 \text{ GW}_{th}$  reactor power, should have regained sensitivity correlating to a value between tests 1 – 3 ( $0.94$  and  $0.87 \text{ nA MW}^{-1}$  for the uranium and thorium nodes). Instead, transient 2908 was observed to have a diminished sensitivity of  $0.71$  and  $0.64 \text{ nA MW}^{-1}$  for the uranium and thorium nodes, respectively. These results can be better represented and understood as shown in Fig. 6-12, whereby each test is plotted corresponding to the reactor power achieved. Based on these results, it is reasonable to conclude that other effects such as fissile layer degradation may have contributed to a loss of sensitivity after consecutive high-power reactor pulses. Unfortunately, these detector nodes were damaged and decommissioned before additional testing could be performed to diagnosis this result. Until further testing with consistent operating parameters can be performed, this conclusion will remain as a working theory as to why non-linear detector response was observed during the experiment campaign. It is important to note that uranium to thorium current ratios stayed consistent through the testing at  $1.114 \pm 0.030$  (2.68%).

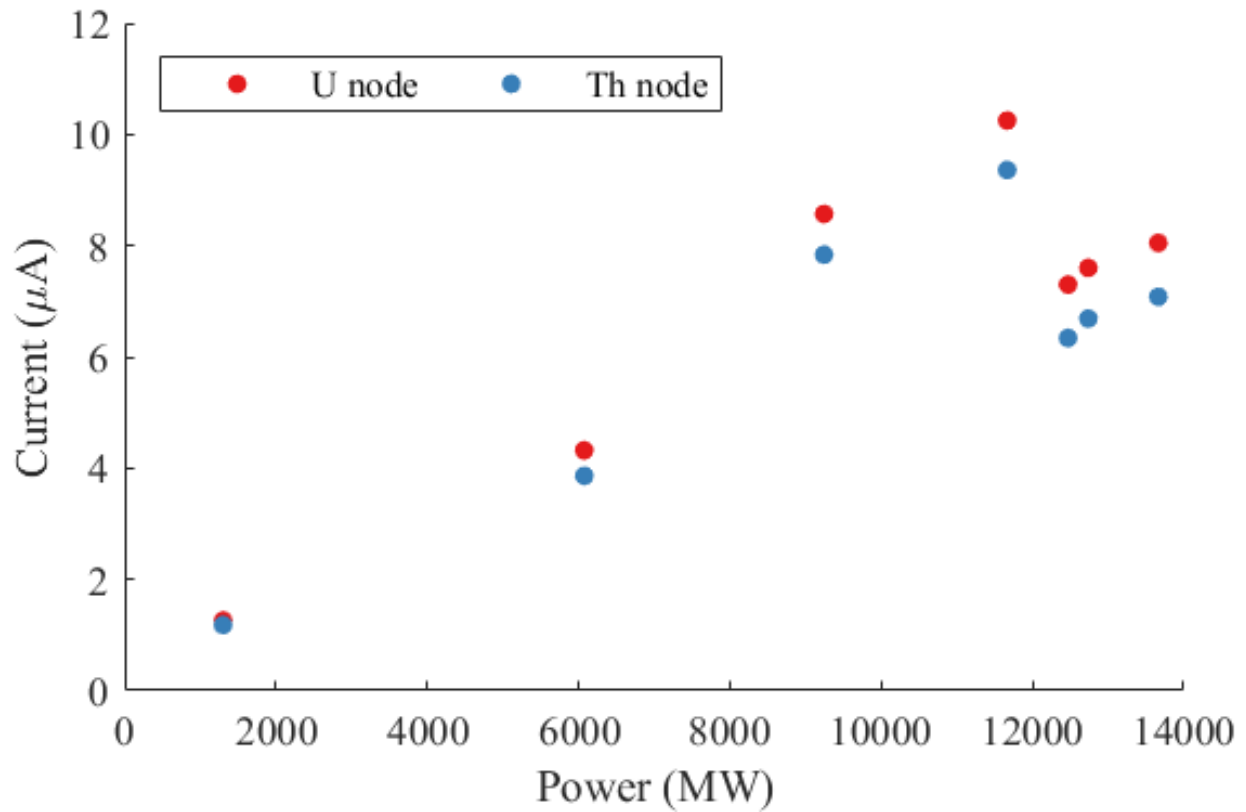


Fig. 6-12. Graph of MPFD node current compared to reactor power level. Note that experiments 2, 6 and 7 are outlier values located at 12.47, 12.74, and 13.66  $GW_{th}$  respectively.

### U vs. Th Maximum Response Peak Shift

Throughout testing, both MPFD nodes were able to track the reactor pulses within  $\leq 20\%$  deviation of the ex-core sensor measurements, even with the aforementioned operating parameter adjustment. A summary of MPFD node maximum count rates and FWHM results are shown in Table 6-6, in comparison to ex-core sensor results. Among the test results from all 7 transients, a pattern of delayed response between the uranium and thorium node was observed. This observation is most likely due to processing electronics delay as evidenced in transient 2907 (shown in Appendix G), wherein a common signal artifact is observed on both the uranium and thorium node with similar shape and amplitude.

Table 6-6. Summary of MPFD data compared against each of the pulse transients.

| Trans # | Ex-core sensor            |             | MPFD Node 2 (U)   |           |            | Peak Offset (ms) | MPFD Node 3 (Th)  |           |            |
|---------|---------------------------|-------------|-------------------|-----------|------------|------------------|-------------------|-----------|------------|
|         | Power (MW <sub>th</sub> ) | FWHM M (ms) | Peak Current (μA) | FWHM (ms) | FWHM diff. |                  | Peak Current (μA) | FWHM (ms) | FWHM diff. |
| 2900    | 1301                      | 299.7       | 1.26              | 346.0     | 15%        | 76               | 1.18              | 361.0     | 20%        |
| 2901    | 12469                     | 114.1       | 7.30              | 128.5     | 13%        | 73               | 6.34              | 130.2     | 14%        |
| 2904    | 9244                      | 90.5        | 8.57              | 102.6     | 13%        | 53               | 7.84              | 103.9     | 15%        |
| 2905    | 11666                     | 90.1        | 10.25             | 102.9     | 14%        | 75               | 9.36              | 103.8     | 15%        |
| 2906    | 12738                     | 92.2        | 7.60              | 101.7     | 10%        | 12.5             | 6.69              | 103.0     | 12%        |
| 2907    | 13662                     | 91.5        | 8.05              | 103.4     | 13%        | 68.5             | 7.08              | 104.7     | 14%        |
| 2908    | 6075                      | 88.7        | 4.32              | 103.6     | 17%        | 8                | 3.86              | 105.9     | 19%        |

Upon examination of the data, there is no apparent correlation between power level and peak offset, or test sequence and offset. At present time, further experimentation is necessary to determine if non-intuitive reactor dynamics or erroneous electronic influence is the cause of this effect. In all pulse transients, the uranium node responded and reached peak count rate prior to the thorium node achieving peak count rate. Raw node data and reactor ex-core sensor data for each transient are provided in Appendix G.

## Summary

The tests at TREAT mark a new upper limit of operability for the MPFD, with a new maximum power level of 13.7 GW<sub>th</sub>. These tests provided valuable insight into the maximum neutron flux the detectors can handle, but also highlighted deficiencies in the design that will need to be addressed. The chief concern is the detector longevity during frequent high neutron flux, short duration tests. As mentioned previously, over the course of one month of testing, the detector nodes experienced 7 transients between 1.3 and 13.7 GW<sub>th</sub>, which resulted in depreciated detector performance. Detector sensitivity dropped 26.6% for the uranium node and 29.9% for the thorium node when comparing responses between transient 2900 and 2908 as seen in Fig. 6-11. The cause of this depreciation is unclear and could be the result of fissile layer delamination. Additional high neutron flux testing will be necessary to adequately diagnosis this result to determine the causing factor. Future fabrication should include fissile layer annealing to reduce delamination probability. Additionally, consecutive tests should be conducted at a consistent peak power level to determine if detector sensitivity degrades proportional to fluence or reaches an asymptotic value.

### 6.3.2 Planned ATRC Testing

Much of the discussion in Section 3.1 centered around simulations for an upcoming experiment at the ATRC reactor. Experimental deployment was planned for early 2021 but was impacted by the recent COVID-19 pandemic which occurred throughout 2020 and parts of 2021. Therefore, detailed explanation of the planned experiment will be given with commentary regarding expected outcomes of the test campaign. An instrument testing assembly was developed and will house two 4-node MPFD arrays alongside two Rh-SPNDs and a flux wire deployment tube, in which flux wires will be inserted and removed throughout testing. The local neutron environment will be corroborated using activation wires much like in the UWNR testing, but these wires will be collocated during testing rather than pre-deployed prior to the experiment. MPFD nodes are equally spaced in the axial dimension to span the 49.5" length of the fuel region of the ATRC core. To achieve this, the two MPFD arrays are displaced; one to occupy the upper portion of fuel region, and the other to occupy the lower region. A single node from each array will be collocated at the center plane of the fuel region and will be used to correlate the response between the two MPFD arrays. The two Rh-SPNDs will also be collocated at the fuel center plane throughout the experiment. Fig. 6-13 depicts a conceptual diagram illustrating the deployment depths of the various sensors to be used during experimentation.

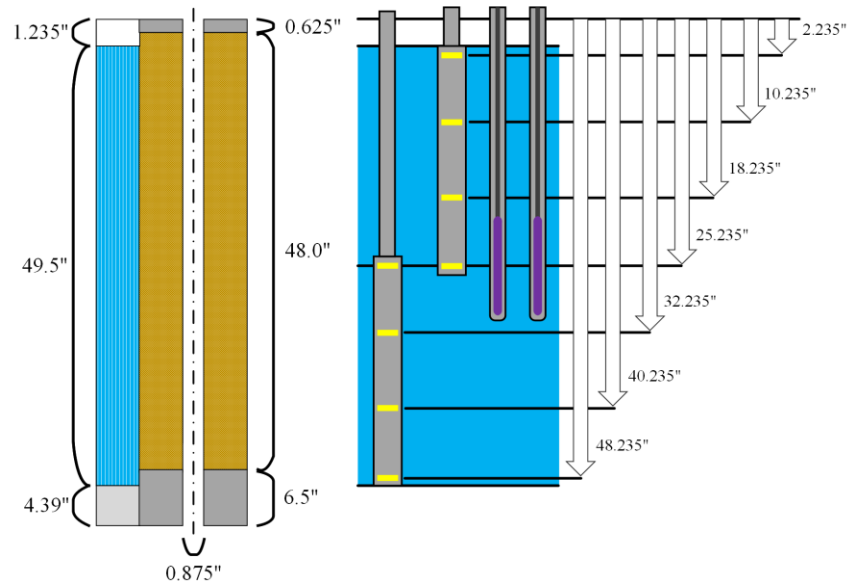


Fig. 6-13. Conceptual diagram illustrating deployment depth of the various sensors as part of the experiment. The fuel region (light blue) center plane, located at 25.235" depth for the upper grid plate will be where one node of each MPFD array (yellow) as well as the Rh-SPNDs (purple) will be collocated.

The instrument package will be deployed in the B-8 irradiation port, shown in Fig. 6-14, which is situated between the fuel and the Outer Shim Control Cylinders (OSCC) located in the northern portion of the core. The goal of the experiment is to determine local neutron flux modification dependency on the nearby hafnium OSCC. Three rotation angles, inward ( $5.73^\circ$ ), nominal ( $58.55^\circ$ ) and outward ( $150.1^\circ$ ), were chosen to determine the effect. Tests will be conducted at static OSCC angles as well as transitory conditions, whereby control drum angles will be altered during a predetermine duration. If the local neutron flux magnitude and axial profile can be recorded accurately, experimenters may begin to use this irradiation location, with higher confidence of the localized neutron environment.

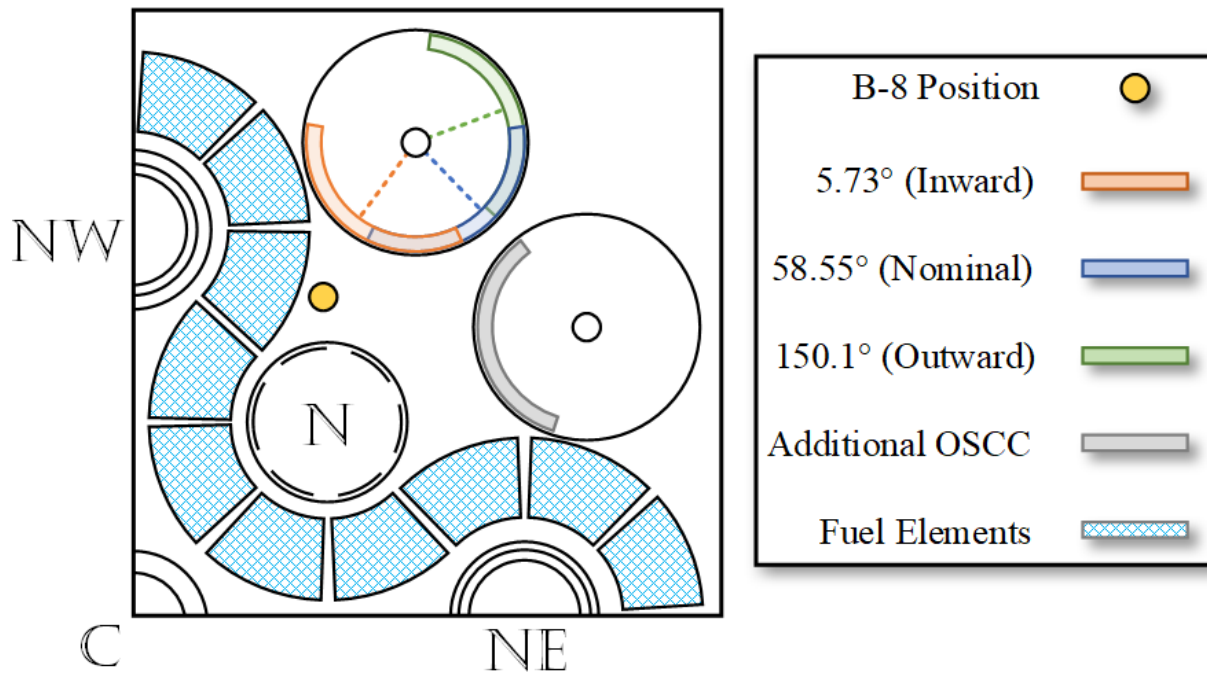


Fig. 6-14. Diagram of ATRC core northern region. Note the diagram only includes relevant reactor features for concept of the experiment in the B-8 irradiation location. Note, the dashed lines in upper OSCC indicate the center point of the hafnium control drum at the various angles ( $5.73^\circ$ ,  $58.55^\circ$  and  $150^\circ$ ).

### Axial Profile Response Based on Rotation Angle

As part of the planned experiment, 8 MPFD nodes will be dispersed along the height of the fuel region of the cord. The node responses will then be used to develop an axial profile of the neutron flux population throughout the fuel region. As mentioned in Section 3.1.3, simulations work was performed to estimate neutron flux and detector response for various axial positions and

OSCC drum angles. The results of the simulations, as shown in Fig. 6-15, were used to estimate the ideal node locations for deployment. The goal is to then use the nodal response to interpolate between nodes to build a neutron flux distribution for axial positions and intermediate drum angles.

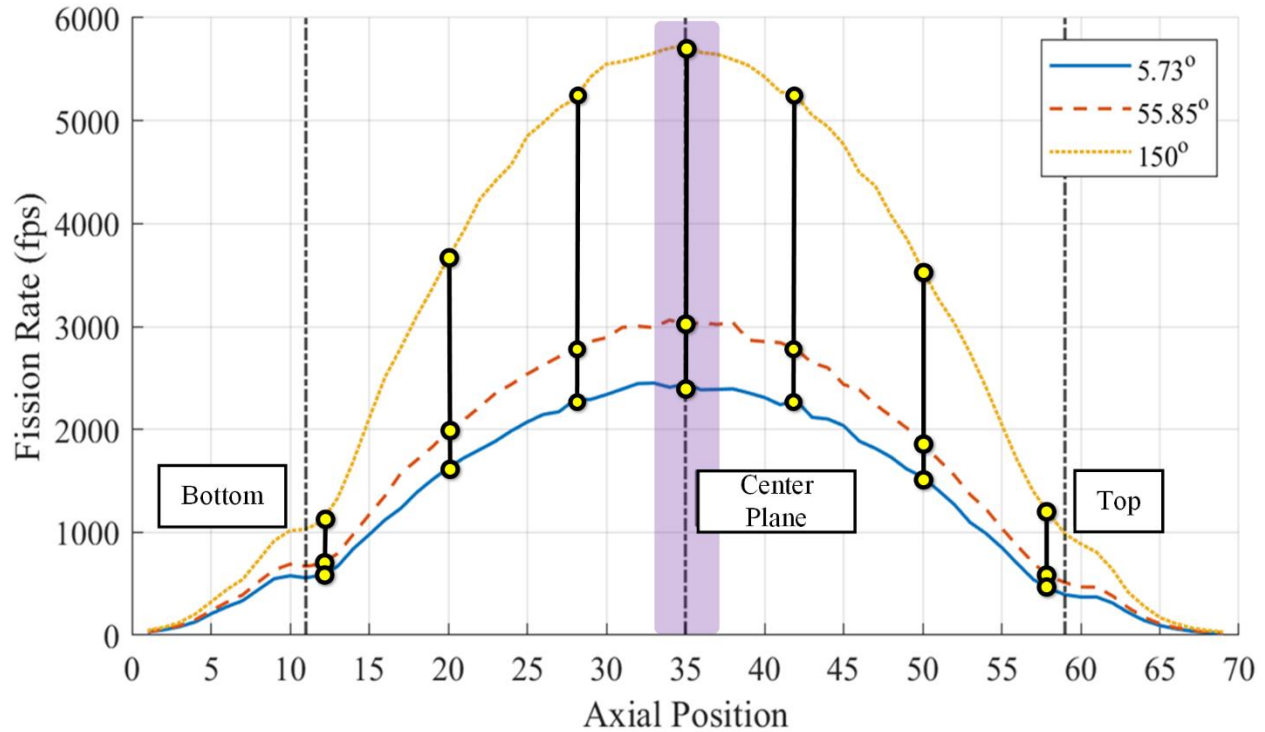


Fig. 6-15. Plot of simulated MPFD fission rates for axial positions and OSCC drum angles. MPFD node locations (yellow) and SPND emitter (purple) are superimposed to illustrate sensor locations.

Placement of the MPFD nodes will be located at  $\pm 23''$ ,  $\pm 15''$ ,  $\pm 7''$ , and  $0''$  relative to fuel center plane (shown as positions 12/58, 20/50, 28/42, and 35 in Fig. 6-15). The axial minimum and maximum node placements ( $\pm 23''$ ) allow for the largest span of the fuel region without impact of the thermal flux peak due to the axial reflectors. Using the data collected from these 8 nodes, cosine function fitting can be used to perform inter-node interpolation.

The concluding remarks of the MPFD research will be presented herein, focusing on contributions to the science, as well as commentary about the application of the MPFD platform and a general assessment of the current technology readiness level (TRL) will be provided. Proposed future development goals will be provided for follow on work.

## 7.1 Contributions to the Science

Prior MPFD research focused on taking the MPFD platform from benchtop testing to in-core deployments. Customized electronics were designed and implemented into the MPFD measurement system, and testing was performed to characterize preliminary response to various neutron flux conditions. Additionally, previous MPFD research successfully developed a method of coating uranium and thorium fissile deposits for use in the manufacturing of detectors [6]–[9]. The research presented herein built off the prior accomplishments by wholistically analyzing the design of the detector (fissile coating, chamber shape, electric field, components for manufacturing, etc.) to develop and optimize the design of the MPFD. These developments were inter-woven with periodic experimental testing that informed researchers about potential drawbacks of the design. During this research, the MPFD platform has now been experimentally tested in a larger variety of in-core conditions compare to prior research. Testing such as reactor pulsing demonstrated the fast response of the detectors, whereas steady state flux tilting performed at UWNr demonstrated the ability of the platform to measure localized perturbations in neutron flux. Moreover, high power pulse testing at TREAT, demonstrated the ability for the detector platform to survive and successfully track extreme neutron flux environments ( $10^{17}$  n cm<sup>-2</sup> s<sup>-1</sup>). The development of the technology is not complete and further research must be performed to commercialize the technology, but this research has provided several contributions to the science of the MPFD technology that will be mentioned.

### 7.1.1 Maximum Power Level Testing

Two MPFDs were tested, one with natural enrichment uranium and one with natural enrichment thorium, at a new maximum power level of approximately 13.7 GW<sub>th</sub>, which occurred at the Transient Reactor Testing (TREAT) facility during summer 2018. Both MPFD nodes

tracked the 13.7 GW<sub>th</sub> reactor pulse which had a FWHM of 115.8 ms. The uranium and thorium nodes respectively measured the reactor pulse to have a FWHM of 128.5 ms and 130.2 ms [10]. During this reactor transient, MPFDs experienced a maximum neutron flux estimated to approximately  $10^{17}$  n cm<sup>-2</sup> s<sup>-1</sup>. These reactor pulse transients were part of a larger in-core testing campaign at TREAT. As testing continued, the MPFD nodes showed signs of detector performance degradation, but continued operating for several other high-power pulses ranging between 1 - 13 GW<sub>th</sub>. During each of these high-power tests, the MPFDs were able to track reactor power level across approximately 3.5 decades of power.

### **7.1.2 Three-Dimensional Core Mapping**

A set of 24 MPFD nodes were dispersed throughout the University of Wisconsin-Madison Nuclear Reactor (UWNR) core providing real-time spatially-discrete three-dimensional neutron flux monitoring. During tests at UWNR, MPFDs were utilized to produce a rich dataset which will be used to validate code as part of the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program. Experimental tests included steady state operations with flux tilting, and transients such as square wave, ramp, and pulsed reactivity insertions.

### **7.1.3 Fast Transient Tracking**

Reactor pulsing experiments were conducted at 3 separate research reactors (UWNR, KSU and TREAT). For each deployment, the measured FWHM of the MPFD closely tracked reactor pulse transients response within  $\pm 15\%$ . During these tests, MPFD achieved successful tracking resolution of 0.25 ms. Each test provided unique outcomes for the research. Tests at KSU demonstrated the first successful tracking of two pulse transients (\$2.50 and \$2.77 reactivity) with 11.46% and 8.61% deviation from Fuchs-Nordheim predicted FWHM and gave experimental insight into necessary operating characteristics for future tests. The UWNR tests signified the first experiment during which MPFD nodes were placed in a three-dimensional set to measure a reactor pulse. Previously, simultaneously deployed MPFD nodes had only successfully measured axial core locations. Finally, testing at TREAT allowed for the study of node response degradation and facilitated the opportunity to push the detectors to the highest flux recording to date. Over the course of the seven high power TREAT tests, ranging from 1 – 13 GW<sub>th</sub>, it was observed that MPFD response reduced by 26.6% for a MPFD node with a uranium fissile layer and 29.9% for a MPFD node with a thorium fissile layer.

#### **7.1.4 Electric Field Optimization**

During detector development for the UWNR experiments, electric field simulations were conducted, and the diamond chamber geometry was confirmed to have a detrimental electric field orientation that precludes its use in multi-node MPFD arrays. This discovery led to the adoption of a new chamber geometry (clover design) that allows for more successful multi-node operation in a common-cathode geometry. This new design also optimizes charge carrier trajectories inside the detector chamber while migrating in the electric field.

#### **7.1.5 Device Calibrations**

Extensive calibration efforts were conducted as part of the experiment campaign at UWNR. Calibration measurements were conducted in a single reference location using a variety of activation foils. This calibration procedure represents the first attempt to experimentally correlate MPFD response to the local neutron energy spectrum. Detection efficiencies were determined for MPFD array 3 to approximately be  $(22.1 \pm 1.64) \times 10^{-12}$  cps/nv for pulse-mode operations, and  $(6.89 \pm 0.25) \times 10^{-21}$  A/nv for current-mode operations, constituting a 0.129% and 22.98% detection efficiency for pulse and current mode respectively.

#### **7.1.6 Instrument Deployment Design**

For the upcoming Laboratory-Directed Research & Development (LDRD) experiment at ATRC, extensive MCNP simulations were conducted to determine the fission rate and fission fragment deposition spectra in the MPFD for the given deployment location (B-8 Position). These simulations were used as the basis when selecting the fissile mass for use in the MPFD. Additionally, simulations were used to estimate non-neutron signal impact from gamma rays, alpha particles, and beta particles. These values were compared against fission fragment energy deposition estimates and were used to build an estimated energy deposition spectrum.

#### **7.1.7 Improved Detector Fabrication Techniques**

A very-large scale integration (VLSI) technique known as photolithography was used to produce high precision Ti/Pt electrode contacts for use in the electrodeposition process. This technique has also improved the manufacturing repeatability of the detectors, and will significantly reduce the cost per piece, opening the door for mass-production of the devices. Previous methods had limited production to between 48 and 256 disks per evaporation, in comparison to the new

photolithography method which can yield an estimated 600 disks per evaporation with low feature variance; 0.005 mm standard deviation for a feature diameter of 0.756 mm.

## **7.2 Application of MPFDs**

The primary focus of MPFD research has been the development as a support instrument in research experiments. As such, design methodology has been focused on accommodating research reactor environments and custom experiment conditions. While the MPFD platform is not limited to only research reactor applications, significant design modifications will be required to perform reliably in the commercial power reactor market.

### **7.2.1 Research Reactors**

Future research reactor applications of the MPFD should focus on discrete spatial sensitivity experiments, where high fidelity point detection of neutron flux is desired. MPFDs, in comparison to other active neutron flux monitoring technologies, have the distinct advantage of providing highly localized neutron flux measurement capabilities. Because of this characteristic, linearly arrayed MPFDs have the potential to fill the role of flux wires, providing customers with the ability to measure axial flux profiles in real-time, which can quickly and accurately characterize irradiation locations. To aid in this effort, future MPFD research could focus on the development of multi-node arrays with more than 4 nodes.

### **7.2.2 Power Reactors**

To accommodate the harsher environment of power reactors, development of the MPFD platform will need to focus on higher temperature/pressure stability, compatibility with corrosive materials, as well as identify the proper mixture of fertile and fissile isotopes to increase detector response stability over the course of its deployment lifetime. As the nuclear industry continues development of next generation reactors such as Small Modular Reactors (SMRs), microreactors, and space reactors, MPFD design methodology should pivot to pursue adaptability in these unique environments.

## 7.3 TRL Estimation

The MPFD technology has been demonstrated to have the capability to serve in a multitude of neutron monitoring roles, both steady state and fast transient, as well as in high and low flux environments. The technology has advanced to large-scale deployments and coupled with specialized read-out electronics. Based on these achievements, and common technology readiness assessment methodology, shown in Fig. 7-1, the status of the MPFD is TRL 7, as it has been pushed beyond the prototype stage and tests have now been conducted at engineering scale. To continue the natural progression of the technology, research and development must now focus on finalizing optimization of the design and pursuing commercialization efforts. To that end, key customers of the detector must be identified to utilize their interest in the technology to determine application scope.

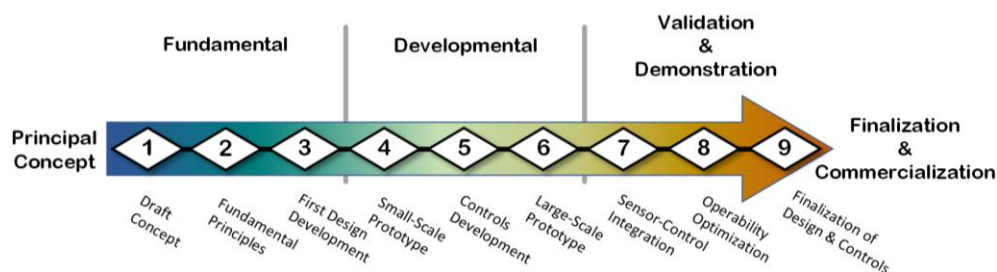


Fig. 7-1. Technology Readiness Level (TRL) scale depicting the development stages of a technology from conceptual idea to final commercialization.

## 7.4 Future of the MPFD Design

As mentioned in the Section 7.2, MPFDs at their present development stage are suitable for use in research settings for tests such as flux profile measurements and low fluence materials testing. Over the course of the last decade of MPFD research, the technology has matured from a benchtop prototype to a large-scale deployed and demonstrated three-dimensional neutron flux mapping platform. Custom-made electronics have been tailor-designed to work in conjunction with the MPFD sensor and provide real-time data to the experimenter while in use. The next step in development must focus on providing a manufacturing method that allows for MPFDs to have a high throughput and a low variance in sensitivity. Once accomplished, the MPFD platform can be easily commercialized and begin use as an “off-the-shelf” advanced sensor, providing high precision, spatially discrete measurements of neutron fluxes.

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### Peer Reviewed

D. M. Nichols, M. A. Reichenberger, A. D. Maile, M. R. Holtz, D. S. McGregor, “Simulated Performance of the Micro-Pocket Fission Detector in the Advanced Test Reactor Critical Facility,” *ANS Nuclear Science and Engineering*, vol. 195, pp. 1098–1106, 2021

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As part of the MPFD research, the exact uranium or thorium compound deposited on the Ti/Pt layer was never confirmed using chemical analysis methods but based on density estimates provided in Section 4.2.2, it was assumed that the fissile layer is primarily composed of  $\text{UO}_2(\text{NO}_3)_2$  with a density of  $2.81 \text{ g cm}^{-3}$ . Thorium fissile layers were assumed to be  $\text{Th}(\text{NO}_3)_4$  and have a density of  $2.80 \text{ g cm}^{-3}$ .

Simulations were performed with two prominent fission fragments were assumed, ( $^{95}\text{Sr}$  at 100 MeV and  $^{139}\text{Xe}$  at 70 MeV) and utilized to characterize escape probability from the fissile layer for the two compounds fissile layer compounds. Using TRIM, each fission fragment was simulated in both  $\text{UO}_2(\text{NO}_3)_2$  and  $\text{Th}(\text{NO}_3)_4$  to produce Bragg ionization curves and energy loss curves:

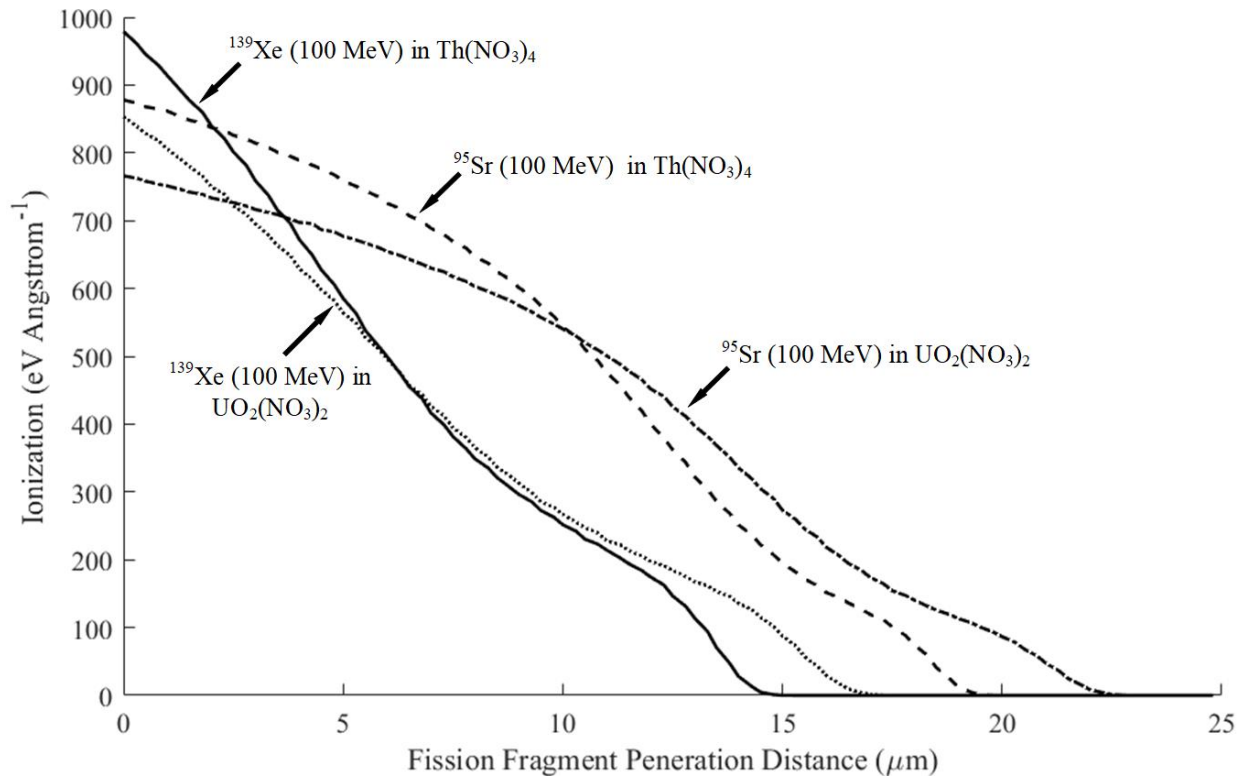


Fig. A-1. Bragg ionization curves for the two chosen fission fragments in uranyl nitrate and thorium nitrate.

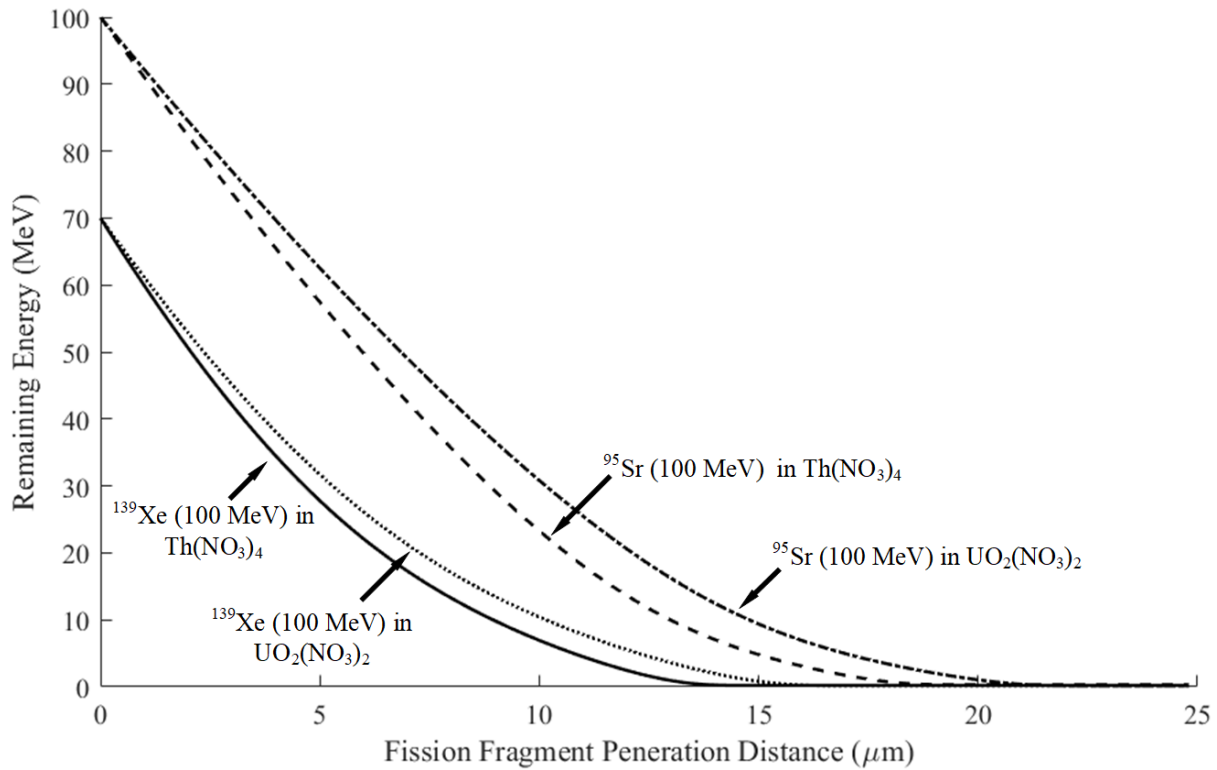


Fig. A-2. Plot of remaining fission fragment energy as a function of traversed distance in the fissile layer.

From these results, it is observed that fission fragments have a high probability of escaping the fissile layer with a thickness of less than 14  $\mu\text{m}$  for both uranyl nitrate and thorium nitrate. As seen in topographic maps of electrodeposited MPFD samples, shown in Fig. 4-13, most of the fissile layer appears to be less than 10  $\mu\text{m}$  in thickness. To determine fissile layer thickness impact on fission fragment escape probability, a series of MCNP simulations were performed with a library of 505 fission fragment isotopes according the MCNP source term described in Section 3.1.2:

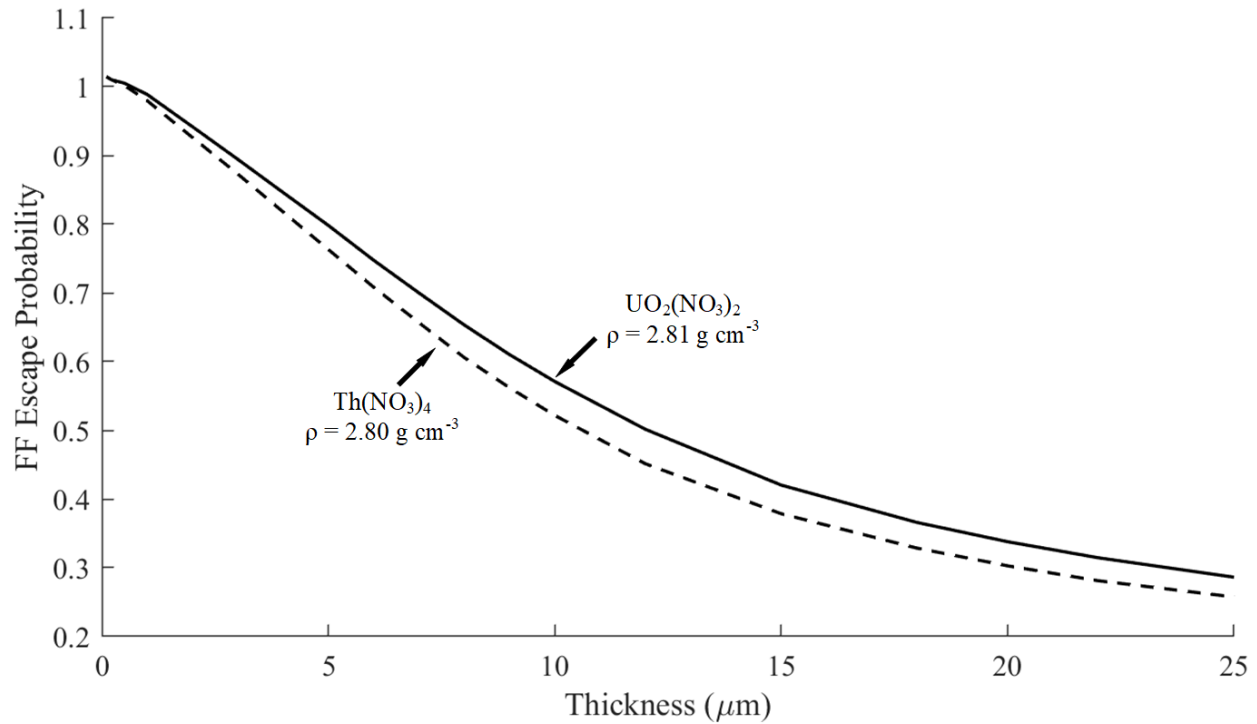


Fig. A-3. Fission fragment escape probability as a function of fissile layer thickness.

It is important to note that the fission fragment escape probability depicted in the Fig. A-3 considers no energy threshold for a fission fragment to be counted. From the above plot there are two notable observations: (1) for thicknesses less than  $0.5 \mu\text{m}$ , fission fragment escape probability can exceed 100%, and (2) a 50% fission fragment escape probability occurs at approximately 10 and  $12 \mu\text{m}$  for uranyl nitrate and thorium nitrate respectively. For the first observation, a fission fragment escape probability of greater than 100% seems nonintuitive. This result is based on the small probability that with a small fissile layer, there can exist a scenario whereby one fission fragment is emitted toward the gas, and other fragment can backscatter of the substrate and enter the gas. Therefore, it is possible that two fission fragments can enter the chamber, but only for minimally thin fissile layers.

Additionally, a series of simulations using the standard MPFD geometry, depicted in Fig. 3-2, and with varying thickness and composition of the fissile layer were performed to determine estimated real fraction of particles that would produce signals as a function of LLD. Total neutron flux was simulated for ATRC B-8 position at  $700 \text{ W}_{\text{th}}$  with OSCC at a drum angle of  $150^\circ$  ( $8.8 \times 10^9 \text{ n cm}^{-2} \text{ s}^{-1}$ ). Gas pressure in the chamber was simulated at 60 psig and the fissile layer was set to a diameter of  $0.250 \mu\text{m}$ . The crossing rates for various fissile layer thicknesses and certain LLD

settings were determined and are shown in Fig. A-4 and Fig. A-5. Crossing rate as a function of LLD in comparison to the ideal fission rate are shown in Fig. A-6.

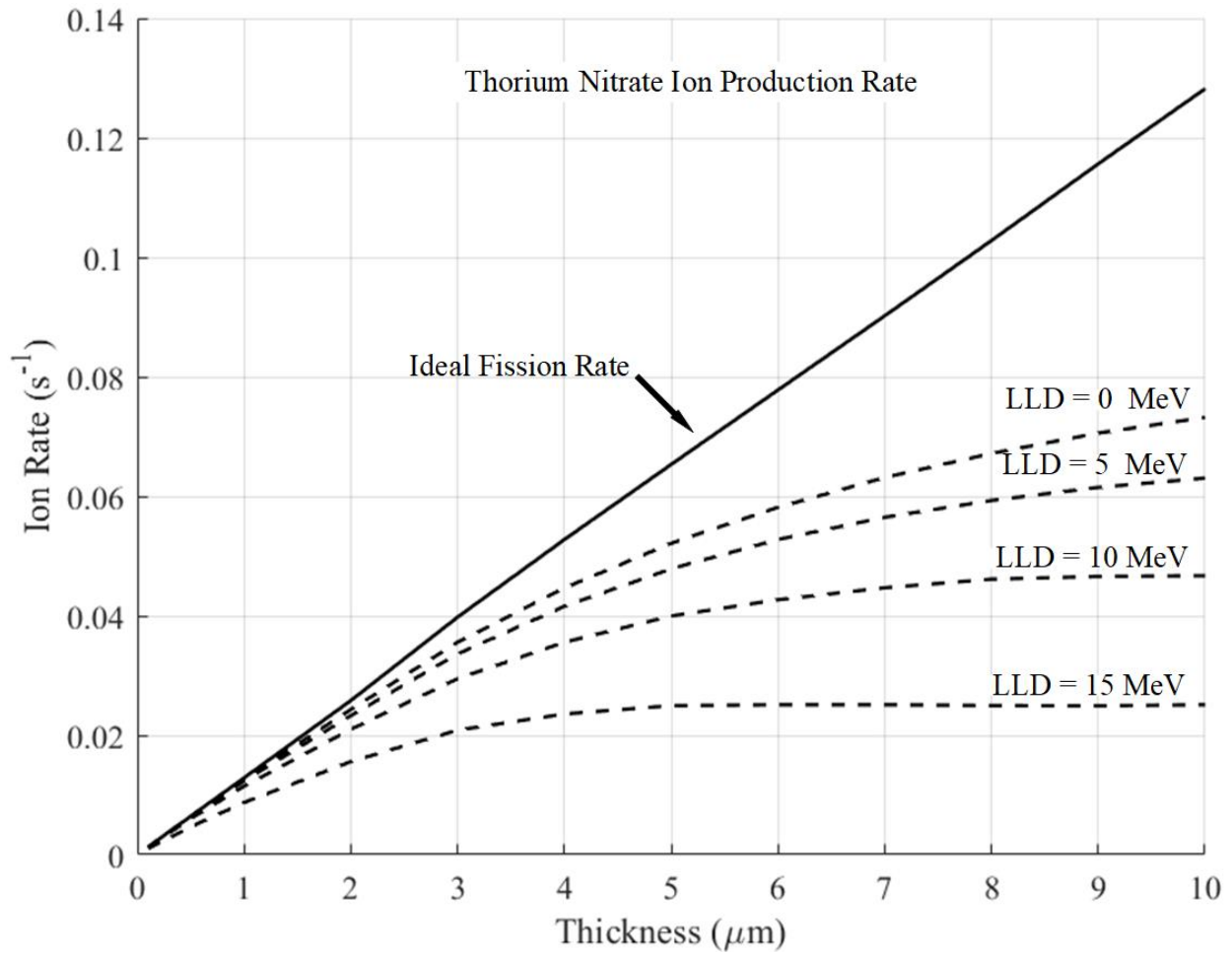


Fig. A-4. Fission fragment crossing rate in thorium nitrate as a comparison of ideal conditions with no self-attenuation and for a variety of energy deposition spectra LLDs.

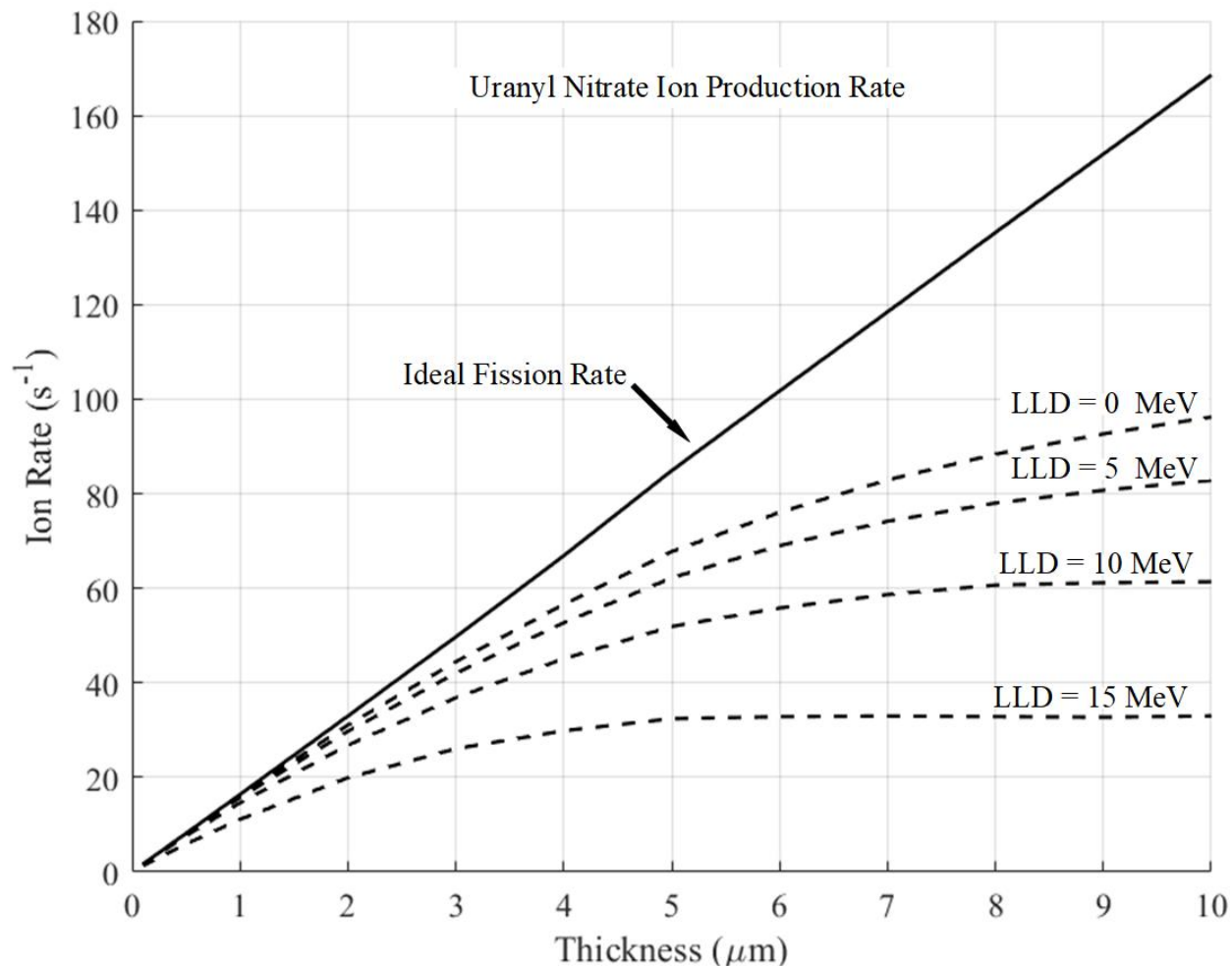


Fig. A-5. Fission fragment crossing rate in uranyl nitrate as a comparison of ideal conditions with no self-attenuation and for a variety of energy deposition spectra LLDs.

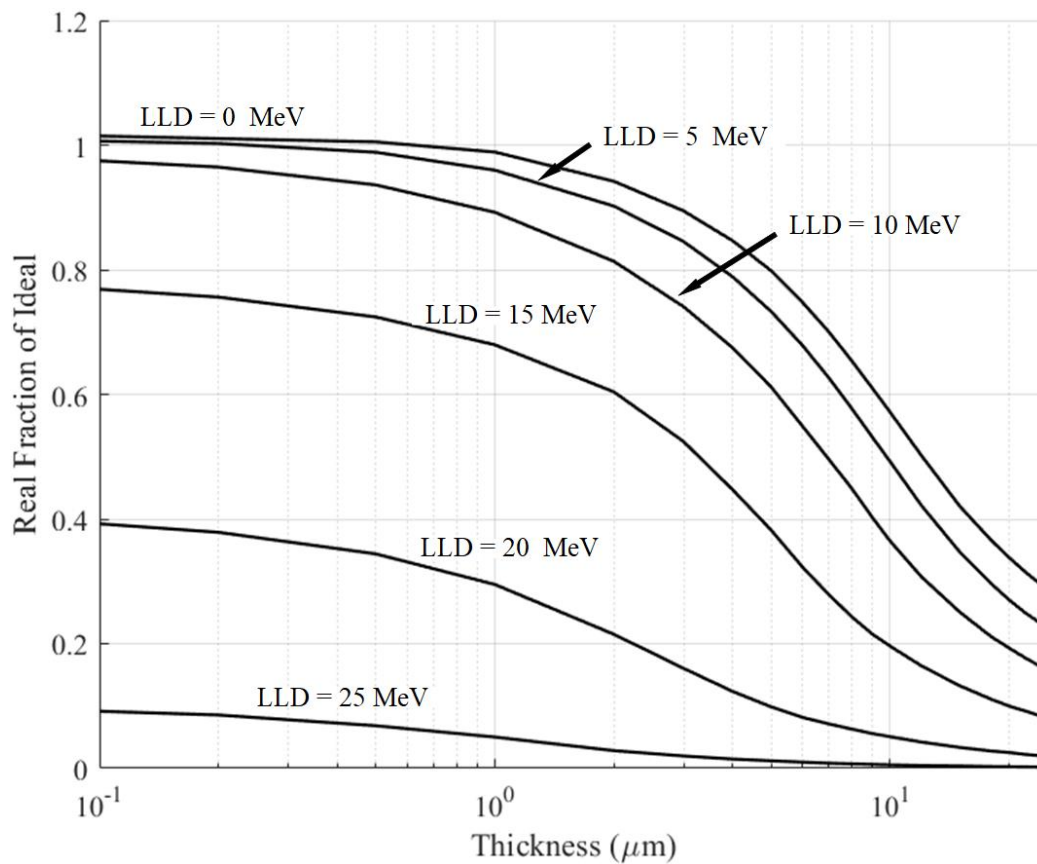


Fig. A-6. Ratio of crossing rate as a function of LLD in comparison to ideal (no self-attenuation). Note, that this plot is applicable for both uranyl nitrate and thorium nitrate.

## **APPENDIX B     PHOTORESIST RECIPE FOR ALUMINA WAFERS**

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NOTE: This recipe is for AZ2070 negative photoresist

### 1) Wafer Preparation:

- a) Spin Rinse Dry; 50, 100, 100 seconds
- b) Bake in vapor prime oven for 20 minutes at 250°C
- c) Let sit for five minutes

### 2) Photolithography Process:

- a) Nitrogen gas is used to blow off any particles on the surface
- b) Deposit volume of 4-5 ml Negative photoresist AZ2070 in the center of wafer with Samco™ Transfer Pipettes
- c) Start Spinner , with two steps settings:
  - i) 1000 rpm for first five seconds
  - ii) Acceleration of 1200 rpm to 3000 rpm and hold for 30 seconds
  - iii) Let sit inside spinner for 60 seconds when run is completed
- d) Soft bake for 3 minutes at 100°C on hot plate
- e) Let sit in for five minutes

### 3) Alignment and Exposure :

- a) Align Mask with wafer
- b) Contact Mode with intensity setting of 325 mJ cm<sup>-2</sup>
- c) Expose time of 15 seconds
- d) Let sit for five minutes
- e) Hard bake at 115°C for 1.5 minutes
- f) Let sit for five minutes

### 4) Photoresist Development:

- a) Wafers are submerged into MIF 300 developer
- b) Continuously fully submerge and remove wafers from solution for approximately 60 submersions to ensure proper development
- c) Move into DI water cascade for a minimal time to quickly wash off MIF300 and transfer to Spin Rinse Dry; 50, 100, 100 seconds
- d) Check under microscope to ensure proper development and desired features are present, if not repeat step 4.b-d

*Notes:* In the event of overdevelopment, large imperfections in features, or any other mistakes. Place undesired wafers into KWIK Strip to strip the photoresist and restart the procedure from the top. Plasma Asher may be required to completely remove any residual photoresist on wafers.

### 5) Surface Clean/Etch for Metallization:

- a) BOE etch for two minutes to ensure clean surface for metal adhesion (200 nm)

- i) Wet the wafers with DI water before submerging into BOE
- b) DI water cascade for eight minutes
- c) Spin Rinse Dry; 300, 200, 300 seconds

6) Metals Deposition:

- a) Releasing vacuum from the Electron Beam Chamber
  - i) Ensure correct metals and respective material data are input for evaporations
  - ii) Ensure the deposition crystal are adequate for prolong depositions
- b) Activate rough pump to initiate first step of pump down
  - i) Allow to pump down before proceeding
- c) Once rough vacuum is achieved, activate high vacuum pump, and allow it to further pump down to  $<5 \times 10^{-6}$  Torr
  - i) Double check the temperature in the chamber to ensure appropriate conditions before proceeding with evaporation, it should be 7-10 Kelvin
- d) Select metal and begin to evaporate with desired thickness
  - i) Titanium for 50 Å
  - ii) Platinum for 500 Å

7) Metal Liftoff:

- a) Place evaporated wafers into KWIK Strip for 1-2 hours with heating element to liftoff photoresist and remove excess metals around desired features
- b) Observe under microscope to confirm successful metallized features

## APPENDIX C ELECTRODEPOSITION SOLUTION PREPARATION

**\*\*Prior to mixing uranyl nitrate solution, ensure the Orion 3 Star pH meter has been calibrated in within 2 months of the date of use.\*\***

**\*\*Confirm Mettler Toledo scale is probably calibrated and balance.\*\***

The following materials are required to properly synthesize the solution:

| Chemical                   | Vendor                                        | Formula                                                                          | Mol wt<br>(g cm <sup>-3</sup> ) | Density<br>(g cm <sup>-3</sup> ) | Qty     | units |
|----------------------------|-----------------------------------------------|----------------------------------------------------------------------------------|---------------------------------|----------------------------------|---------|-------|
| Thorium Nitrate Hydrate    | Strem Chemicals, Inc.                         | Th(NO <sub>3</sub> ) <sub>4</sub> ·(H <sub>2</sub> O) <sub>x</sub>               | 480.06                          | N/A                              | 51.25   | mg    |
| ----- or -----             |                                               |                                                                                  |                                 |                                  |         |       |
| Uranyl nitrate hexahydrate | International Bio-Analytical Industries, Inc. | UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ·(H <sub>2</sub> O) <sub>6</sub> | 502.13                          | 2.807                            | 51.25   | mg    |
| ----- and -----            |                                               |                                                                                  |                                 |                                  |         |       |
| Ammonium nitrate           | Fisher Scientific                             | NH <sub>4</sub> NO <sub>3</sub>                                                  | 80.43                           | 1.725                            | 1801.25 | mg    |
| Deionized water            | -                                             | H <sub>2</sub> O                                                                 | 18.02                           | 0.998                            | 12.5    | mL    |
| Ethanol                    | Fisher Scientific                             | CH <sub>3</sub> CH <sub>2</sub> OH                                               | 46.07                           | 0.805                            | 1.25    | mL    |
| Final Volume               |                                               |                                                                                  |                                 |                                  | 18.75   | mL    |

Glassware necessary to complete this procedure are listed below:

- 25 mL graduated cylinders (qty: 2)
- 1 cc syringe with needle nose tip (qty: 1)
- 50 mL beaker (qty: 1)

1. Label a clean 50 mL beaker with: solution name, experimenter initials, current date
2. Using a small weigh boat, measure out 1801.25 mg of ammonium nitrate
3. Add ammonium nitrate to 50 mL beaker and confirm material deficit using scale
4. Using a small weigh boat, measure out 51.25 mg of uranyl or thorium nitrate
5. Add uranyl or thorium nitrate to 50 mL beaker and confirm material deficit using scale
6. Fill a graduated cylinder with 12.5 mL of deionized water and then add to 50 mL beaker
7. Fill a graduate cylinder with 5 mL of 190 proof ethanol
8. Using the needle nose syringe, extract 2 mL of ethanol from graduate cylinder
9. Add 1.25 mL of ethanol to 50 mL beaker and record material deficit
10. Confirm final solution volume to be 18.75 mL, add deionized water to volume if needed
11. Add miniature magnetic stir rod into 50 mL beaker and set on stir plate
12. Cover beaker with parafilm and proceed to moderately stir mixture (100 rpm) for at least 5 minutes and until solution appears homogenous with no particles
13. Uncover beaker and confirm pH to be between 3.2 – 3.4 using pH probe
14. Recover beaker and continue to stir until solution is needed

## APPENDIX D

## ALPHA SPECTROMETER CALIBRATION

### Ortec Alpha Suite™ Technical Specifications:

Energy Resolution:  $\leq 23$  keV FWHM for  $^{241}\text{Am}$  alpha particles with a detector-to-source spacing equal to the detector diameter

Detector Efficiency:  $\geq 25\%$  is achievable with close detector-to-source spacing.

Background: Above 3 MeV,  $\leq 1$  count/hour based on a BU-020-450-AS detector.

Vacuum range: 1 – 13 Torr

| Source            | Assay Date | Assay Activity (nCi) | Half-Life (y) | A(t) (nCi) |
|-------------------|------------|----------------------|---------------|------------|
| $^{241}\text{Am}$ | 10/1/2003  | 102.9                | 432.7         |            |
| $^{148}\text{Gd}$ | 10/1/2003  | 100                  | 74.6          |            |

#### $^{241}\text{Am}$ Energies:

5388 keV (1.66%)    5442.8 keV (13.1%)    5485.6 keV (84.8%)    5544.5 keV (0.37%)

#### $^{148}\text{Gd}$ Energies:

3182.7 keV (100%)

#### Equation for Sample Tray Locations:

$$\text{distance}(\text{slot}_x) = 4(x - 1),$$

Example:  $\text{slot}_2 \approx 4$  mm ,  $\text{slot}_7 \approx 24$  mm

$$\text{Solid angle}(\text{slot}_x) = .5 \left( 1 - \frac{4(x-1)}{\sqrt{4(x-1)^2 + 12^2}} \right),$$

Example:  $\text{slot}_2 \approx 0.34$ ,  $\text{slot}_7 \approx 0.05$

### Energy and Efficiency Calibration Procedure:

1. Insert uncollimated  $^{241}\text{Am}$  source into first alpha suite chamber
2. Set sample tray to slot 7 (24 mm to detector)
3. Set vacuum pressure to 1500 mTorr
4. Apply bias and run measurement for 10 minutes to obtain a well-defined spectrum
  - a. Confirm energy resolution of the 5485.6 keV peak  $\leq 23$  keV FWHM and record
  - b. Save ASCII file of spectrum
5. Set the MCA calibration for all visible alpha peaks
6. Turn off bias and vent chamber
7. Repeat steps (1) – (7) for uncollimated  $^{148}\text{Gd}$  source
8. Turn off bias and vent chamber
9. Return  $^{241}\text{Am}$  source to chamber
10. Adjust sample tray to slot 2 (4 mm to detector)
11. Set vacuum pressure to 1500 mTorr
12. Apply bias and run measurement for 5 minutes
  - a. Set ROI to all channels  $\geq 250$
  - b. Confirm detection efficiency  $\geq 25\%$  (corrected for solid angle)

- c. Save ASCII file of spectrum
13. Turn off bias, vent chamber, and remove source
14. Repeat steps (1) – (12) for all 16 chambers
15. Turn off bias, vent chamber, and remove source
16. Set vacuum pressure to 1500 mTorr
17. Apply bias and run a background measurement for all chambers simultaneously for a duration of at least 48 hours
  - a. Record number of counts in ROI
  - b. Save ASCII file of spectrum
18. Turn off the vacuum pump and both alpha suite systems when not in use
19. Initial and date calibration sheet

**Example of Completed Calibration:**

| Chamber # | FWHM (keV) | Efficiency (%) | 48-hr BKG counts | Initial | Date     |
|-----------|------------|----------------|------------------|---------|----------|
| 1         | 15.7       | 61.56          | 16               | DMN     | 1/2/2018 |
| 2         | 17.6       | 62.34          | 4                | DMN     | 1/2/2018 |
| 3         | 17.5       | 61.37          | 7                | DMN     | 1/2/2018 |
| 4         | 16.58      | 62.70          | 3                | DMN     | 1/2/2018 |
| 5         | 17.12      | 63.47          | 2                | DMN     | 1/2/2018 |
| 6         | 18.71      | 63.47          | 4                | DMN     | 1/2/2018 |
| 7         | 16.56      | 62.81          | 6                | DMN     | 1/2/2018 |
| 8         | 18.70      | 63.66          | 4                | DMN     | 1/2/2018 |
| 9         | 17.41      | 63.31          | 7                | DMN     | 1/2/2018 |
| 10        | 15.44      | 62.51          | 2                | DMN     | 1/2/2018 |
| 11        | 14.44      | 62.15          | 4                | DMN     | 1/2/2018 |
| 12        | 17.24      | 62.62          | 4                | DMN     | 1/2/2018 |
| 13        | 14.86      | 63.82          | 7                | DMN     | 1/2/2018 |
| 14        | 16.23      | 64.16          | 6                | DMN     | 1/2/2018 |
| 15        | 12.9       | 64.82          | 1                | DMN     | 1/2/2018 |
| 16        | 15.44      | 62.97          | 10               | DMN     | 1/2/2018 |

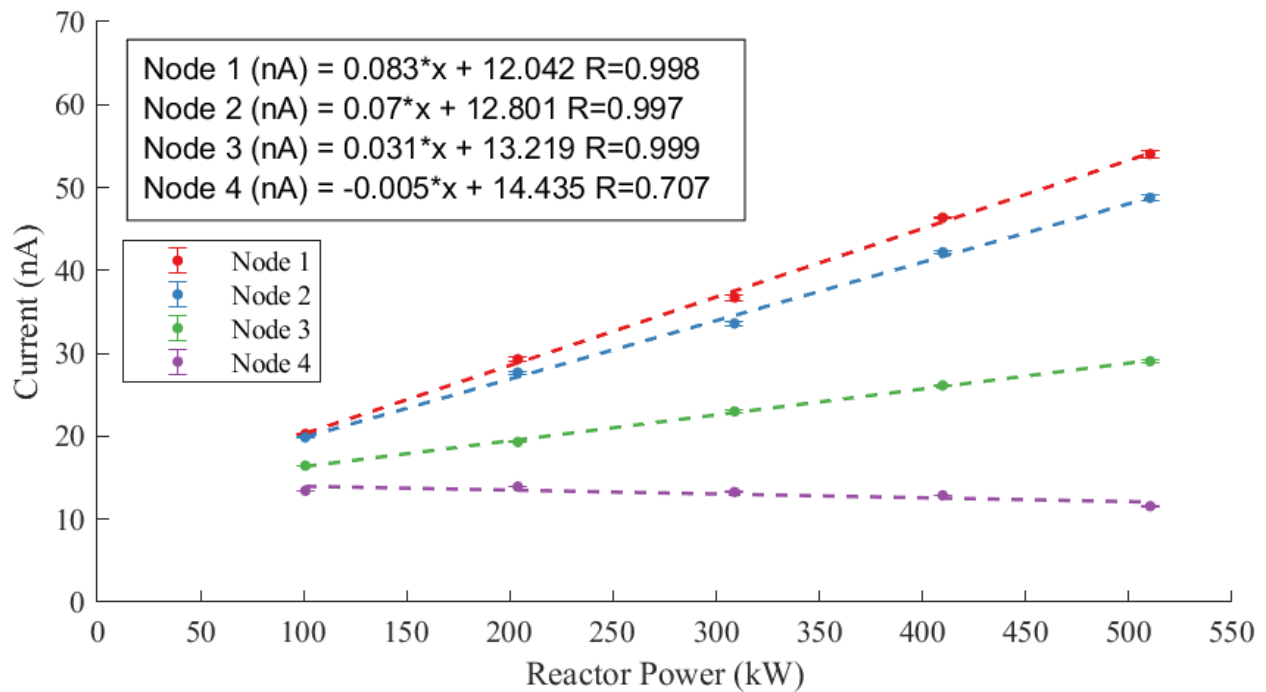


Fig. E-1. Current mode linearity for MPFD array 1.

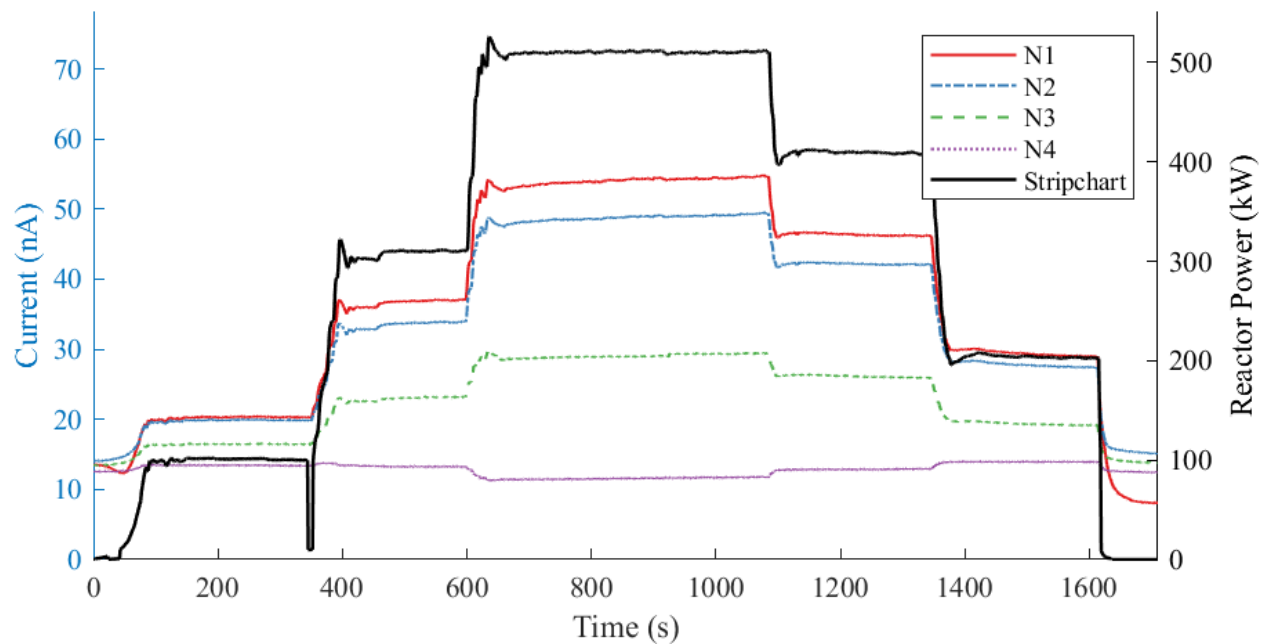


Fig. E-2. Current mode DRT plot of MPFD array 1.

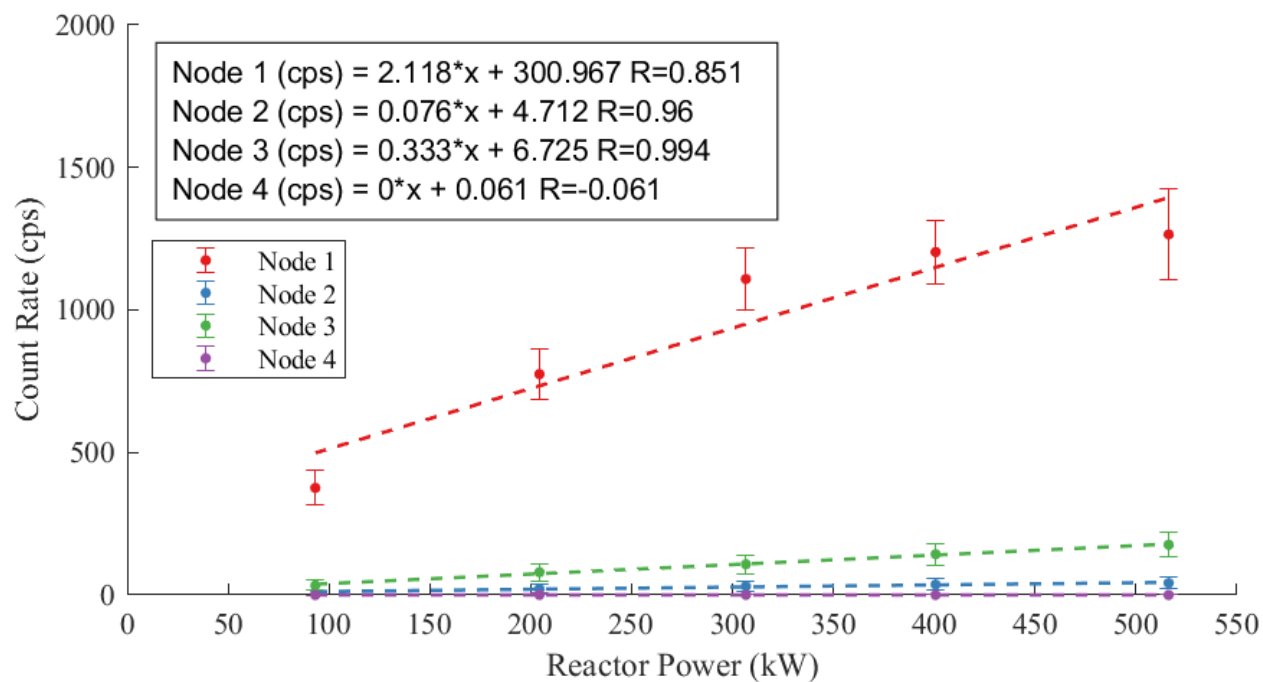


Fig. E-3. Pulse mode linearity for MPFD array 1.

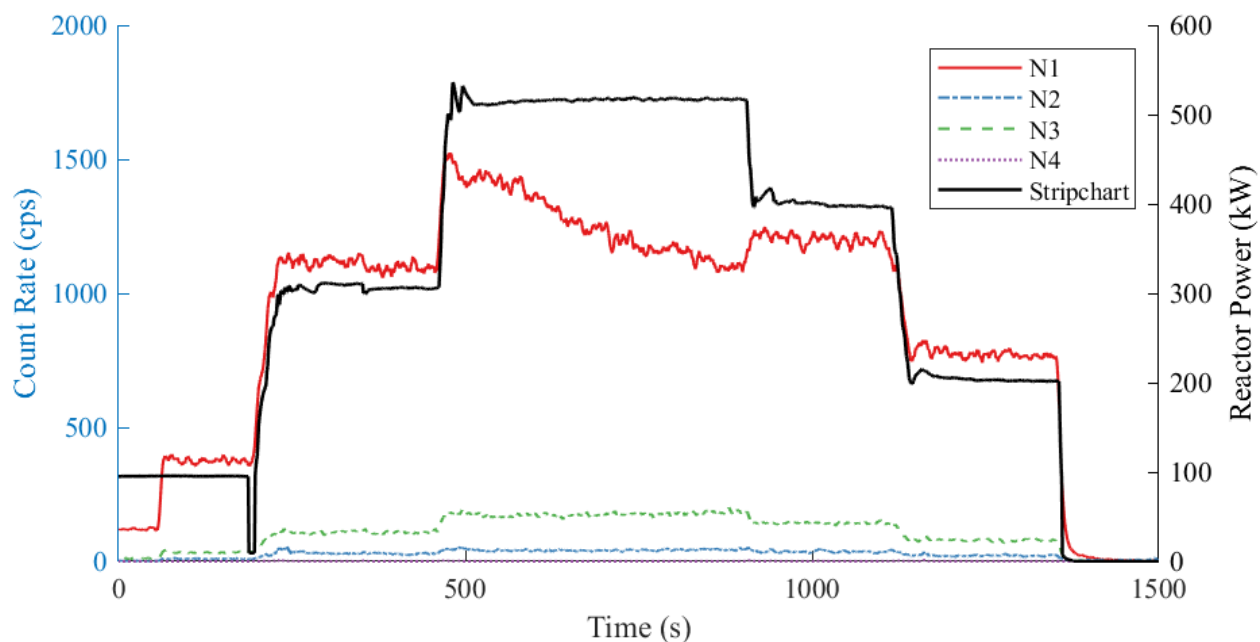


Fig. E-4. Pulse mode DRT plot of MPFD array 1.

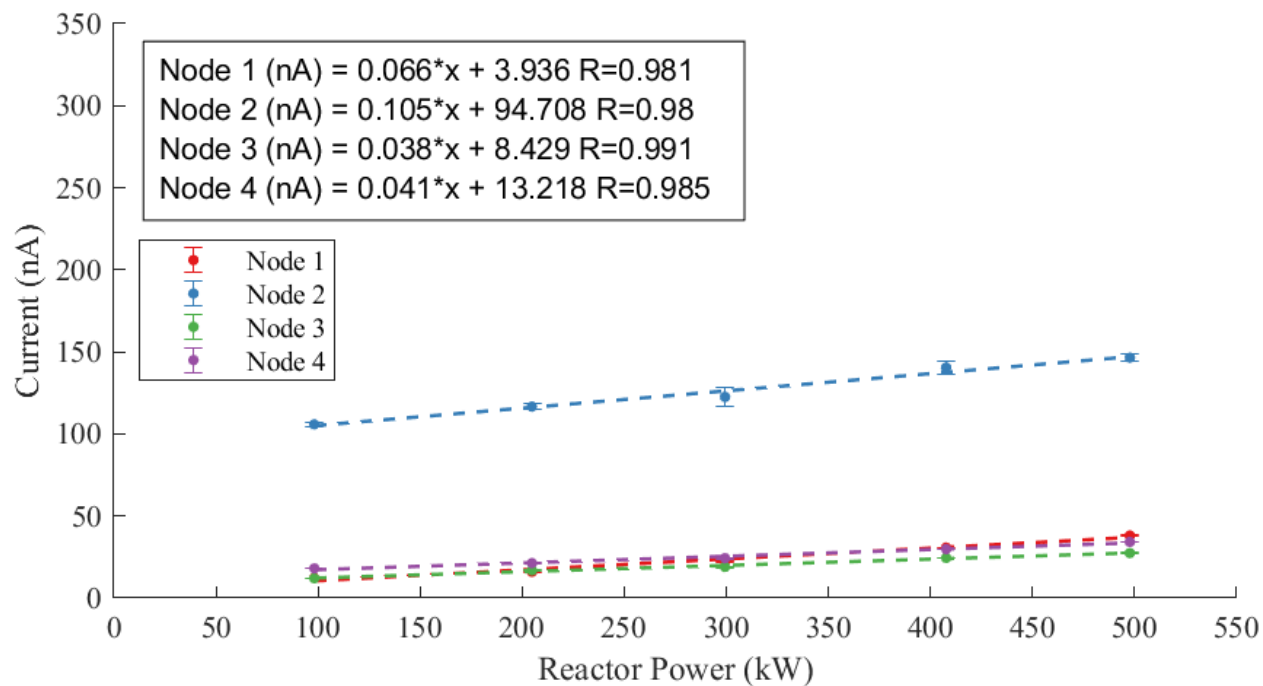


Fig. E-5. Current mode linearity for MPFD array 2.

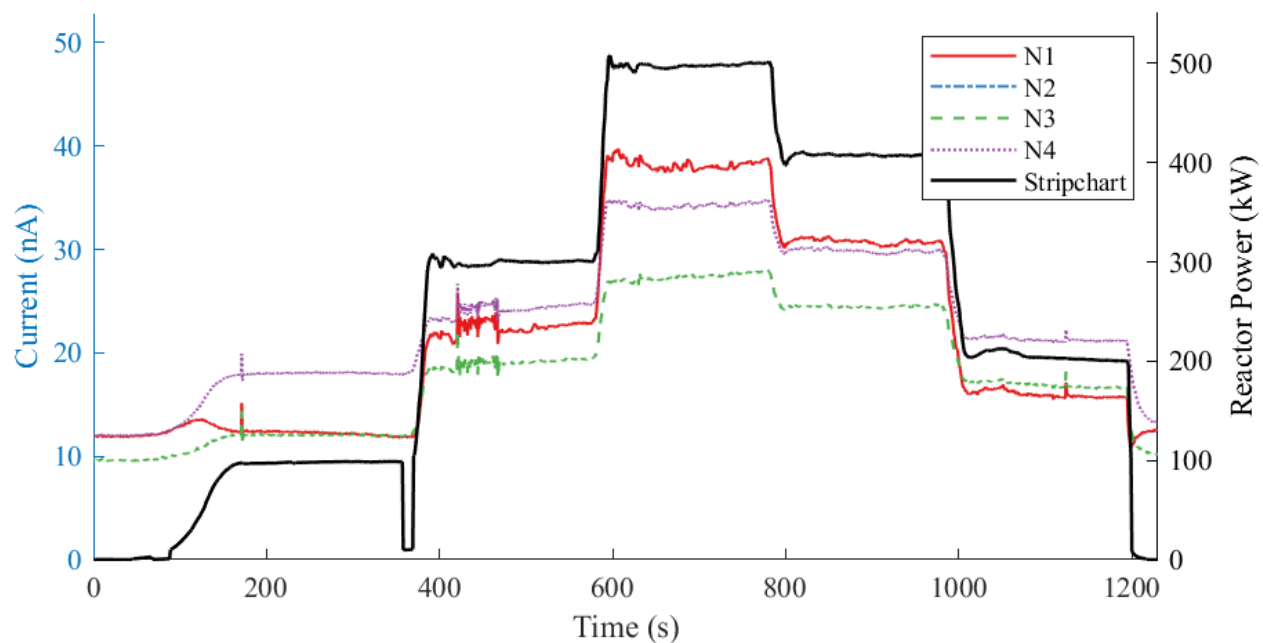


Fig. E-6. Current mode DRT plot of MPFD array 2.

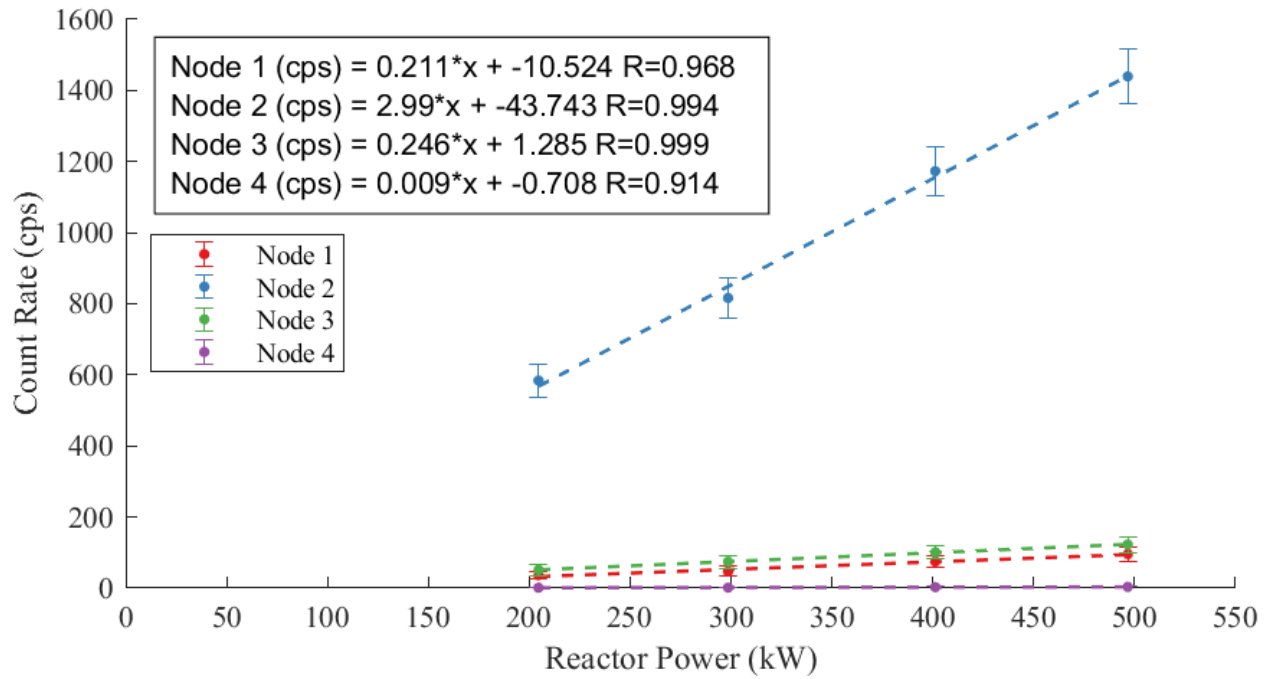


Fig. E-7. Pulse mode linearity for MPFD array 2.

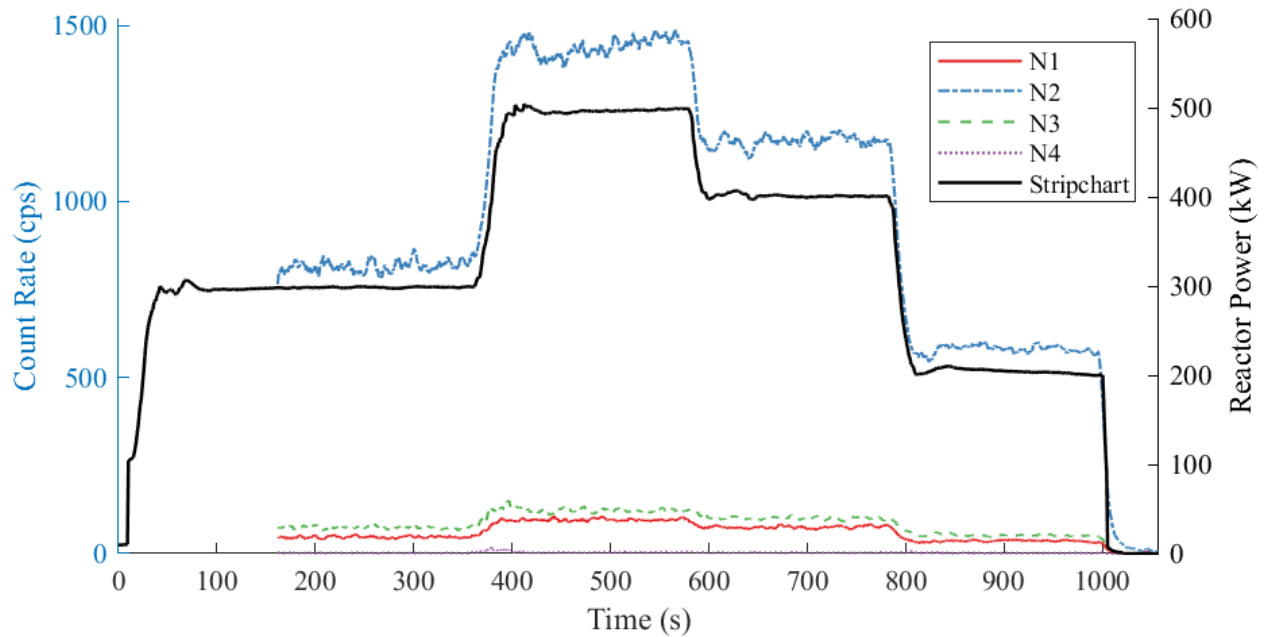


Fig. E-8. Pulse mode DRT plot of MPFD array 2.

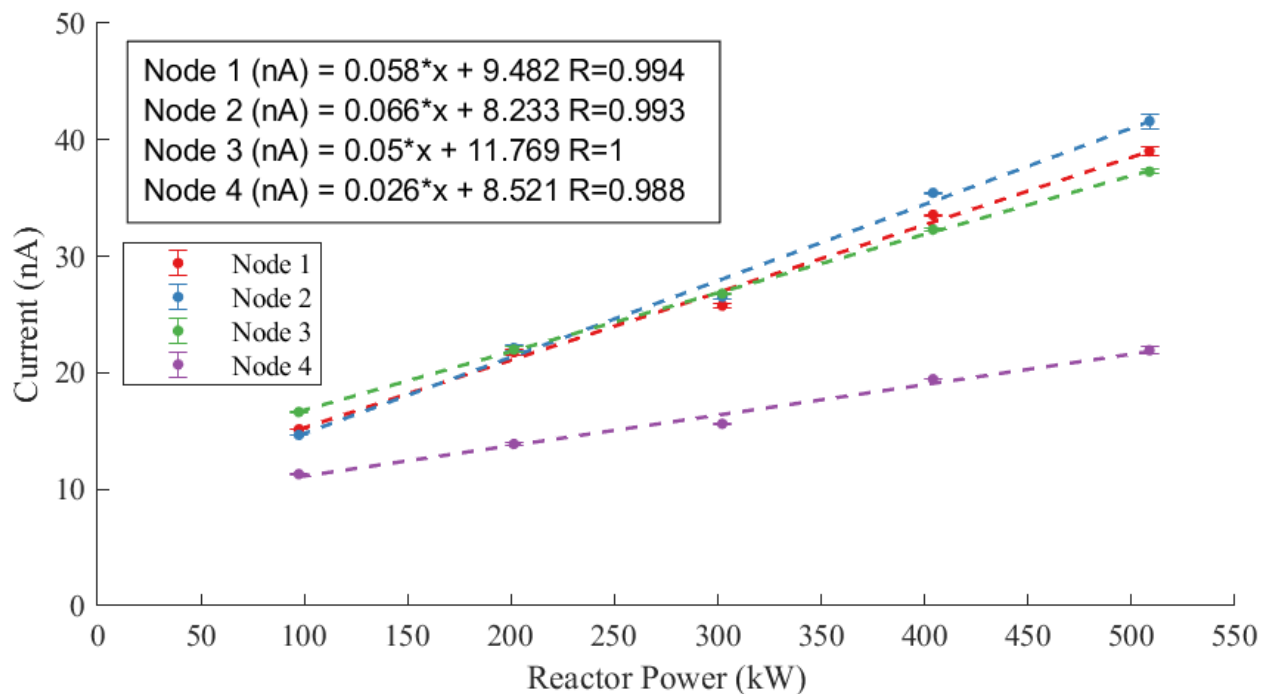


Fig. E-9. Current mode linearity for MPFD array 3.

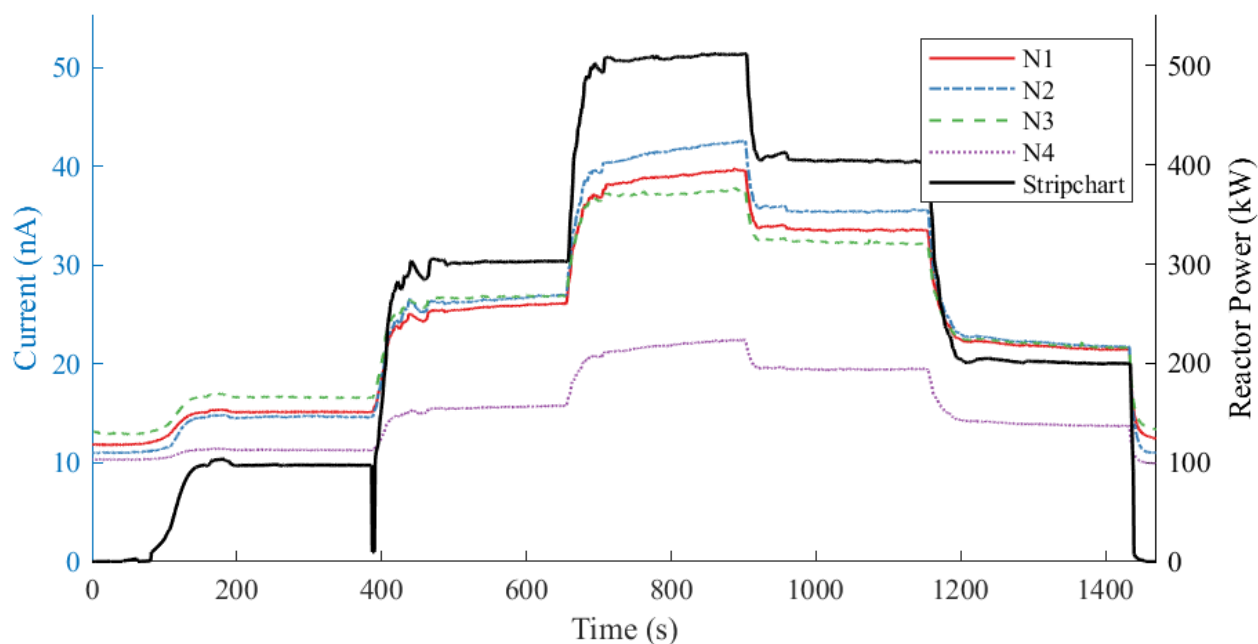


Fig. E-10. Current mode DRT plot of MPFD array 3.

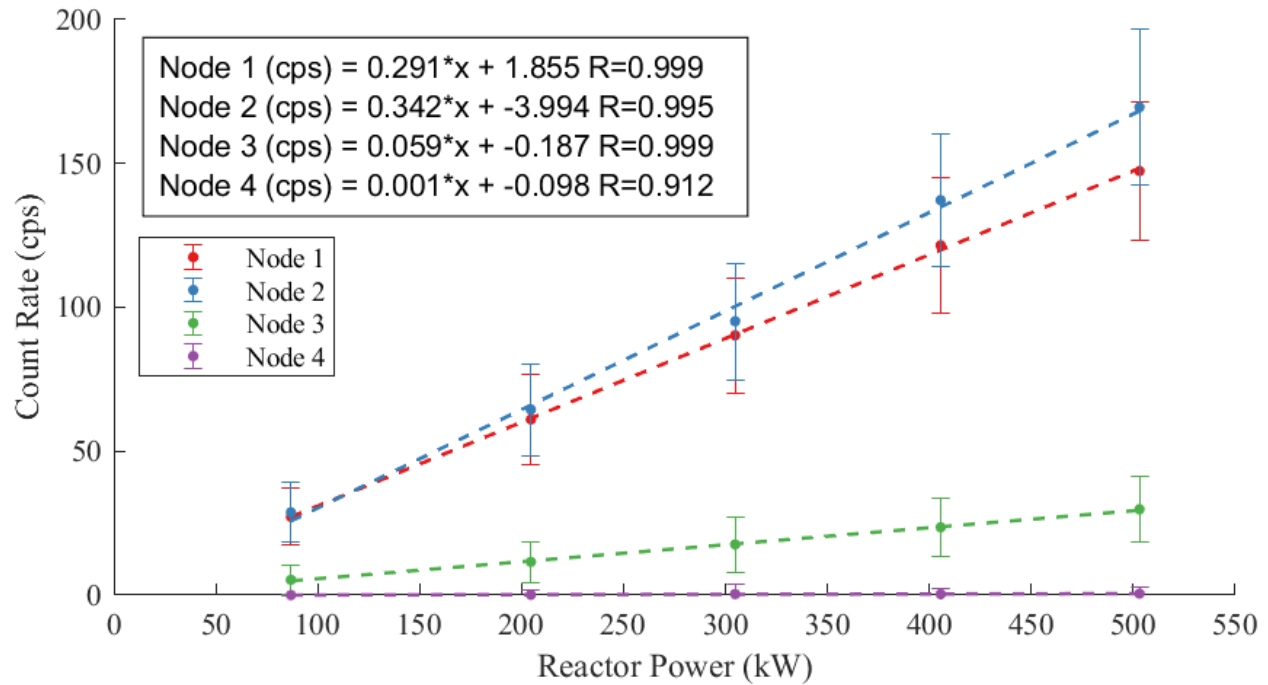


Fig. E-11. Pulse mode linearity for MPFD array 3.

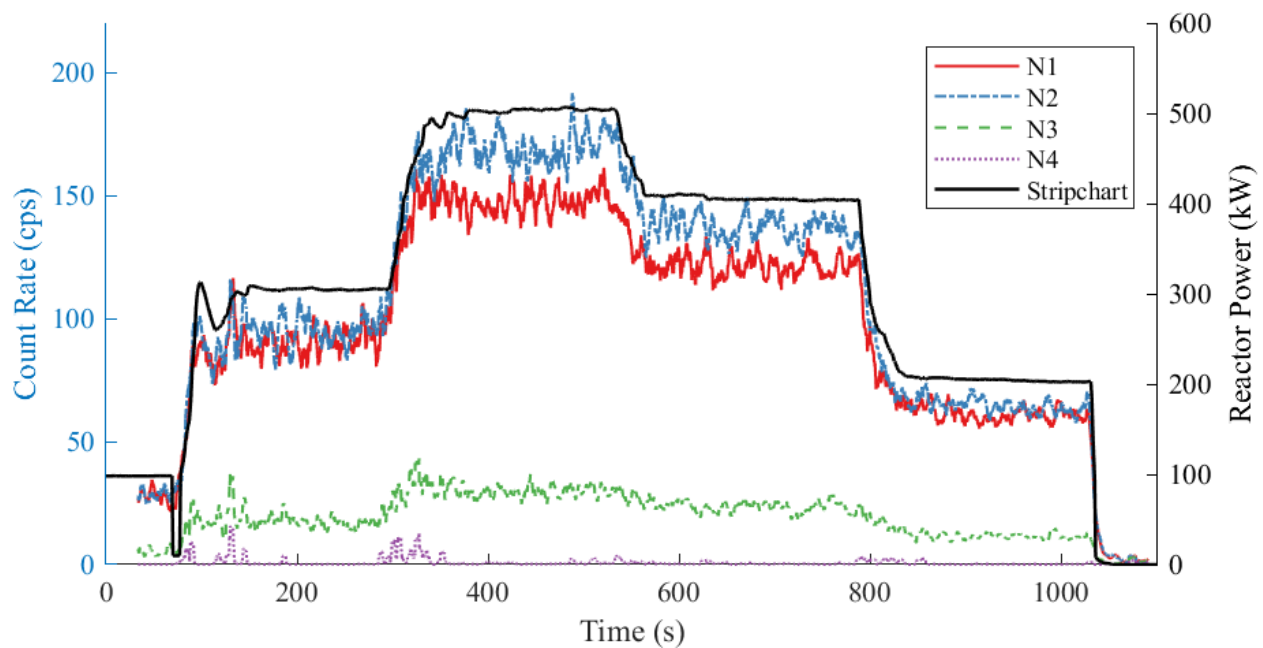


Fig. E-12. Pulse mode DRT plot of MPFD array 3.

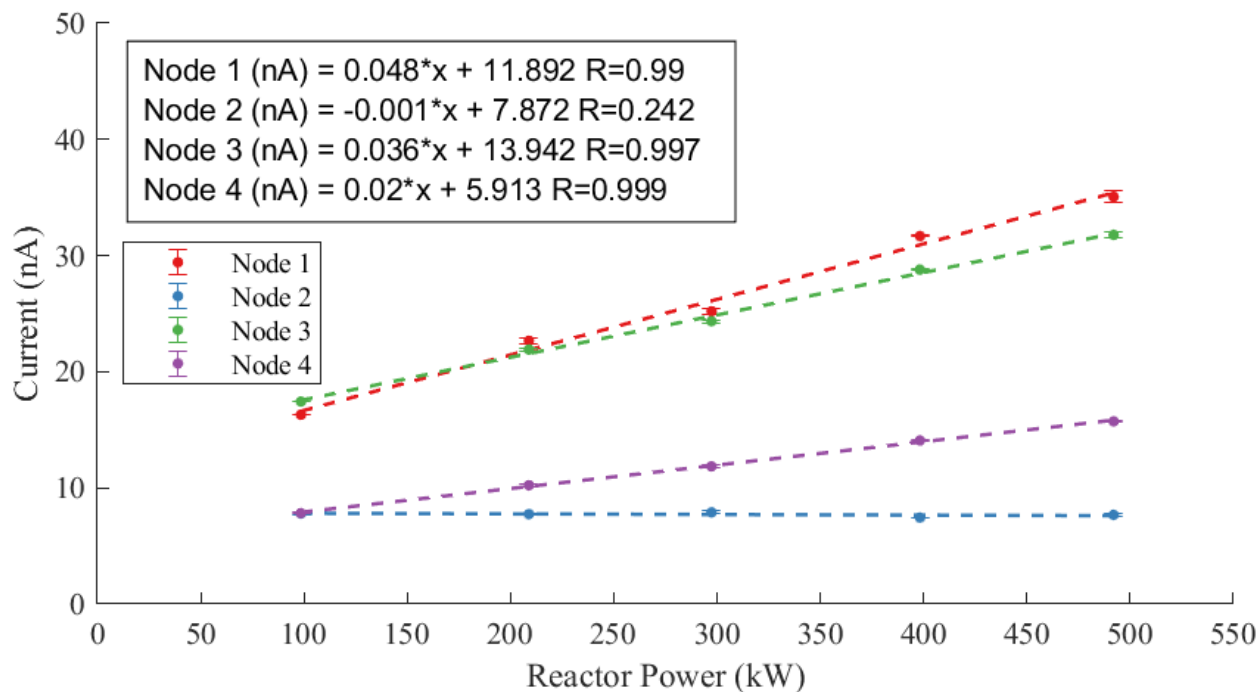


Fig. E-13. Current mode linearity for MPFD array 4.

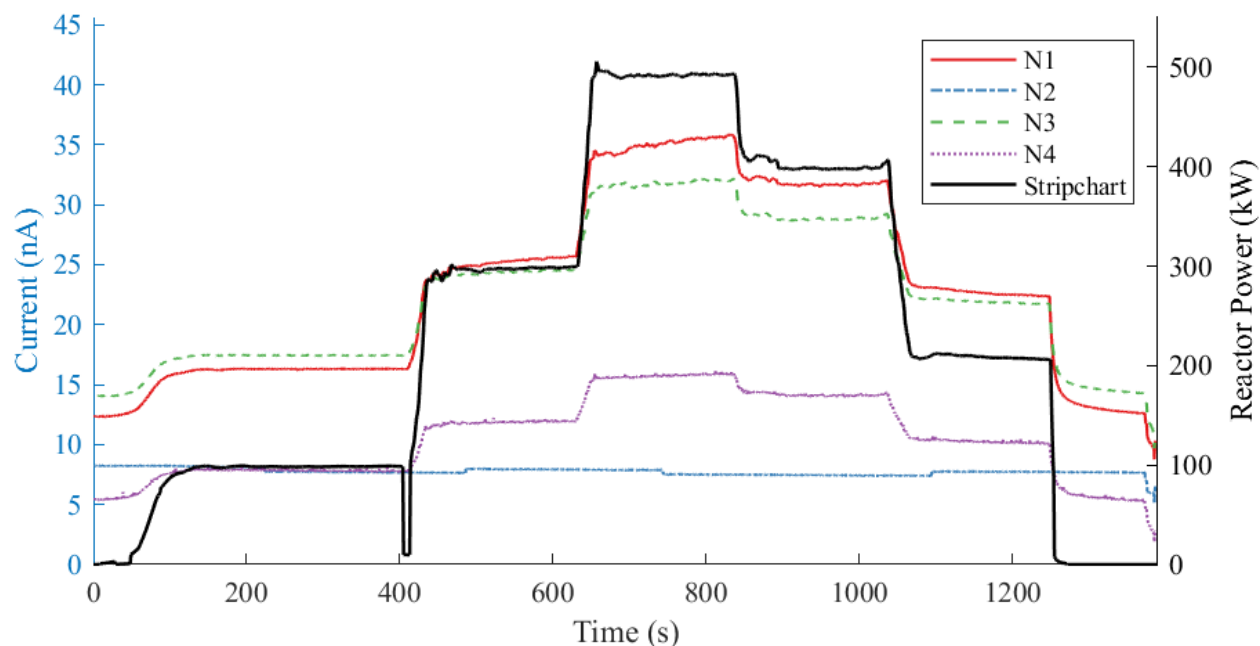


Fig. E-14. Current mode DRT plot of MPFD array 4.

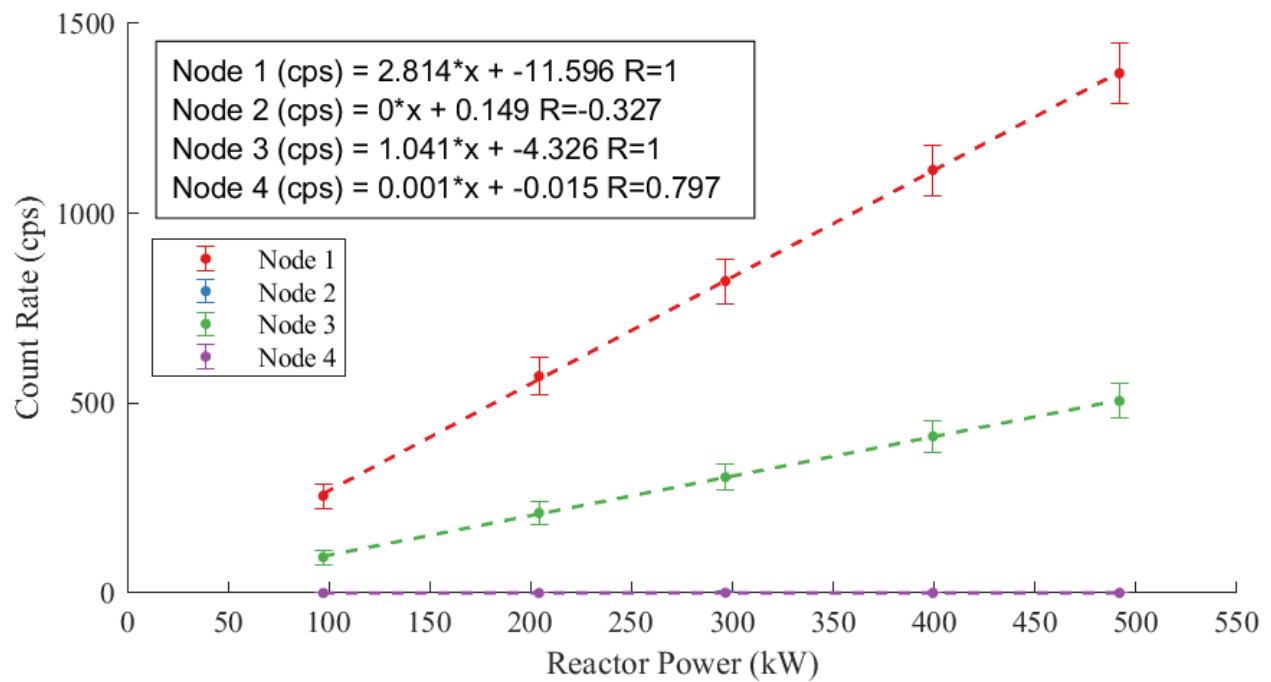


Fig. E-15. Pulse mode linearity for MPFD array 4.

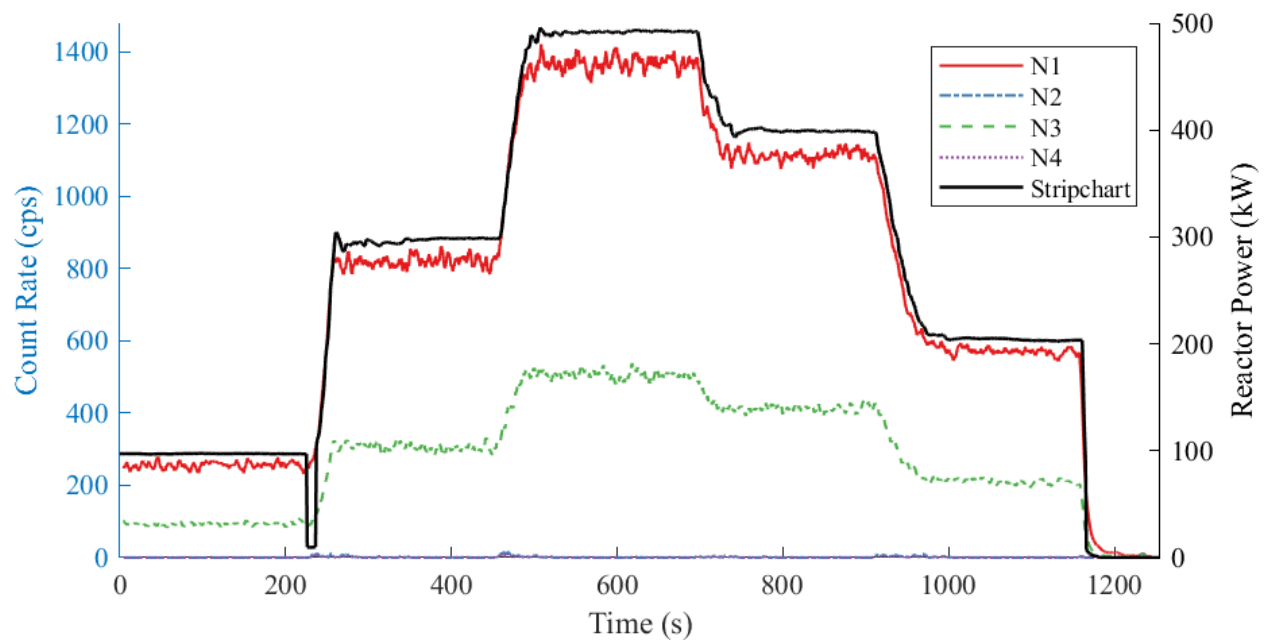


Fig. E-16. Pulse mode DRT plot of MPFD array 4.

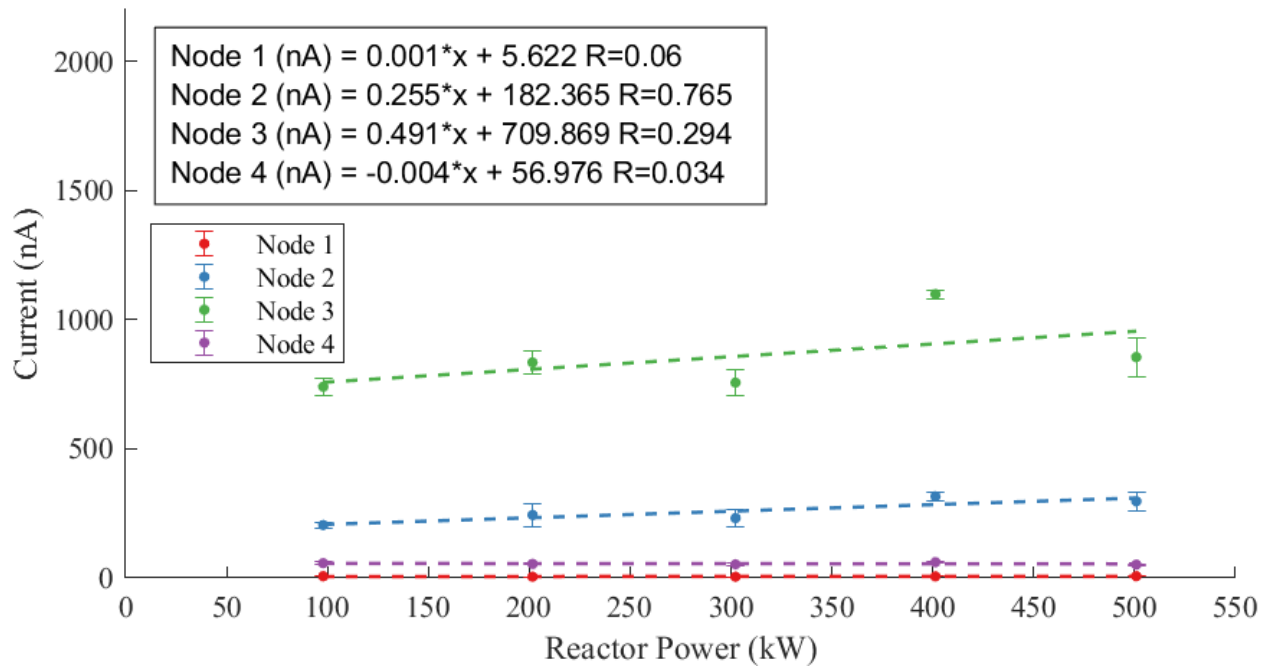


Fig. E-17. Current mode linearity for MPFD array 5.

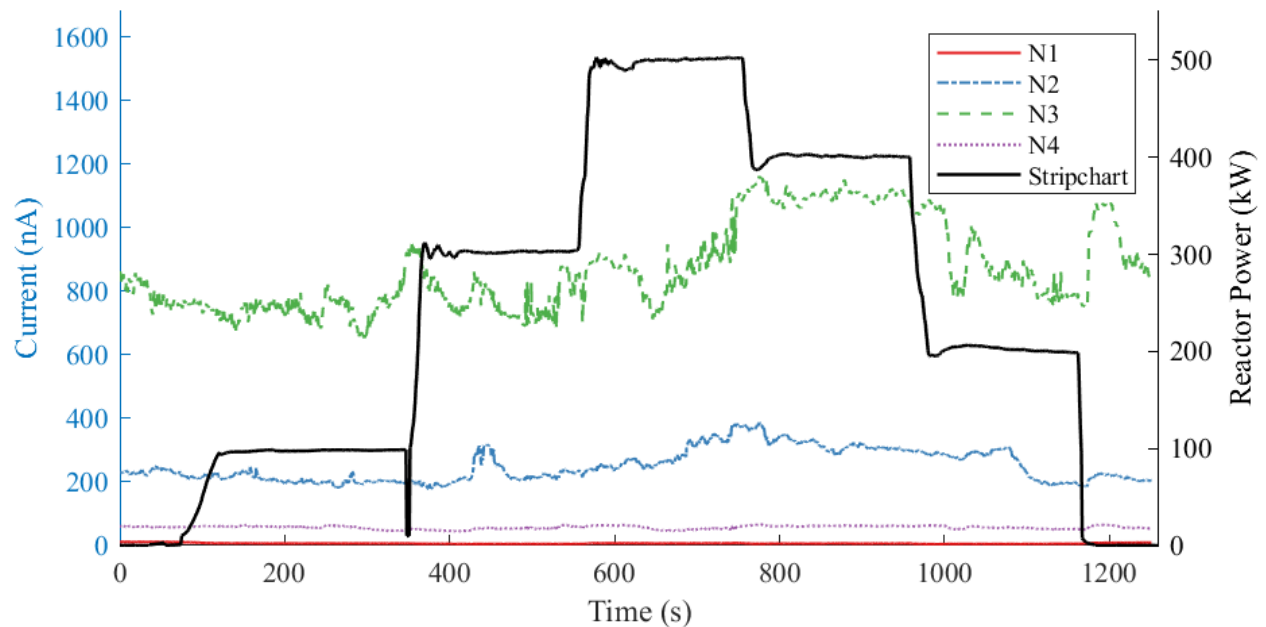


Fig. E-18. Current mode DRT plot of MPFD array 5.

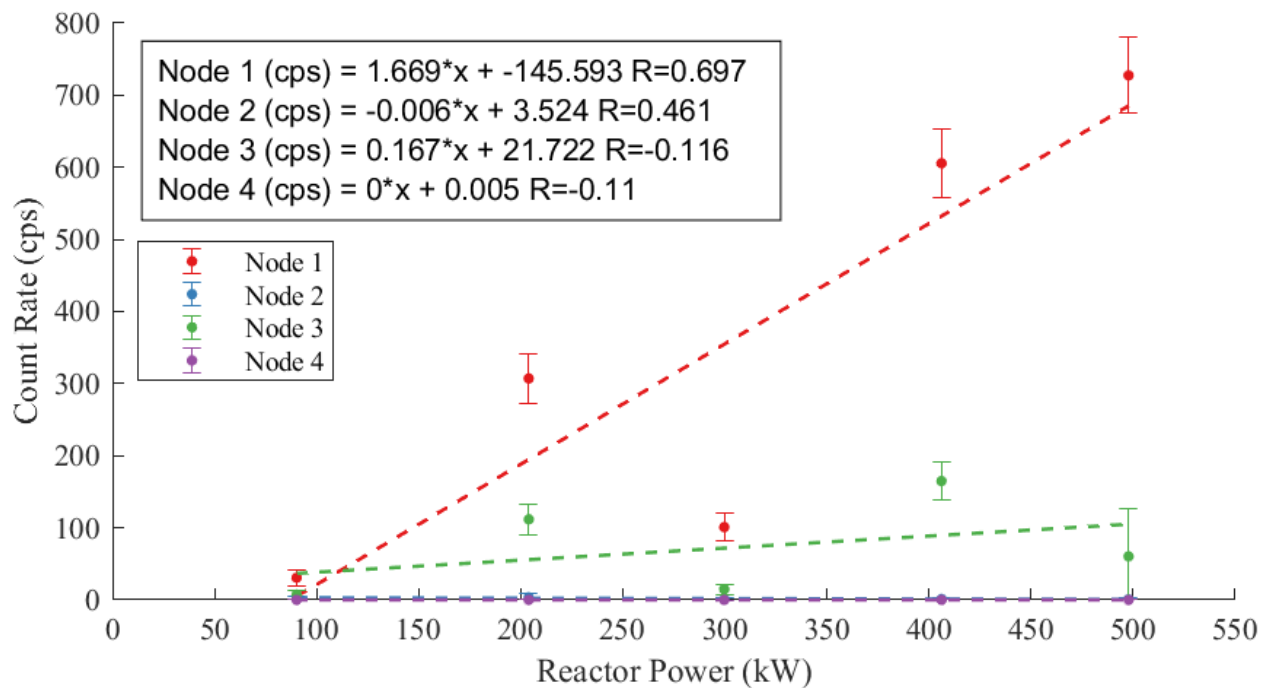


Fig. E-19. Pulse mode linearity for MPFD array 5.

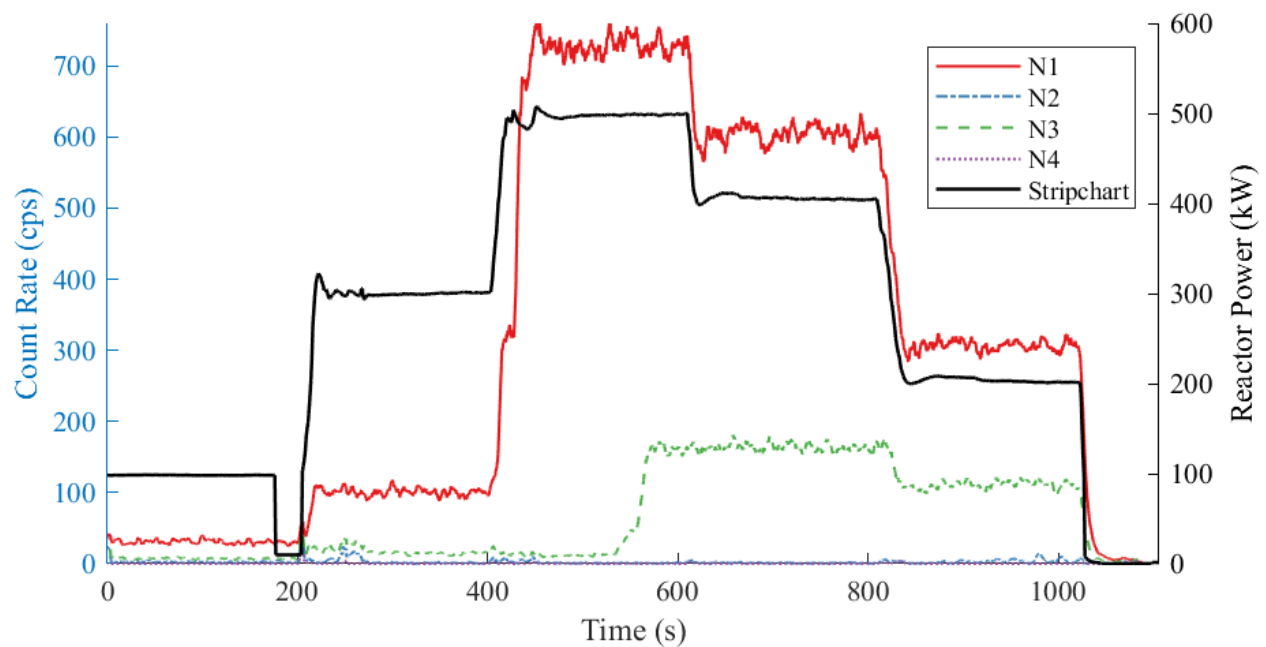


Fig. E-20. Pulse mode DRT plot of MPFD array 5.

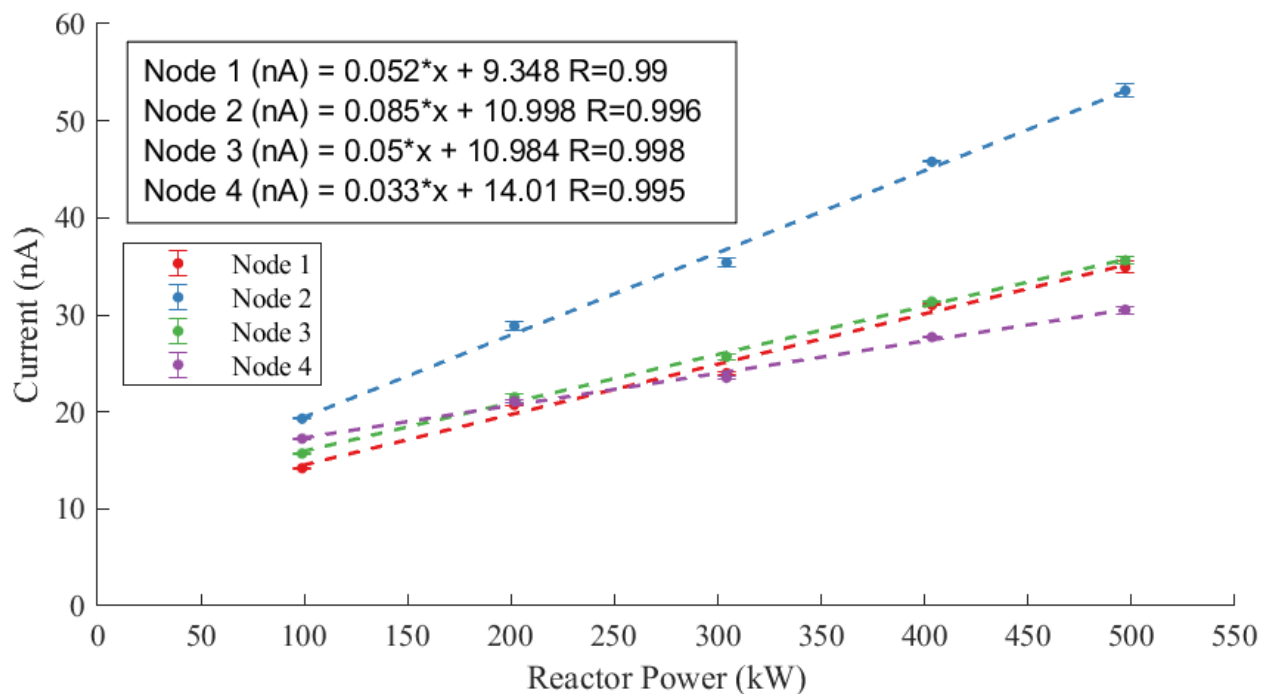


Fig. E-21. Current mode linearity for MPFD array 6.

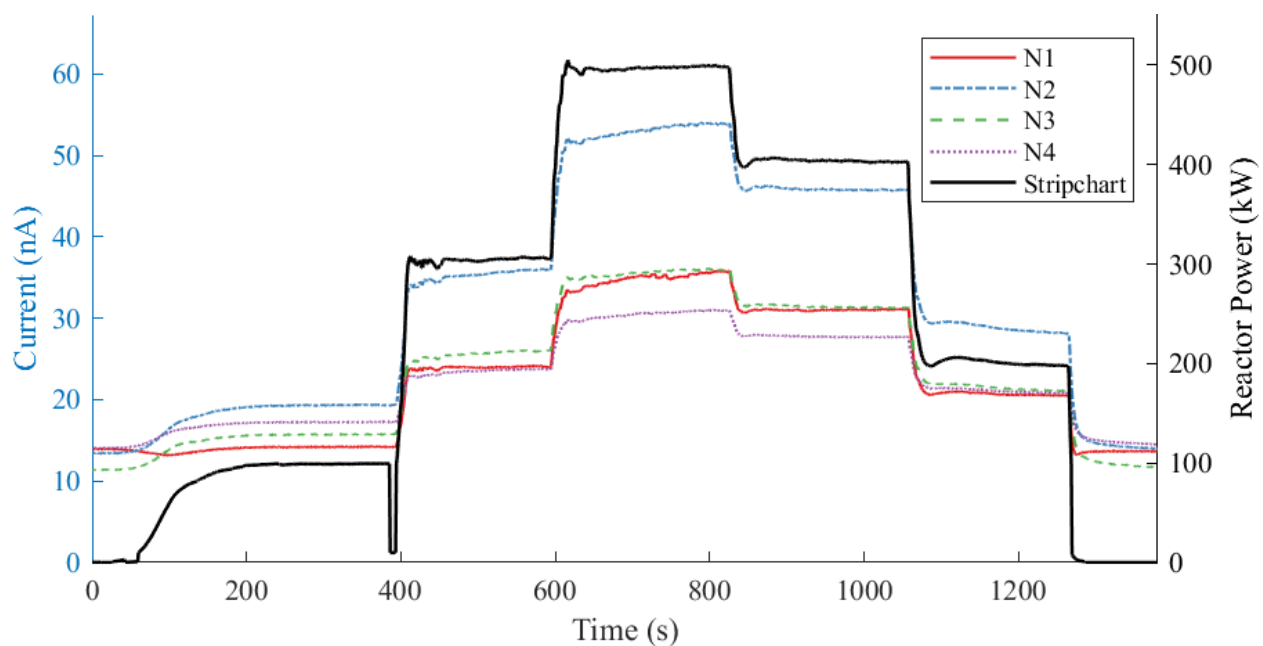


Fig. E-22. Current mode DRT plot of MPFD array 6.

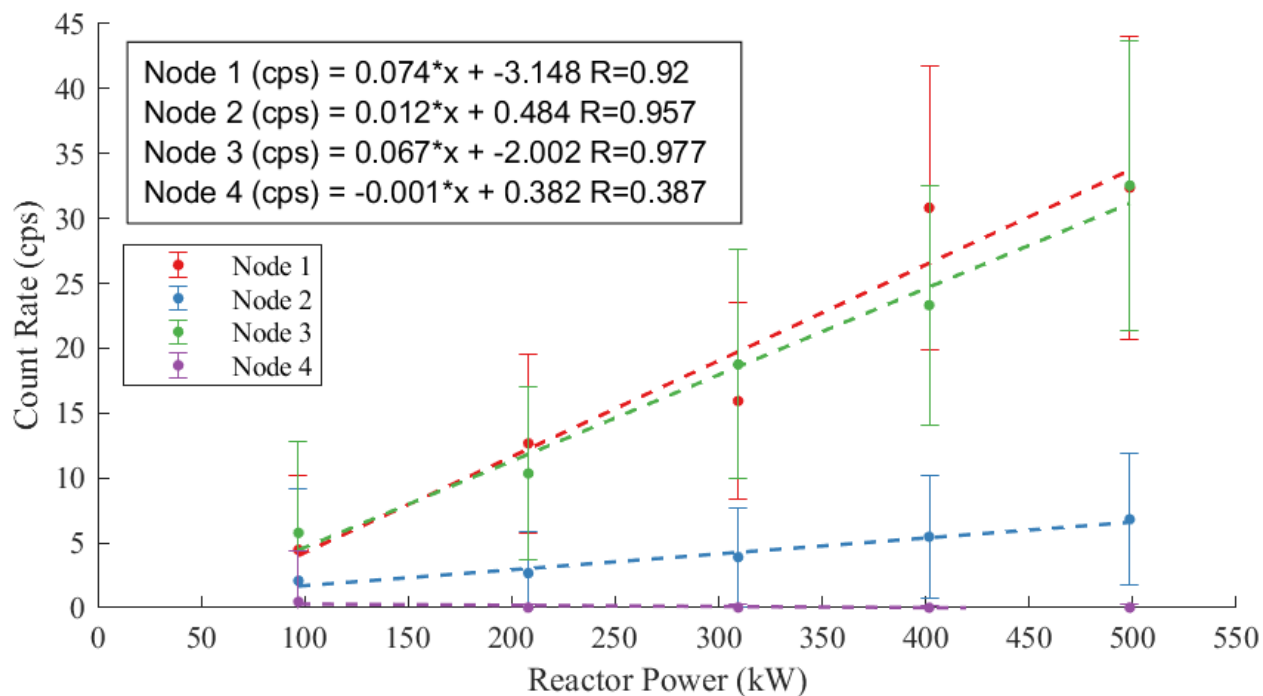


Fig. E-23. Pulse mode linearity for MPFD array 6.

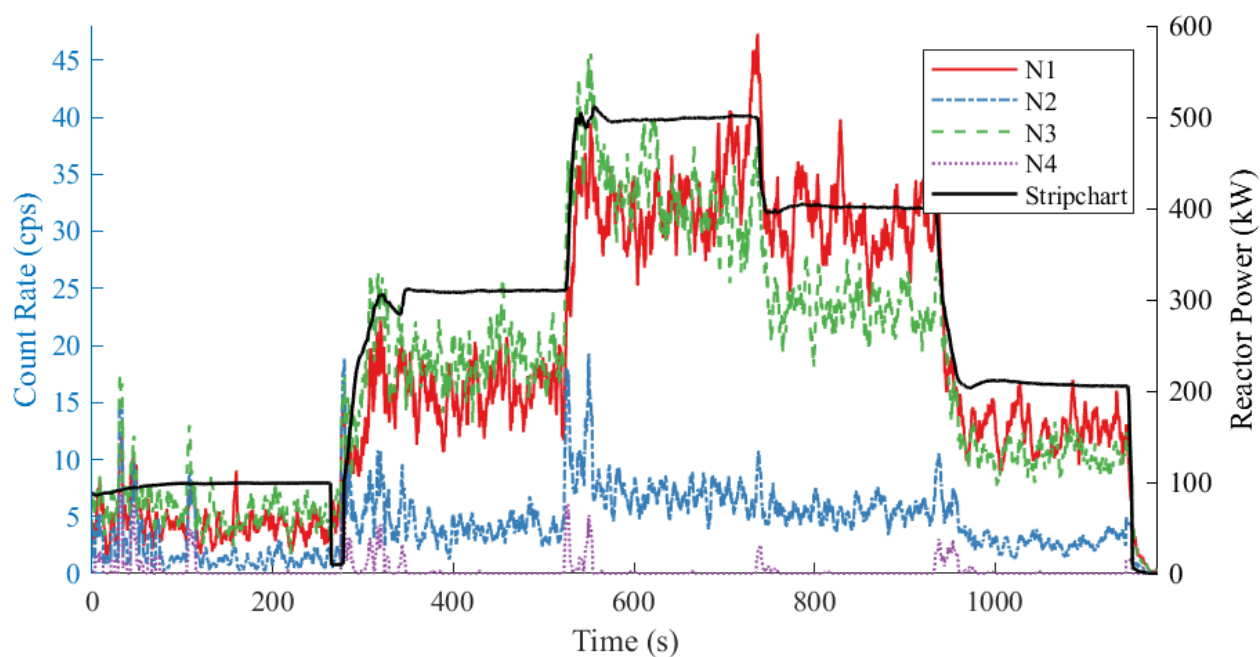


Fig. E-24. Pulse mode DRT plot of MPFD array 6.

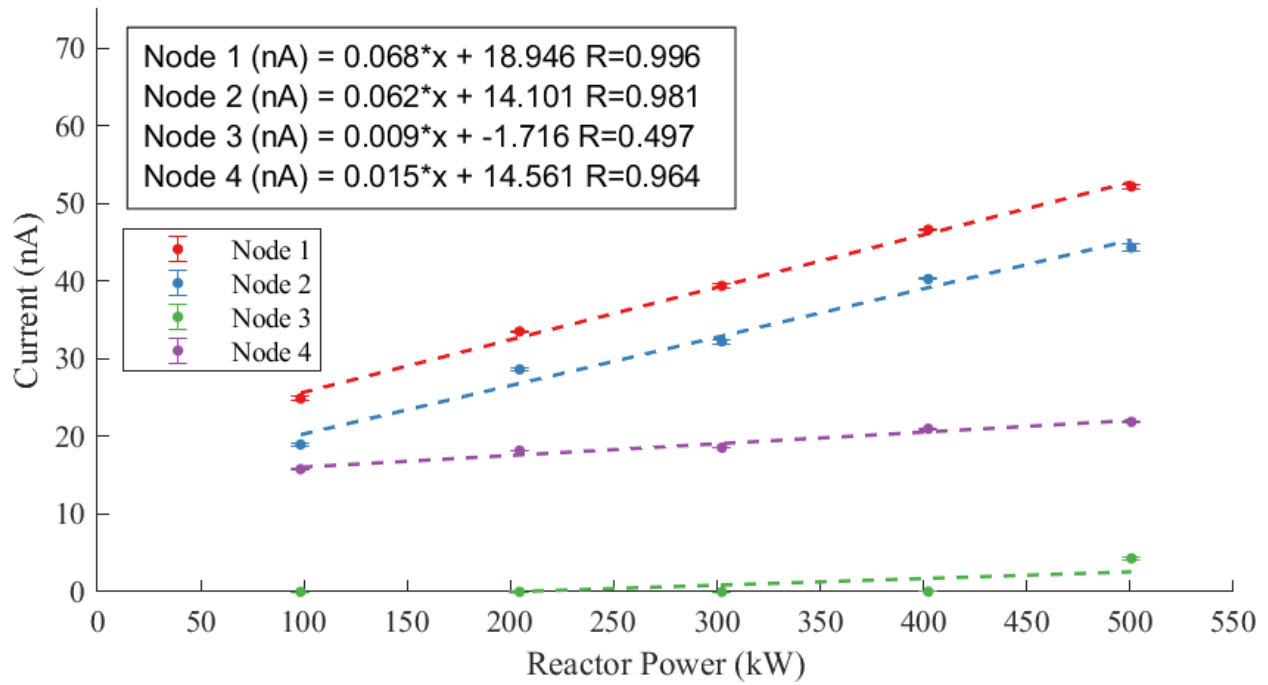


**Fig. E-25. Current mode linearity for MPFD array 7.**

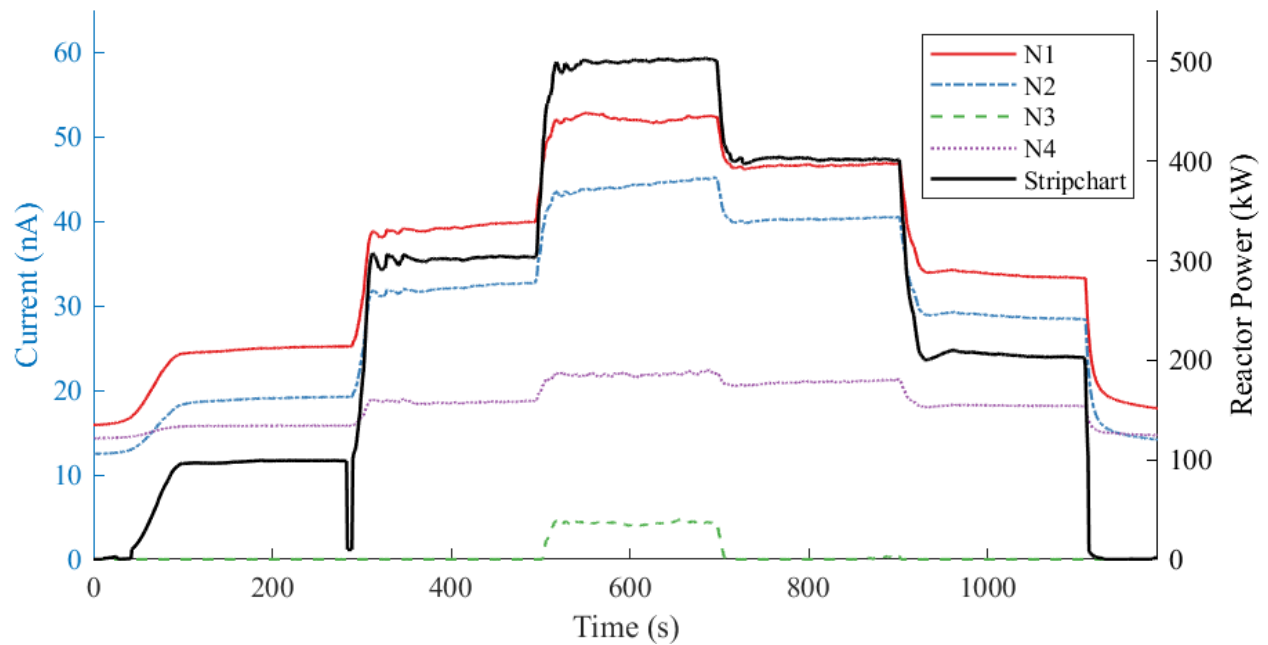


**Fig. E-26. Current mode DRT plot of MPFD array 7.**

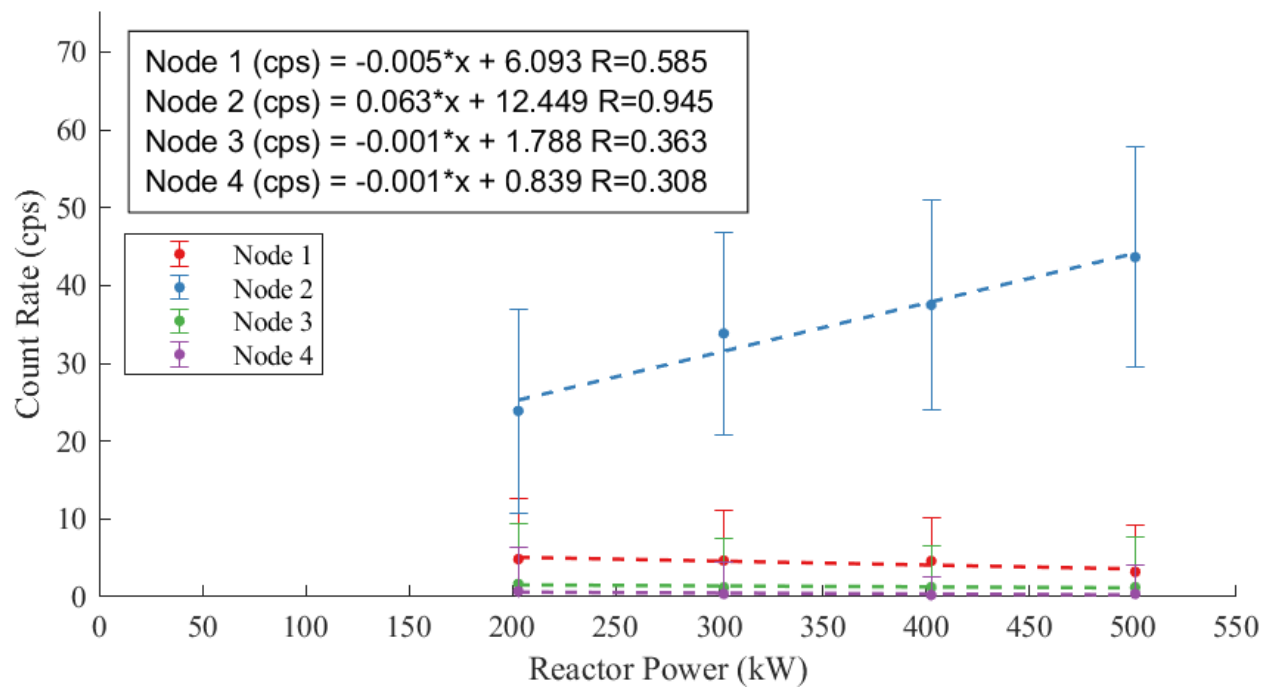


**Fig. E-27. Pulse mode linearity for MPFD array 7.**

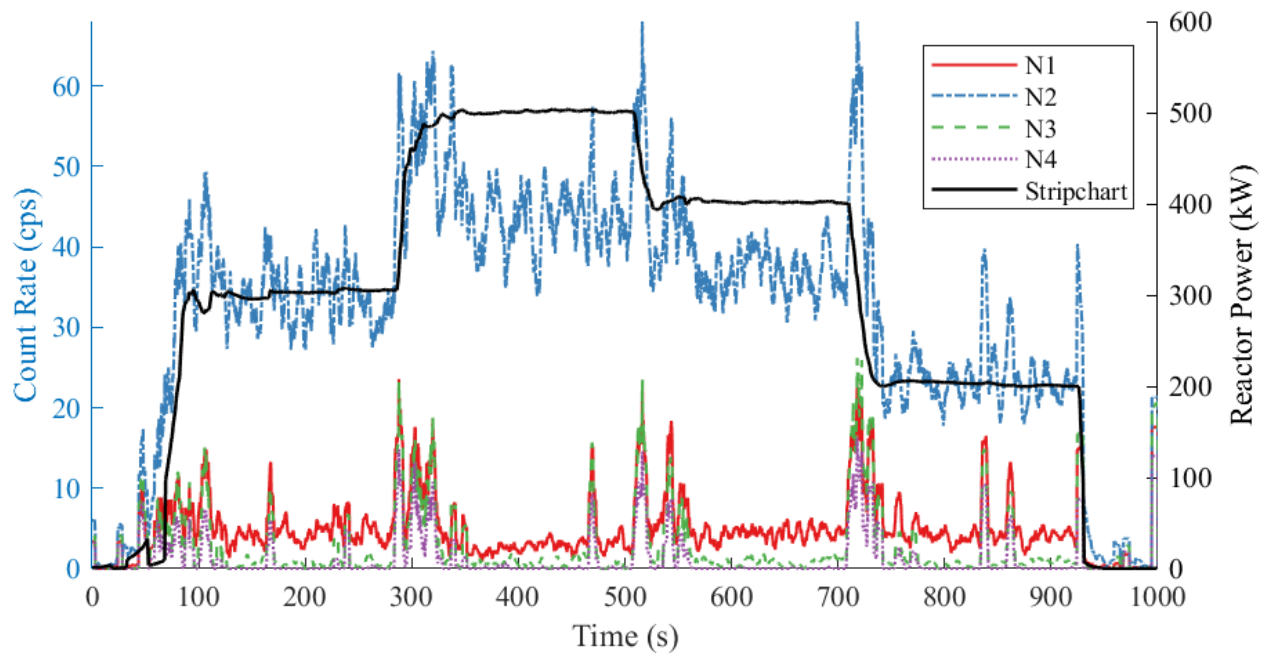


**Fig. E-28. Pulse mode DRT plot of MPFD array 7.**

## APPENDIX F

## UWNR PULSE FWHM DATA

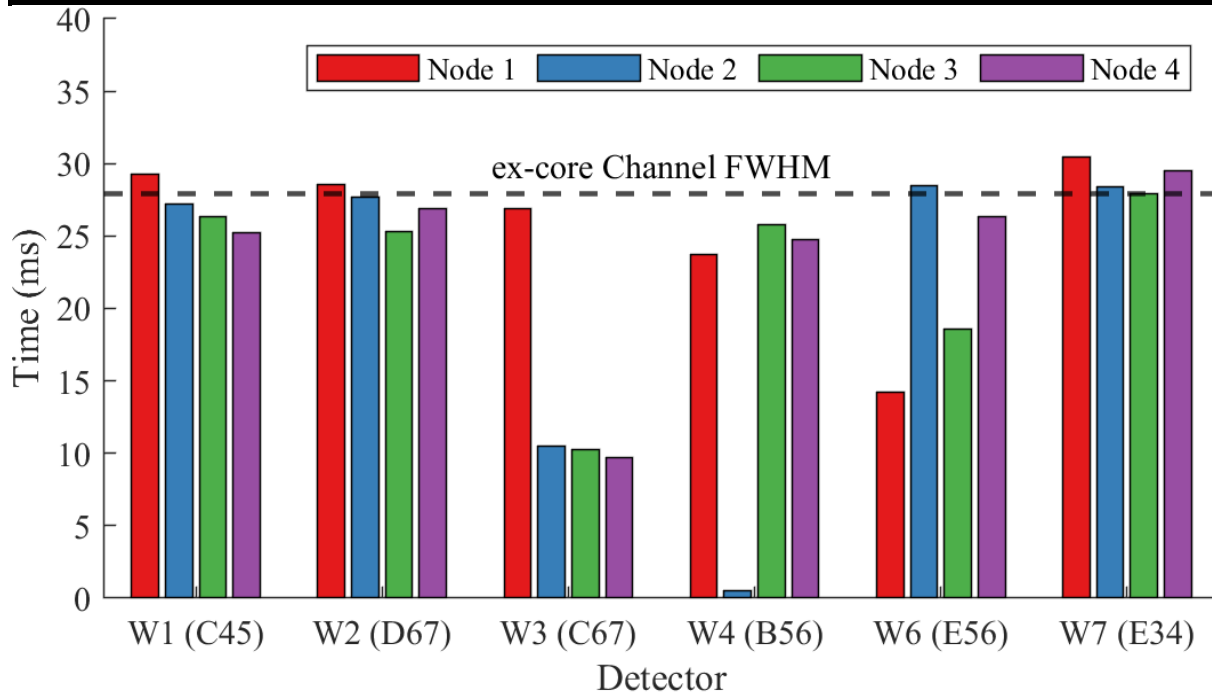


Fig. F-1. \$1.43 Pulse in the C2 configuration.

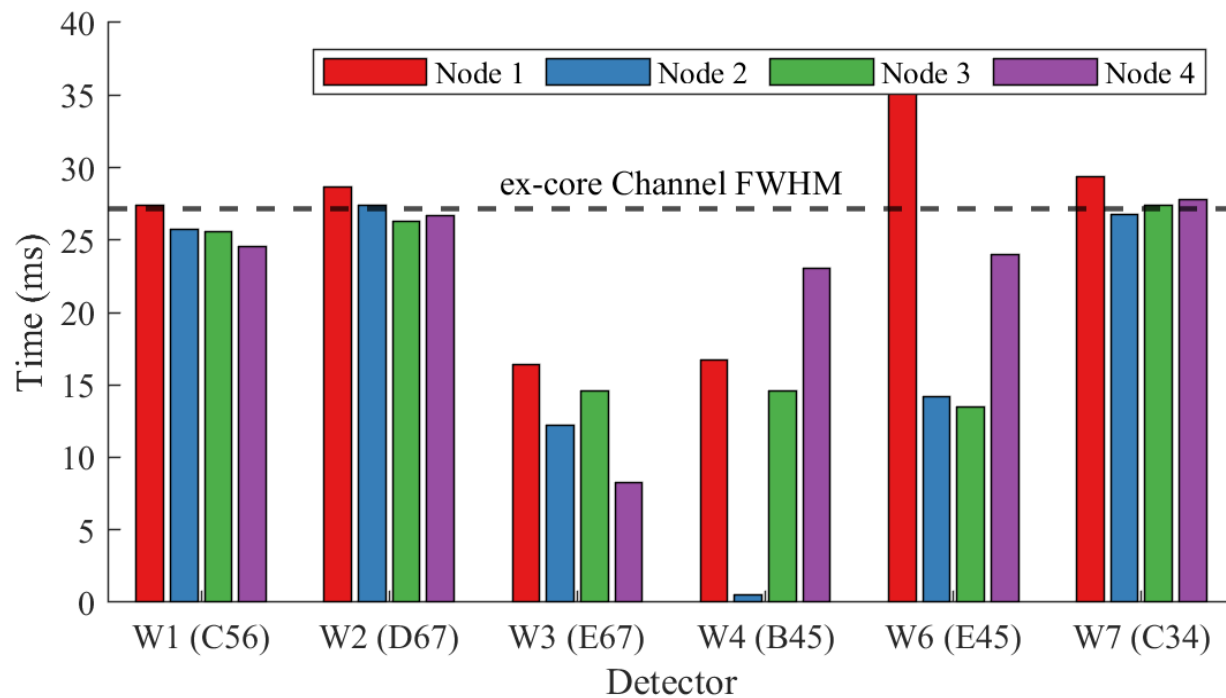


Fig. F-2. \$1.43 Pulse in the C1 configuration.

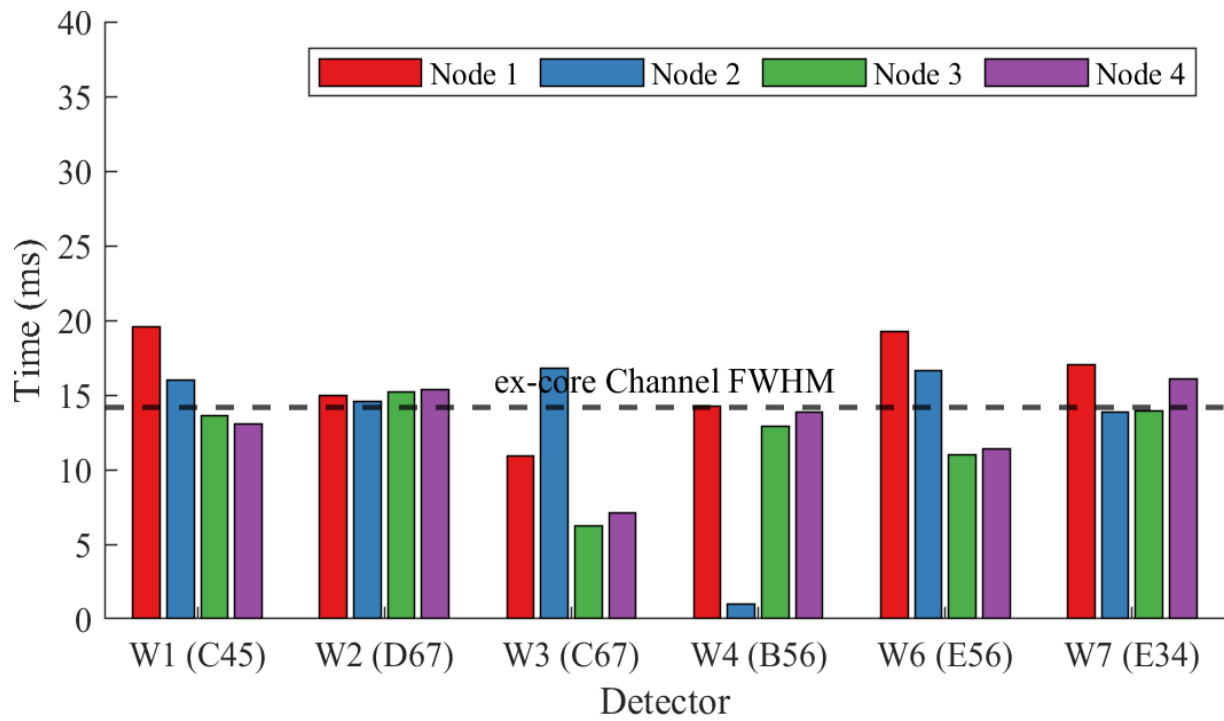


Fig. F-3. \$1.71 Pulse in the C2 configuration.

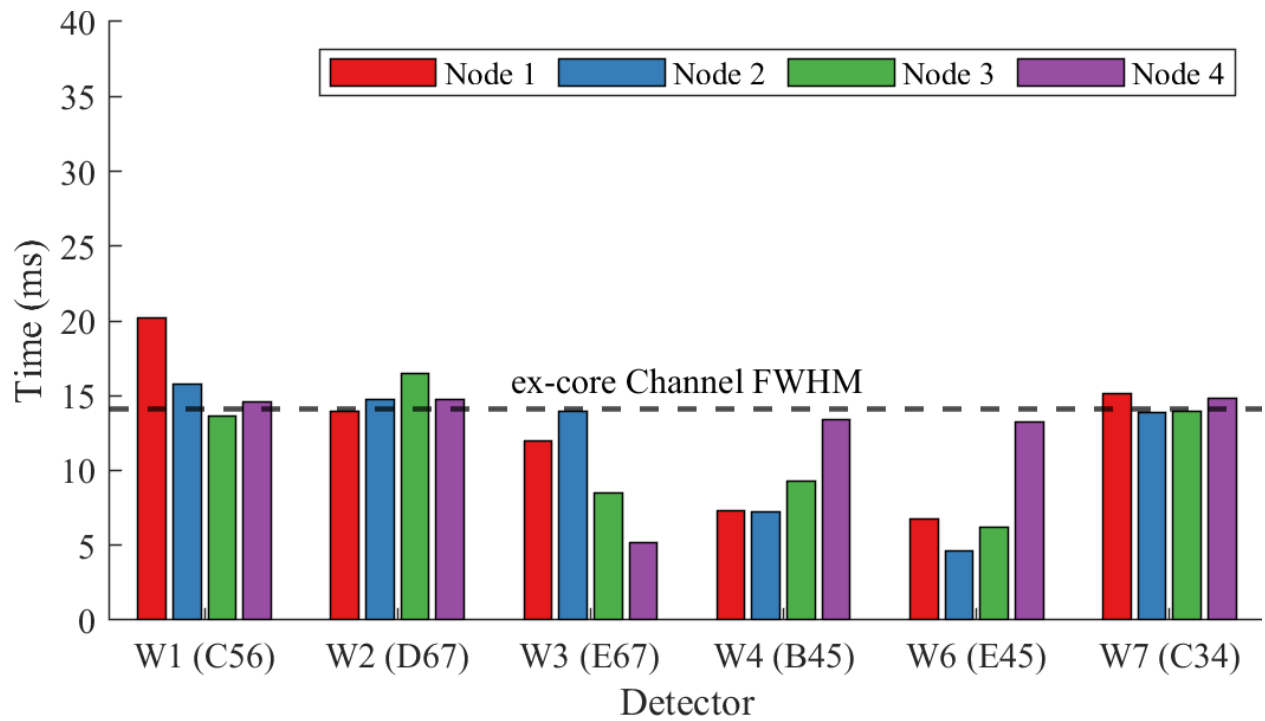


Fig. F-4. \$1.71 Pulse in the C1 configuration.

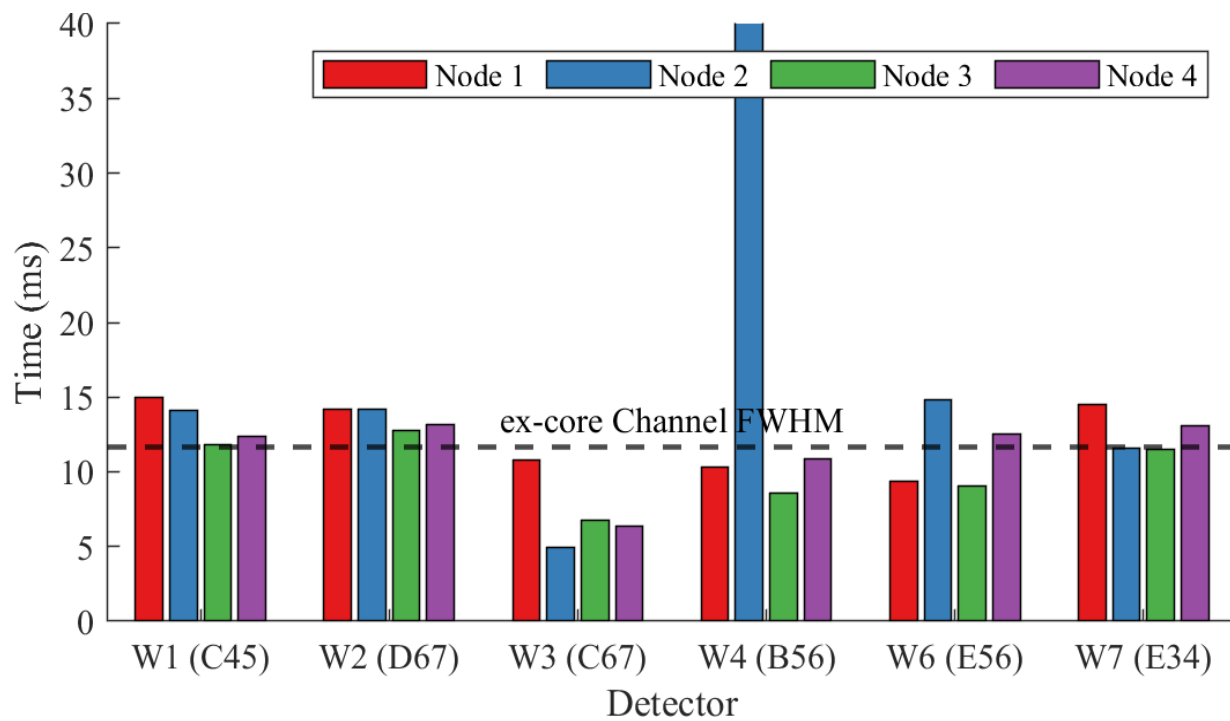


Fig. F-5. \$1.97 Pulse in the C2 configuration.

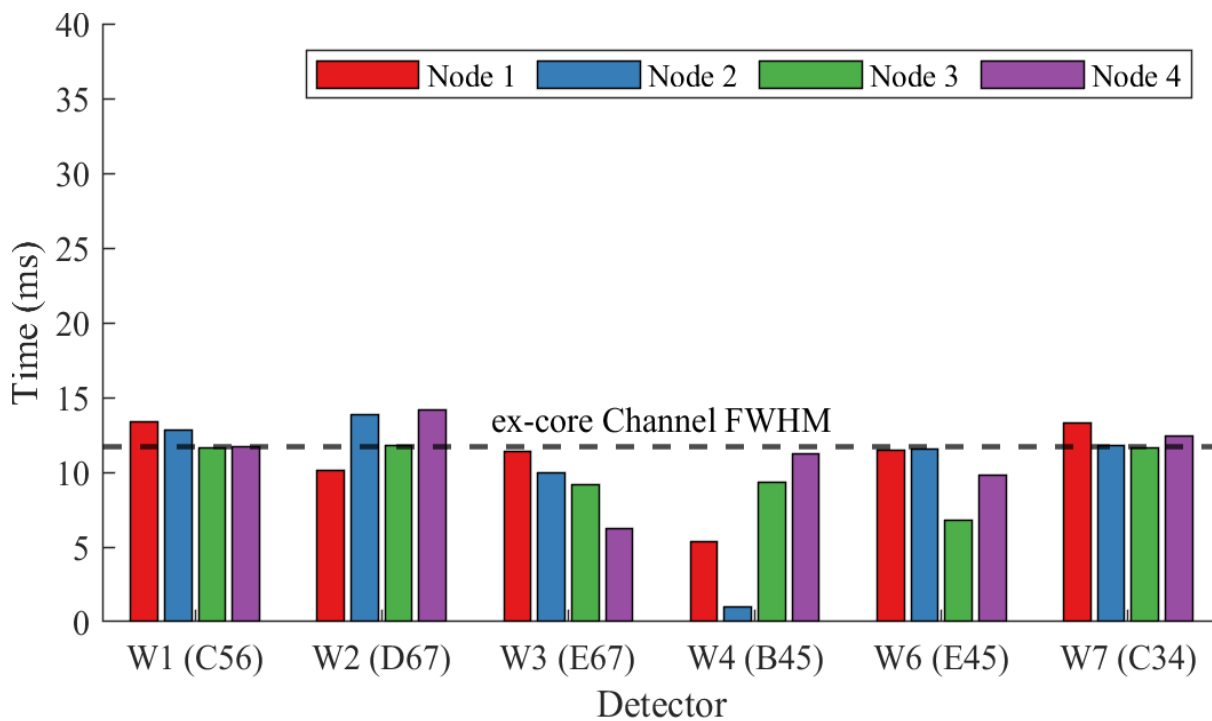


Fig. F-6. \$1.97 Pulse in the C1 configuration.

All plots displayed have been time correlated to have the U-node centered with the reactor ex-core sensor signal.

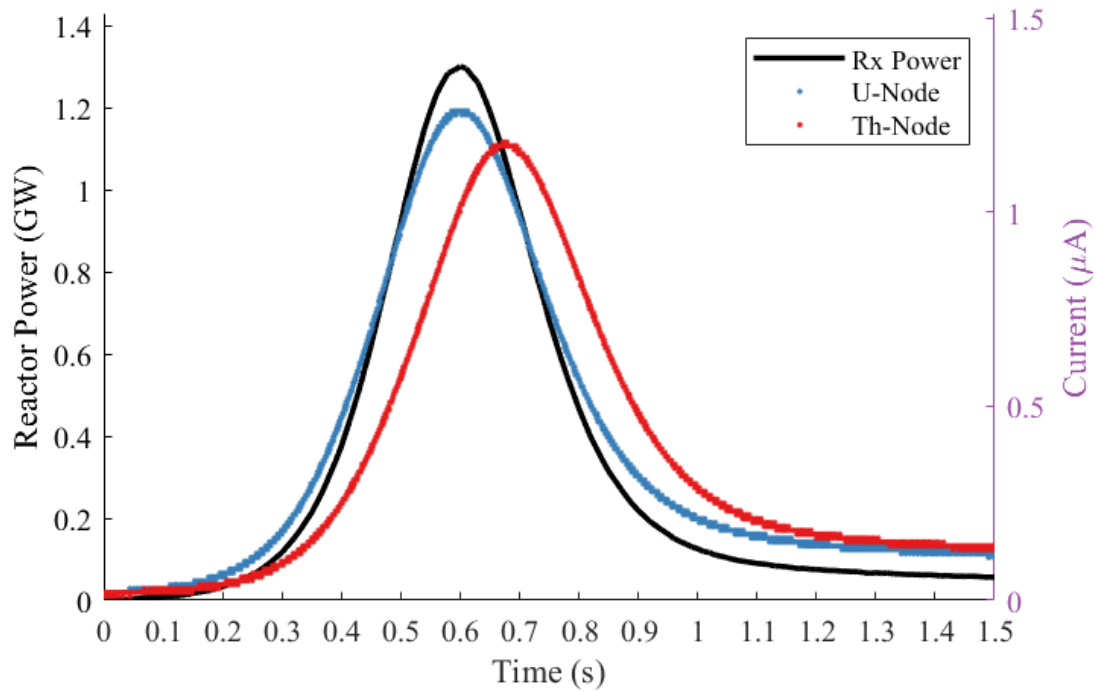


Fig. G-1. Plot of transient 2900 (1.6%  $\Delta k/k$ ).

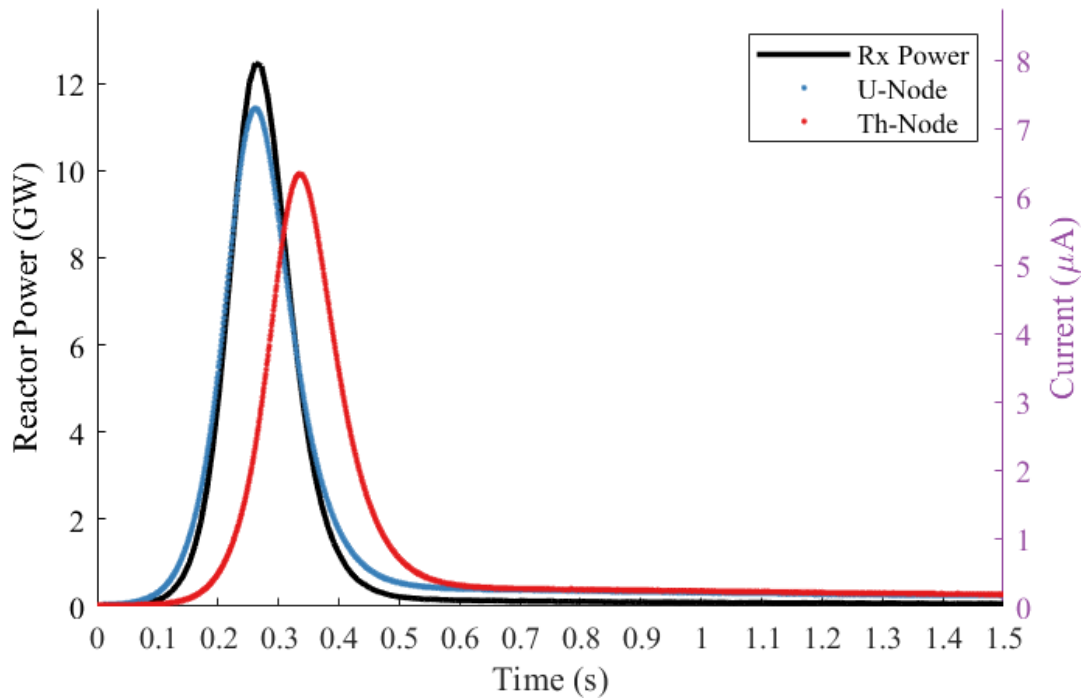


Fig. G-2. Plot of transient 2901 (3.85%  $\Delta k/k$ ).

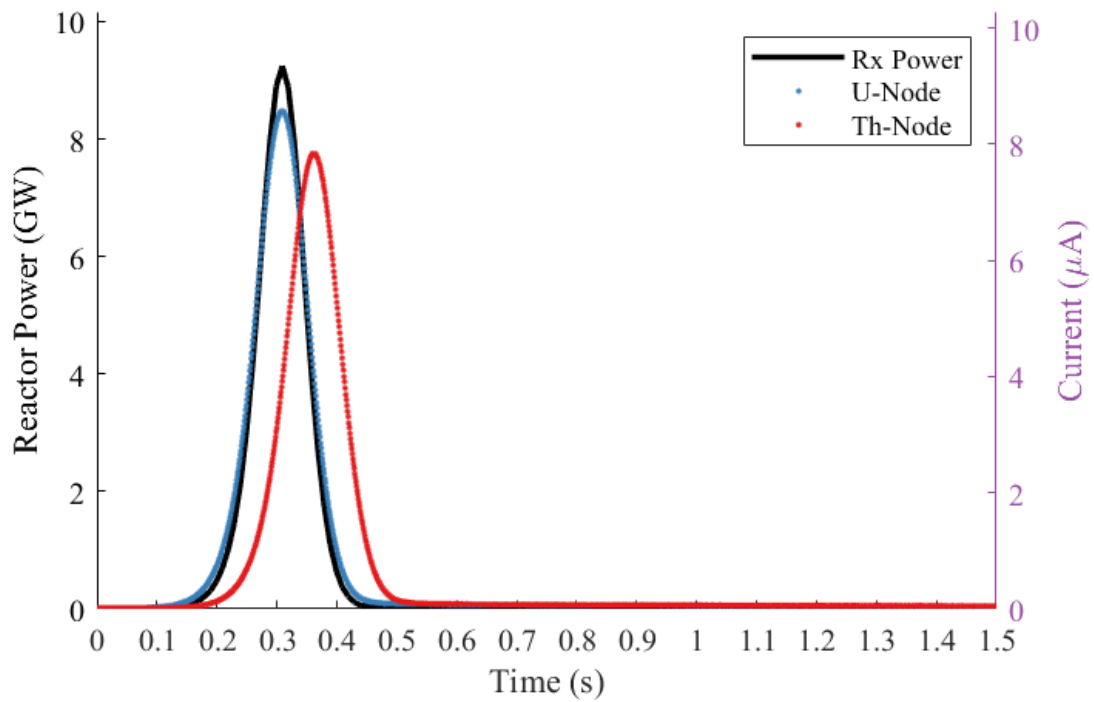


Fig. G-3. Plot of transient 2904 (475-ms NPW).

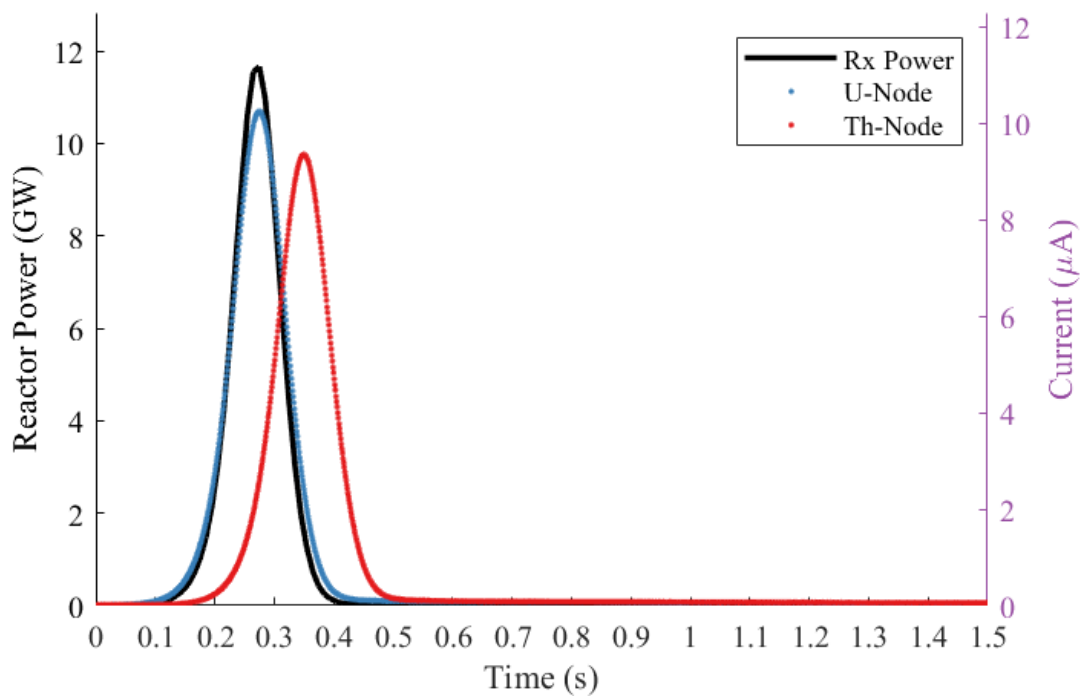


Fig. G-4. Plot of transient 2905 (500-ms NPW).

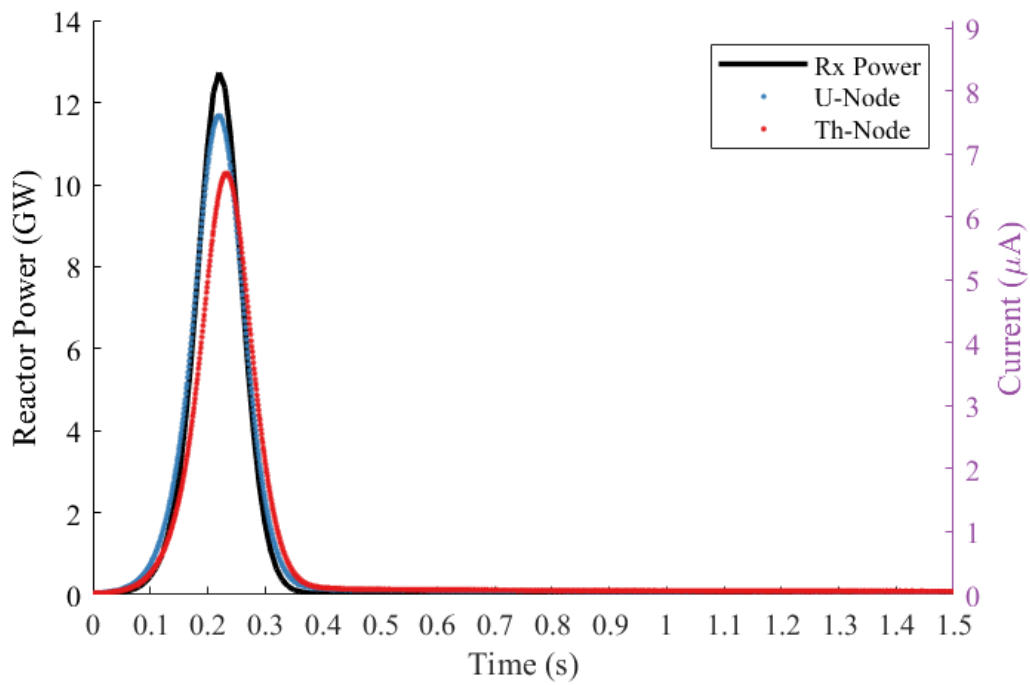


Fig. G-5. Plot of transient 2906 (513-ms NPW).

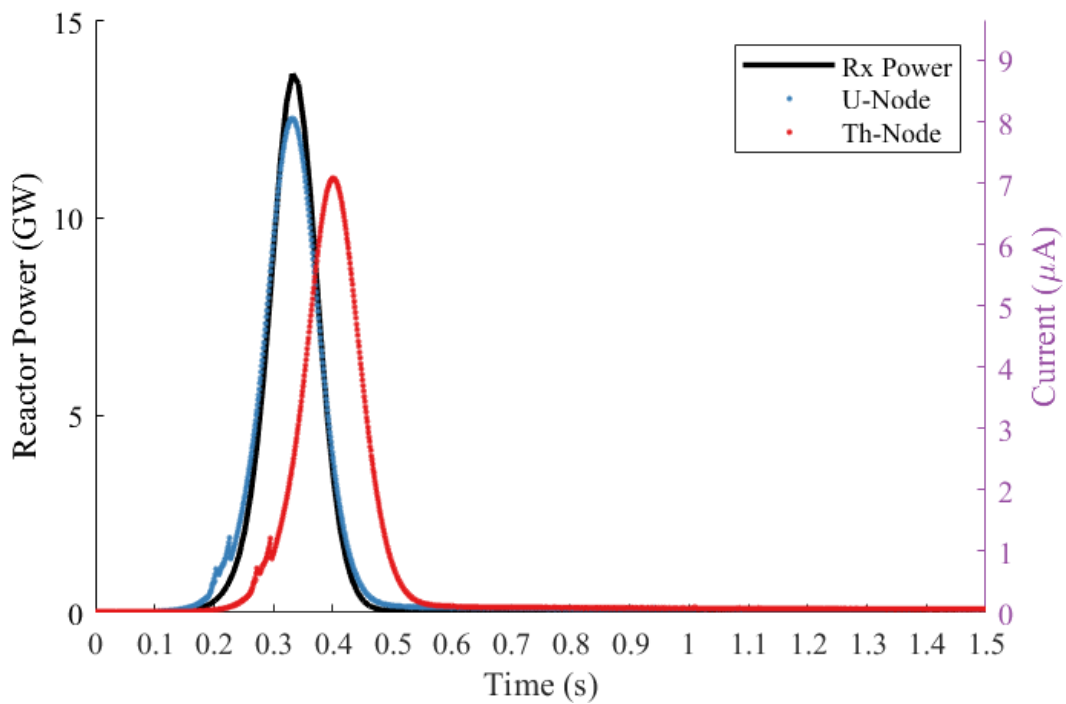


Fig. G-6. Plot of transient 2907 (525-ms NPW).

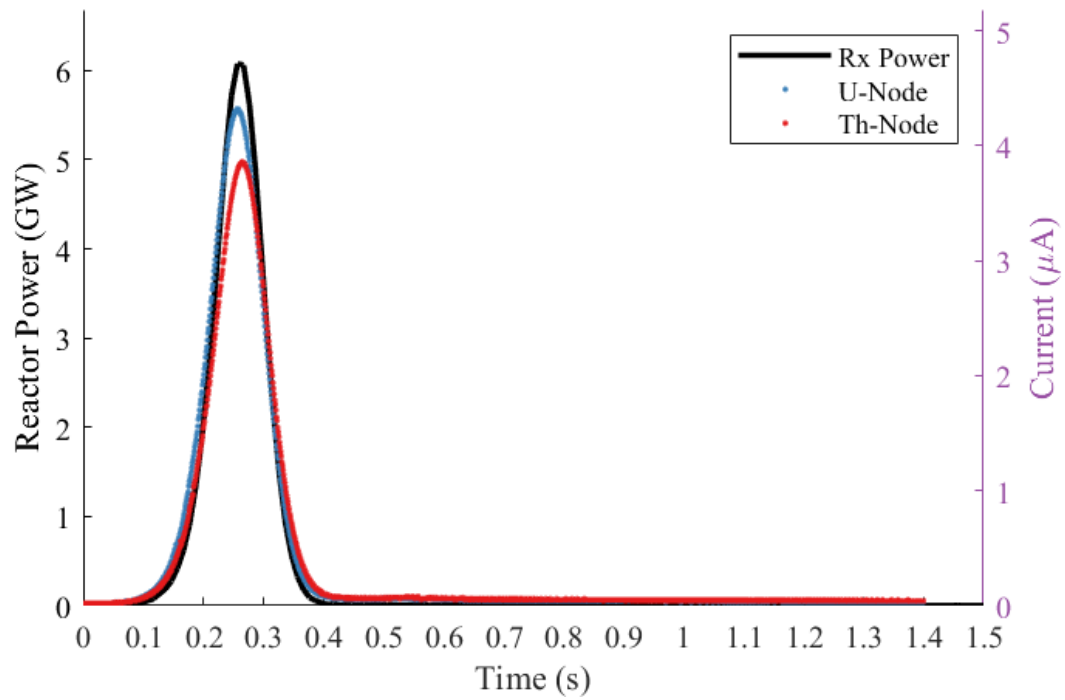


Fig. G-7. Plot of transient 2908 (450-ms NPW).