INL/RPT-23-75886

Performance Benchmark of Commercial and Developmental Fission Chambers in Elevated Temperatures

November 2023

Milestone Report – M3CT-23IN0702018

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SUMMARY

This report documents the testing of two in-core fission chamber technologies for high-temperature irradiation environments. This work is in collaboration with the French Alternative Energies and Atomic Energy Commission (CEA). The fission chamber evaluated by Idaho National Laboratory is the micro-pocket fission detector (MPFD). The fission chamber evaluated by the CEA are the 3 mm miniaturized fission chamber and the 7 mm high-temperature fission chamber.

Demonstrations of the MPFD were performed at the Neutron Radiography Facility and the Massachusetts Institute of Technology Reactor. Demonstrations of the CEA fission chambers were performed at the Ohio State University Research Reactor. All demonstrations were conducted with a heated experiment rig up to 850°C.

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MIT staff: David Carpenter, Michael Ames, and Yakov Ostrovsky.

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ACRONYMS

ATR-C	Advanced Test Reactor Critical
CEA	French Alternative Energies and Atomic Energy Commission
DOE	Department of Energy
FC	Fission Chamber
HT-FC	High-Temperature Fission Chamber
INL	Idaho National Laboratory
KSU	Kansas State University
LDRD	Laboratory Directed Research and Development
MITR	Massachusetts Institute of Technology Reactor
MPFD	Micro-Pocket Fission Detector
NRAD	Neutron Radiography (Facility)
OSURR	Ohio State University Research Reactor
RaCL	Radioanalytical Chemistry Laboratory
RDT	Radiation Detection Technologies, Inc.
SBIR	Small Business Innovation Research
TREAT	Transient Test Reactor (Facility)

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1. INTRODUCTION

A variety of advanced reactor designs have been in development with the support of the U.S. Department of Energy (DOE) for demonstration within the next 15 years [1]. Many of the designs employ novel fuels and moderators that enable higher operational temperatures, 650°C and above, for numerous advantages including increased thermal efficiency and process heat applications [2]. In addition to increasing the operational temperatures, many companies are also focusing on compact designs such as small modular reactors and microreactors, for deployments in remote areas. The high operational temperatures significantly limit the availability of in-core power monitoring sensors; most in-core fission chambers (FC) are rated up to 400°C and only a few are rated up to 600°C. Design compactness also increases ex-vessel temperatures limiting many traditional ex-core monitors; many of which are only rated up to 200°C [3], [4], [5]. Supporting reactor demonstrations and commercial deployments in the near future requires robust sensors capable of operating at high temperatures be designed and evaluated.

The design and evaluation of FCs for high-temperature environments are performed in collaboration with the French Alternative Energies and Atomic Energy Commission (CEA). This report presents the developmental overview of two FC technologies: the Micro-Pocket Fission Detector (MPFD) supplied by Idaho National Laboratory (INL) and the miniaturized FCs supplied by CEA.

Section 2 provides an overview of the MPFD's design and operation in high-temperature irradiations at the Neutron Radiography (NRAD) facility and the Massachusetts Institute of Technology Reactor (MITR). These demonstrations will conclude MPFD development at INL as it moves towards commercialization after receiving a Small Business Innovation Research (SBIR) fund. Section 3 presents the performance of the CEA miniaturized FCs at the Ohio State University Research Reactor (OSURR) where two 7-mm-diameter high-temperature fission chambers (HT-FCs) were supplied by CEA and benchmarked with a previously irradiated CEA 3-mm-diameter FC [6] up to 850°C.

2. MICRO-POCKET FISSION DETECTOR OVERVIEW

Originating from Kansas State University (KSU), the MPFD technology was brought to INL through collaboration as an HT-FC with the potential to perform thermal neutron flux, fast neutron flux, and temperature measurements within a single probe that can operate up to 800°C [7]. The INL-adapted design has gone through different iterations and demonstrated various successes using the Transient Test (TREAT) facility as a testbed. The MPFD was designed to operate in pulse mode to simplify detector calibration, with a large operational range, natural background gamma rejection, and resistance to temperature effects; however, detector noise was a difficult hurdle for reactor deployments. Noise characterization and rejection techniques were employed at the TREAT facility, attempting recover pulse mode operation. This resulted in a recognizable power trace, but the overall result remains unsatisfactory. The high lower-level discriminator settings only allowed approximately 10% of the expected count rate to be accounted, greatly diminishing the quality of the power trace [8]. Overall, successes from operating the MPFD in current mode merits its continual development, deployment, and evaluation through different projects. The latest MPFD improvement shown in Figure 1 was developed under a Laboratory Directed Research and Development (LDRD) project, LDRD 20A44-122FP [9], [10], [11].

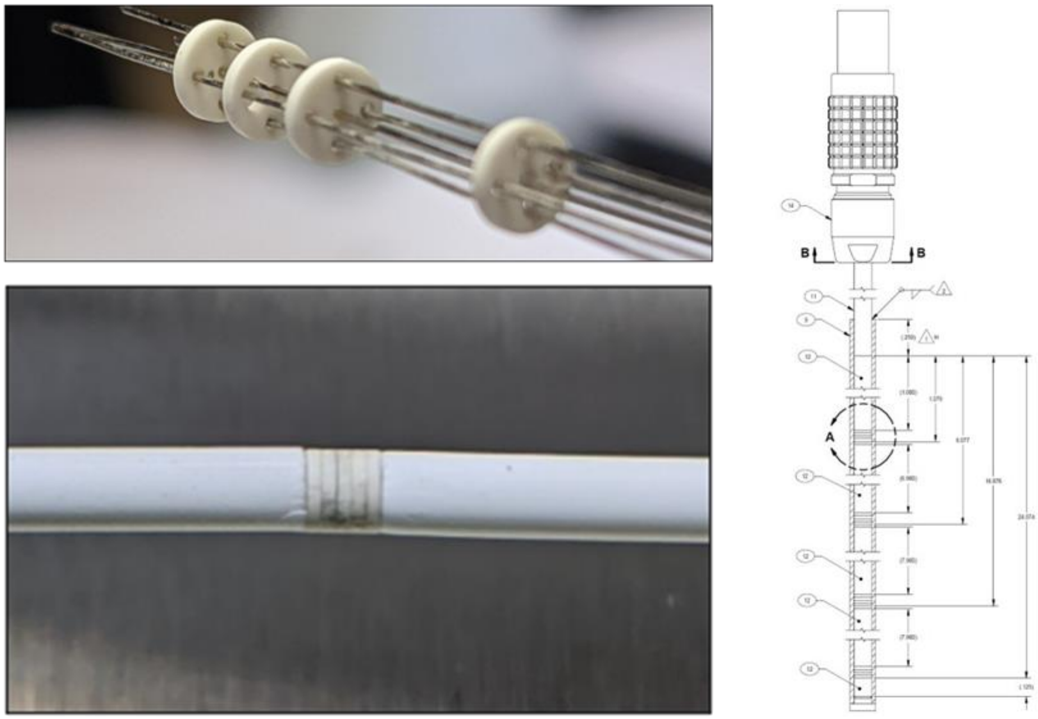


Figure 1. Design of the 4-node MPFD array.

2.1 Micro-Pocket Fission Detector Design Changes and Improvements

Fabrication method improvement was one part of an internally funded LDRD (20A44-122FP) that aimed to design an irradiation fixture for real-time axial neutron flux profile measurements at the Advanced Test Reactor Critical (ATR-C) Facility. The objective of this research was to demonstrate how advanced sensors could be used to significantly reduce the duration and cost of experiments, improve understanding of experimental environments, and enable the verification and validation of simulation and modeling methods. This was accomplished by designing and fabricating a dedicated real-time instrument test train for the ATR-C.

In the past, collaborations with KSU have provided some insight into the electrodeposition method for MPFD components using natural and low-enriched uranium. However, KSU was not able to procure or work with the high enrichment required for continued MPFD development. This project procured the necessary supplies and equipment to facilitate the electrodeposition of enriched uranium onto the small (~250 µm) diameter electrodes required for the MPFDs. The chemical preparation of the electrolytic solution was optimized, starting from the method developed at KSU. Internal collaboration with chemists in INL’s Energy and Environment Science and Technology directorate substantially improved both the chemical composition of the solution and the process to dramatically improve reproducibility and reduce material waste. A high-concentration enriched uranium solution was prepared from the uranium dioxide source material. The chemistry and electrodeposition were performed at the Radioanalytical Chemistry Laboratory (RaCL) at the ATR-C Facility. The RaCL has appropriate work controls and support to safely work with both the powdered uranium dioxide and liquid electrolytic solutions containing highly enriched uranium. The source solution was then diluted to prepare an appropriate amount of working solution that could be pH balanced using precision pipetting for a highly reproducible system. The electrolytic solution was fixed under a microscope to allow the chemists to make electrical contact with the working electrode, as shown in Figure 2.

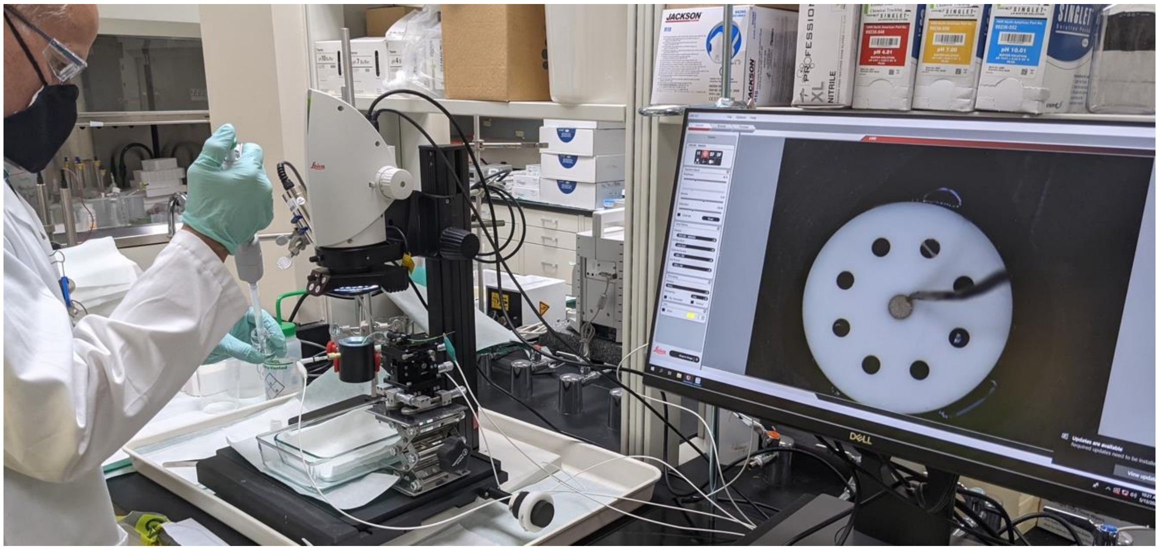


Figure 2. The electrodeposition system at RaCL that facilitates research with highly enriched uranium solution.

With the updated design and fissile deposit improvements made during the LDRD project, two sets of multi-node MPFDs were fabricated for testing as part of the FC evaluation to assess high-temperature compatibility. The two sets of MPFDs were deployed individually at the NRAD facility and MITR.

While maintaining the overall geometry, the NRAD MPFD was designed with only one node at the tip of the probe. The node contains 93% enriched uranium with mass of 2.96E-8 g. The rest of the 24-inch probe length is filled with silica insulation. During manufacturing, the welding process to seal the chamber resulted in a helium leak rate of 1.2×10^{-5} atm cc/s failing the anticipated helium leak rate of 6×10^{-6} atm cc/s. While the leak rate was above the anticipated value, additional evaluation deemed it acceptable.

The MITR MPFD was fabricated utilizing all four nodes within the design. The three nodes closest to the tip of the probe contain 93% enriched uranium with individual masses of 2.13E-8, 1.85E-8, and 2.27E-8 g followed by a fourth node containing no fissile material. The empty fourth node acts as a gamma ion chamber. A new welding procedure was developed after the NRAD MPFD fabrication, and the final chamber seal passed the helium leak check with a rate of 1.8×10^{-7} atm cc/s.

2.2 Neutron Radiography Facility Heated Irradiation

As a part of the comparative assessment of neutron sensors for high-temperature applications, the experiment rig used at the NRAD facility utilizes an insulated 2-inch-outer-diameter aluminum dry tube. The center of the dry tube contains six 0.25-inch-outer-diameter stainless-steel guide tubes surrounding a central cartridge heater [12]. The reactor is brought to 250 kW before increasing the temperature stepwise to 350°C, 550°C, 750°C, and 850°C. Figure 3 shows a plot of reactor power, temperature, and MPFD signal output. The MPFD output is provided by a transistor-to-transistor logic pulse that is proportional to the measured electric current of the MPFD operating in current mode.

The overall MPFD performance did not suggest a measurable neutron-generated signal. This failure was attributed to the previously acceptable leak rate of 1.2×10^{-5} atm cc/s. However, there are still notable measured temperature effects that did not correlate to a setting change performed in operating the MPFD (Figure 4). These effects are in response to temperature changes, particularly during temperature changes between each step beyond 350°C. These temperature effects are similar to the effects identified with the self-powered neutron detectors documented in previous reports and suggest a phenomenon measured in the MPFD mineral-insulated cable [12], [13].

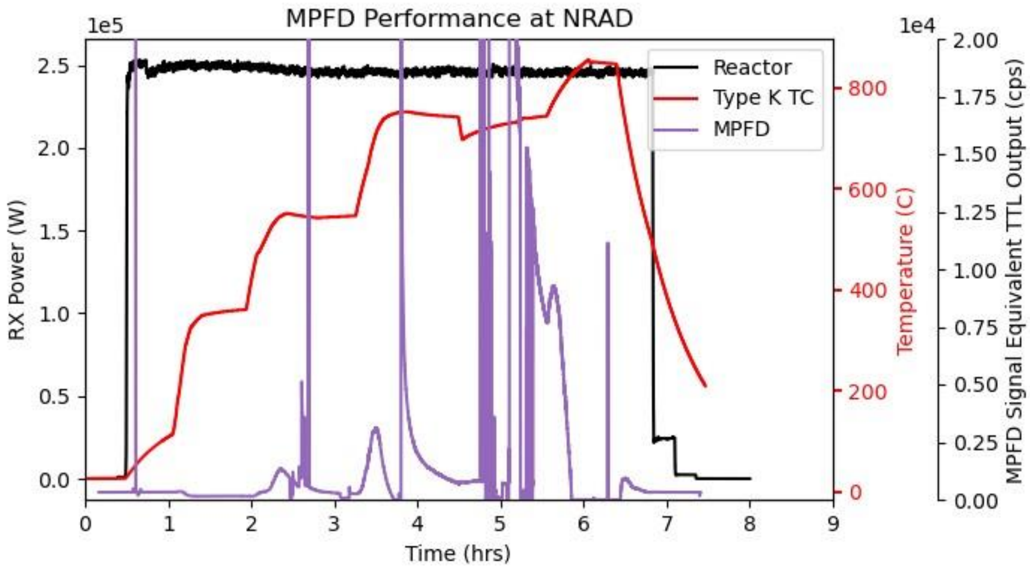


Figure 3. Overview of MPFD performance in high-temperature irradiation in the NRAD Facility.

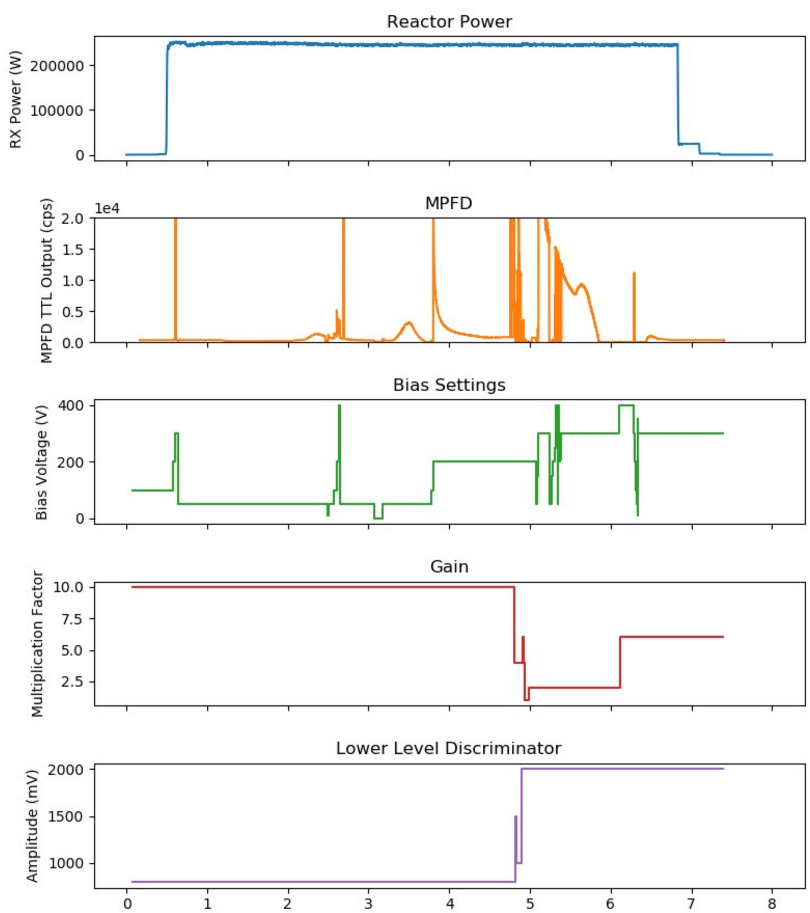


Figure 4. MPFD response to electronic setting changes.

2.3 Massachusetts Institute of Technology Reactor Heated Irradiation

Designed to mimic the NRAD experimental rig, the MITR experimental rig uses a similar six-slotted configuration with a central heat source [13]. However, the overall rig is made of graphite, and the central heat source is a tungsten alloy spine. The heat is provided through gamma heating in the tungsten spine. Heat transfer to the surrounding sensor slots is controlled by varying the composition of flow gas between helium and neon. However, given the reaction rates in the tungsten spine, the minimum temperature of the experiment rig at full power was 600°C. Therefore, detector response testing at low temperatures can only occur at low power.

Unfortunately, the MITR MPFD also showed unfavorable results during irradiation (Figure 5). At zero reactor power, MPFD channel 0, containing fissile material, and channel 3, an empty channel, both showed a maximum saturated signal. The channel remained saturated when the detector was connected and provided zero signals when the channel was disconnected. Only channels 1 and 2, both containing fissile materials, had reasonable outputs of 40 and 66 signal equivalent transistor-to-transistor logic pulses. However, while testing MPFD channels 0 and 3 prior to reactor startup, MPFD channel 1 inexplicably dropped to a zero-signal output. Channel 1 would eventually come back online during reactor startup. Unfortunately, channels 1 and 2 did not show responses indicative of a neutron event with rising reactor power and quickly dropped to a zero signal near 400°C without any further recovery.

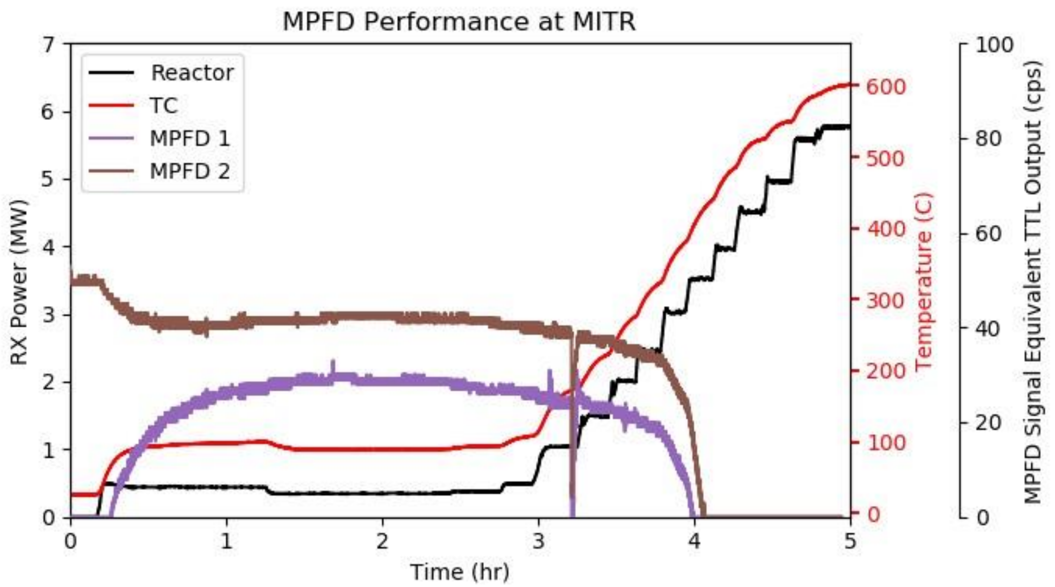


Figure 5. Overview of the MPFD performance at the MITR.

Given the results observed at the MITR, the conclusions are:

1. The MPFD fabrication needs to be improved. While improvements were made to the chamber seal, a relatively high failure rate remained.
2. The noise level of the detector needs to be improved to enable pulse mode. Since the detector can only be operated in current mode, it is difficult to determine whether the signal was induced by the reactor (including gamma contributions) or electrical noise.
3. Communication with the electronic system and detector may be a concern. This is due to the fact that there were no observed signals relating to reactor power at low temperatures (below 100°C) at the start of the irradiation at NRAD and MITR with thermal fluence rates of ~1E12 n/cm^2-s.

These conclusions regarding fabrication were soon addressed by the awarding of the SBIR proposal. An additional outlook of a Phase II proposal to address the detector noise and communication with the electronic system was also discussed, hence concluding and transferring the work from INL.

2.4 Commercialization Efforts Through Small Business Innovation Research

Following the improvements made during prior research, a collaborative Phase I SBIR proposal was submitted to the DOE, led by Radiation Detection Technologies, Inc. (RDT). The proposal was awarded (Award Number: DE-SC0022841) for the period between June 2022 through June 2023. The research and development under this SBIR were led by RDT and supported by INL. The goal of this effort was to elevate the MPFD from laboratory technology to a commercially viable product, satisfying real-time, in-core neutron detector needs within the nuclear reactor community.

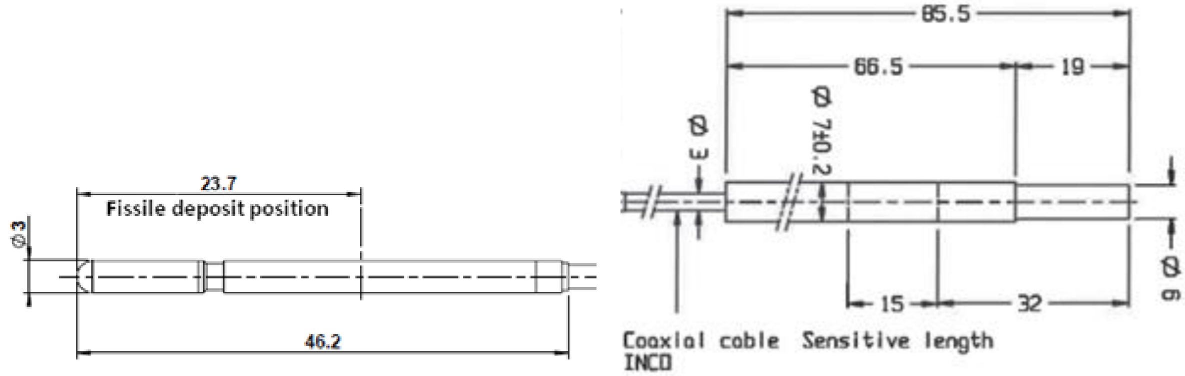
In Phase I, the team addressed several critical manufacturing issues at the subcomponent level that have been prohibitively costly or unreliable in previous work. These issues are represented in the prior sections of this report. Phase I began with a round of stakeholder interviews to refine the design constraints for the MPFD sensor. Based on these design constraints, the team set out to modify the MPFD subcomponents to improve manufacturing yield and reduce costs. One of the main concerns was to address the unreliable methods of purging, backfilling, and sealing with high-purity argon in the detector chamber. A major objective for this project was to devise a reliable and commercially ready method for hermetically sealing the gas chamber. Major achievements include redesigning the chamber and fissile material geometry to improve manufacturing yield and reduce cost while retaining detector performance, proving electroplating feasibility for the fissile-coated neutron conversion wire, and integrating a gas pinch-off tube into the MPFD design that will allow us to evacuate and backfill the MPFD detector chamber with optimum gas fill pressure. The full results of the Phase I SBIR can be found in the final report submitted by RDT to DOE.

A Phase II proposal was submitted but not awarded for continued development. RDT continues to pursue additional funding through public grants and private investment. Continued engagement with RDT in these proposals remained but the opportunities for direct research and development of MPFDs are limited at this point due to the lack of a commercially viable design and fabrication pipeline.

3. FRENCH ALTERNATIVE ENERGIES AND ATOMIC ENERGY COMISSION FISSION CHAMBER OVERVIEW

The CEA has a long history of designing and fabricating miniaturized FCs for material test reactor use. The FCs utilize a coaxial cylinder design with pressurized gas with many versions available for purchase through CEA or the commercial vendor, Photonis. Sub-miniaturized versions can have probe diameters as small as 1.5 mm and larger sizes. The larger sizes are often used for higher sensitivity and temperature ratings up to 600°C.

For the high-temperature testing performed at the OSURR, CEA has supplied two types of FCs: a 3-mm-diameter FC rated for 400°C used in a previous experiment [6] and two 7-mm-diameter HT-FCs rated for 600°C. A schematic overview of the two FC designs is given in Figure 6, and the fissile mass and fill-gas specifications are given in Table 1. All three FCs were operated through the Libera MONACO system. While the system allows the simultaneous operation of pulse, Campbelling, and current modes, the desired FC operational modes were pulse and Campbelling modes due to their natural resistance to leakage currents as a function of temperature. However, the detector noise becomes a larger concern.



(a) (b)

Figure 6. Schematic overview of the (a) 3 mm FC and (b) 7 mm HT-FC.

Table 1. CEA FC fissile material and fill-gas specifications.

Design	Fissile mass	Fissile deposition length	Fill gas
3mm FC	1 µg U-235	10 mm	Argon + 4% N @ 5 bar
7mm HT-FC	170 µg U-235	15 mm	Argon @ 1 bar

3.1 Ohio State University Research Reactor Heated Irradiation

The heated irradiation at OSURR utilizes the 9.5-inch dry tube with a 24-inch-long cylindrical furnace. The furnace has a 2-inch inner diameter to enable sensor testing. The dry tube is positioned to the east of the reactor core. An overlay of the furnace with respect to the temperature and neutron flux profile is given in Figure 1Figure 7. The overall experiment is performed over 3 days covering from ambient to 850°C and reactor power between 100 W and 450 kW. A detailed irradiation plan is given in Table 2 – Table 4; however, the duration only is listed as an estimate.

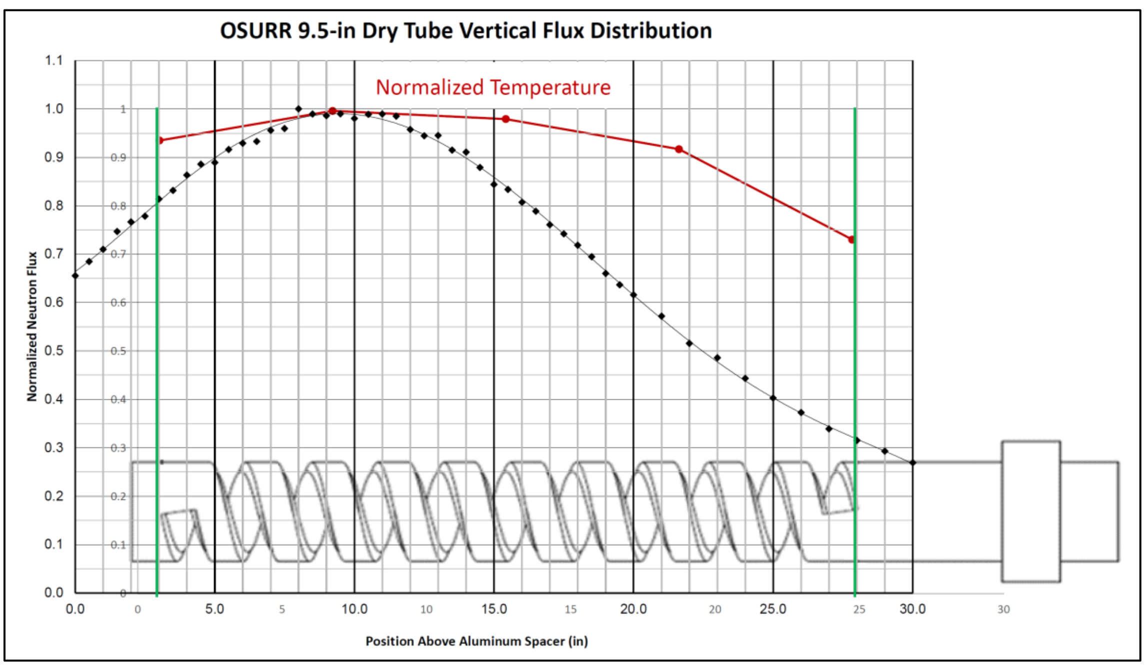


Figure 7. Overlay of furnace length, temperature profile, and neutron flux profile.

Table 2. OSURR day 1 irradiation plan.

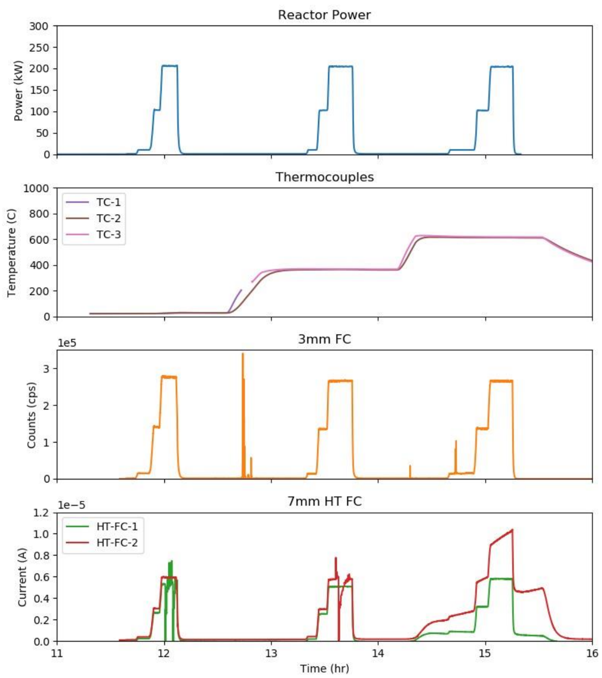
Power	Duration	Temperature
100 W	1 hr	Ambient
100 W 1 kW 10 kW 100 kW 200 kW	15 min each	Ambient
1 kW	1 hr	Heat to 350°C
1 kW 10 kW 100 kW 200 kW	15 min each	350°C
1 kW	1 hr	Heat to 600°C
1 kW 10 kW 100 kW 200 kW	15 min each	600°C

Table 3. OSURR day 2 irradiation plan.

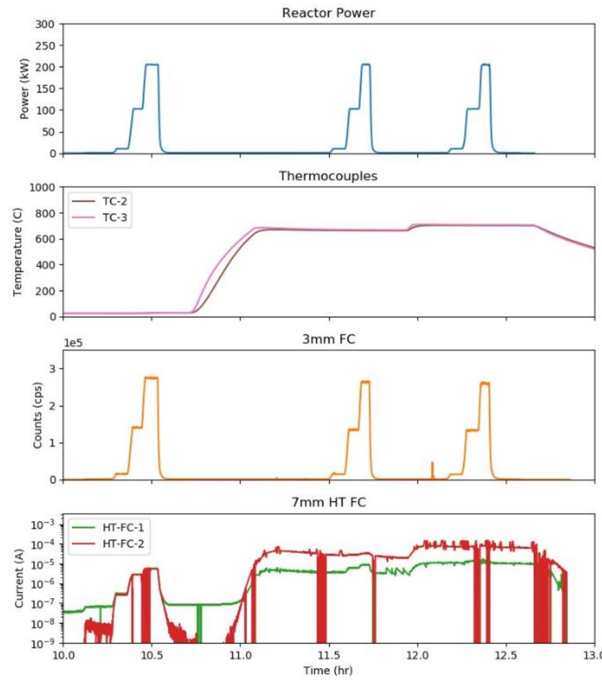
Power	Duration	Temperature
100 W	1 hr	Ambient
100 W 1 kW 10 kW 100 kW 200 kW	15 min each	Ambient
1 kW	1 hr	Heat to 650°C
1 kW 10 kW 100 kW 200 kW	15 min each	650°C
1 kW	1 hr	Heat to 700°C
1 kW 10 kW 100 kW 200 kW	15 min each	700°C

Table 4. OSURR day 3 irradiation plan.

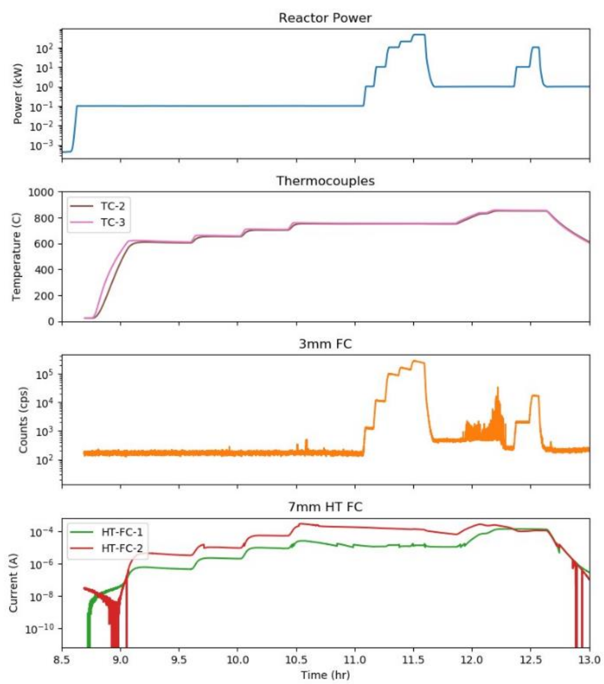
Power	Duration	Temperature
100 W	5 min each	Ambient 600°C 650°C 700°C 750°C
100 W/ 1 kW 10 kW 100 kW 200 kW 450 kW	15 min each	750°C
1 kW	1 hr	Heat to 850°C
1 kW 10 kW 100 kW	15 min each	850°C
1 kW	1 hr	Heater off



(a)



(b)



(c)

Figure 8. Overview of the OSURR irradiation occurring over (a) day 1, (b), day 2, and (c) day 3.

The overview plot of the detector response for the 3 days of irradiation is shown in Figure 8. In general, the 3 mm FC, despite having a temperature rating of 400°C, has the best overall performance. The 7 mm HT-FCs had significant electrical noise issues such that the pulse and Campbelling modes were inoperable. The electrical noise of the HT-FCs was theorized to be from the improper selection of mineral-insulated cable. Since the 7 mm HT-FCs had to be operated in current mode, it was more influenced by the increasing leakage current as a function of increasing temperature. This is further observed by the sensitivity curves at different temperatures given in Figure 9 – Figure 11.

Shown in Figure 9, the 3 mm FC demonstrated strong linearity in response to reactor power up to 700°C. However, signs of detector failure were observed at 750°C. Decrease in detector response over time is shown in Figure 8(c). Further investigation into the mean pulse amplitude and charge deposition (shown in Figure 12) suggests a decrease in chamber gas pressure, indicating a failure of the chamber seal.

Shown in Figure 10 and Figure 11 are the sensitivity curves for 7 mm HT-FCs under different temperatures. Per the rated operational temperature of 600°C, the HT-FC-1 performed satisfactorily with similar sensitivity at 200 kW reactor power. However, given the leakage contributions observed in the lower power levels, the HT-FCs would not be applicable for modular-type reactors.

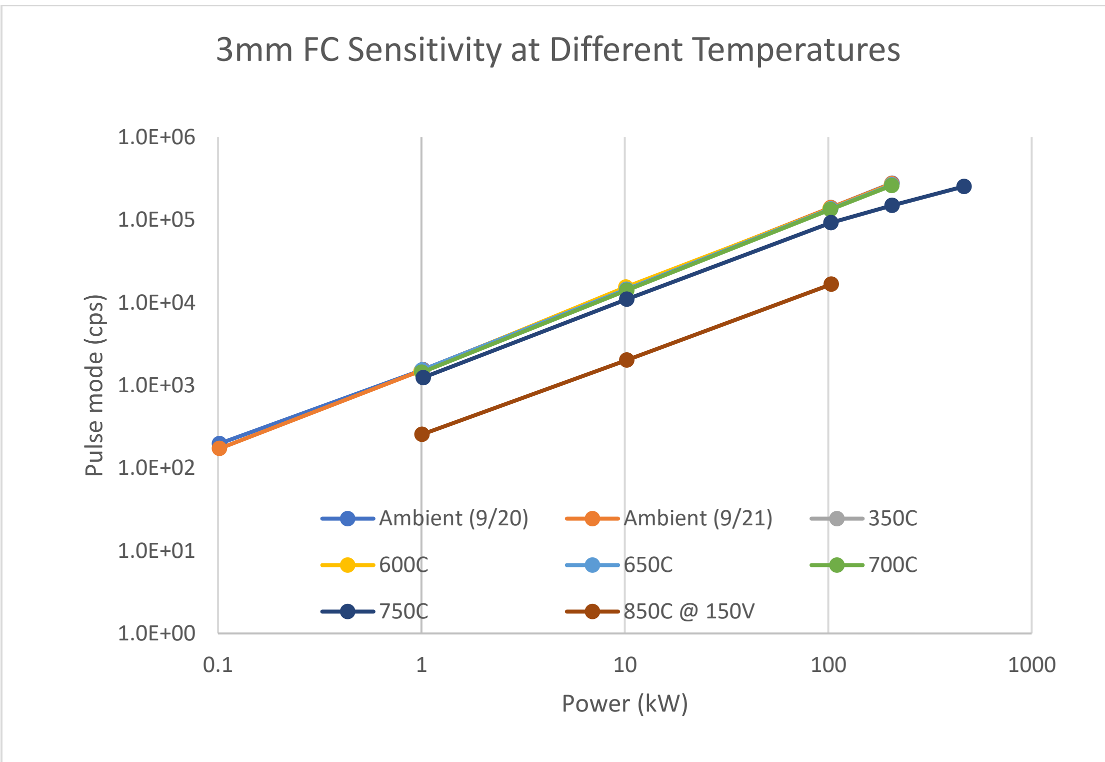


Figure 9. 3 mm FC sensitivity curve at different temperatures.

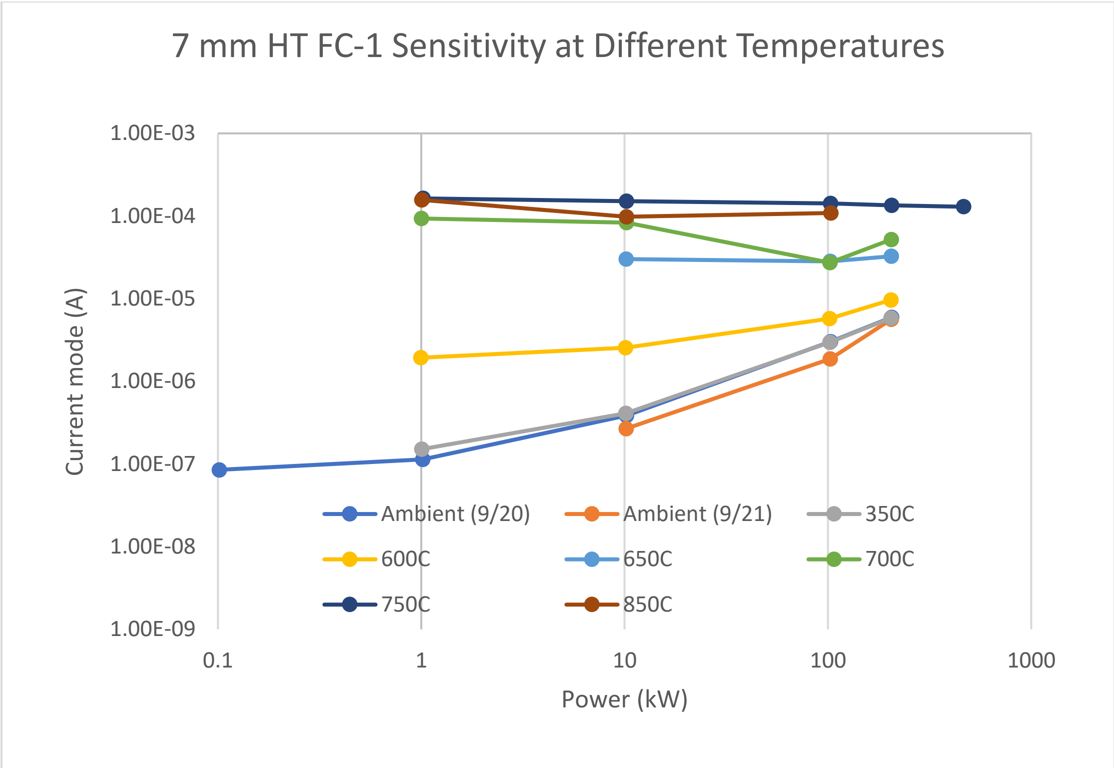


Figure 10. 7 mm HT-FC-1 sensitivity curve at different temperatures.

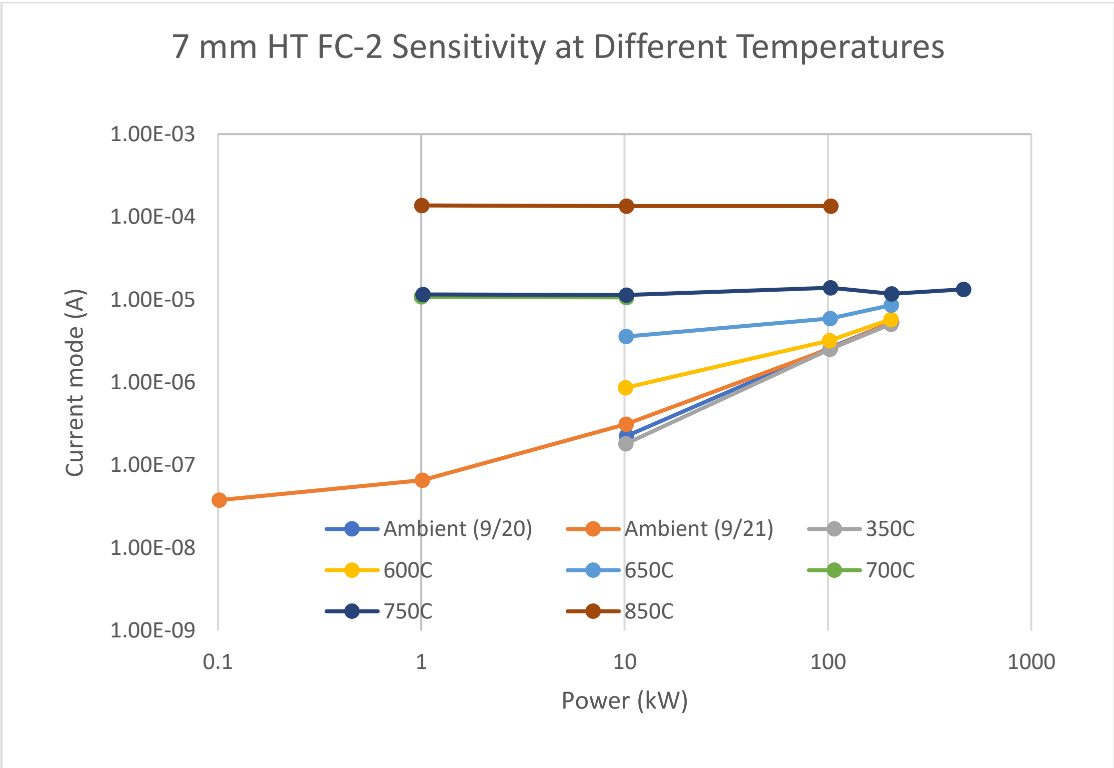


Figure 11. 7 mm HT-FC-2 sensitivity curve at different temperatures.

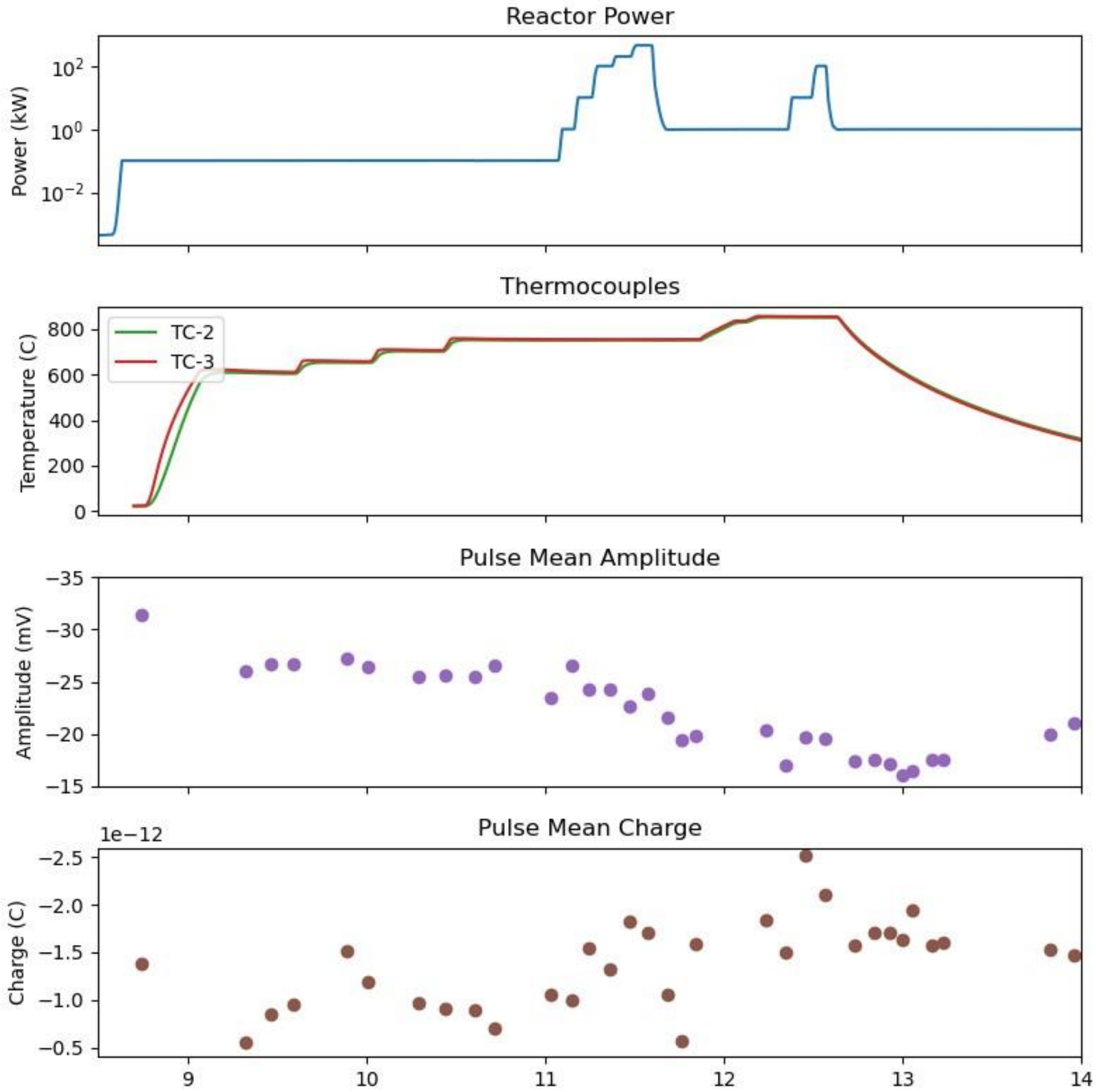


Figure 12. 3 mm FC mean pulse amplitude and charge plot over time.

4. CONCLUSION

In summary, the MPFD design and fabrication was performed at INL with its demonstration at the NRAD facility and MITR with temperatures ranging from ambient to 850°C. Separately, two CEA miniaturized FC designs were tested at the OSURR at temperatures up to 850°C.

Identified limitations and failure modes of both FC technologies in high-temperature applications can be summarized as:

- Susceptibility to background electrical noise needs to be accounted for in the design and fabrication. Identified from the CEA 3 mm fission chamber, the ideal components are the high-transmission mineral insulated cable with good resistance to electromagnetic interference, and a direct termination to a low noise connector.
- With the reduction of background electrical noise, it is ideal to use pulse mode or Campbelling mode operation for all ranges of reactor power monitoring.

    - Separately, as seen from the electric current contributions from temperature changes in the MPFD observed at NRAD to the leakage current contributions in the 7 mm HT-FC at higher temperatures, current mode operation is only applicable for high-flux reactor environments or power range reactor monitoring.
- Gas-chamber seals will need to be evaluated for high-temperature applications. This was the case for the MPFD as constructed at INL as well as the 3mm FC (rated for 400°C). However, given the results produced by RDT in the Phase I SBIR, solutions exist.

Future designs of FCs operating at high-temperature environments are suggested incorporate these design features to minimize failure modes.

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