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Technical note

Micro-Pocket Fission Detectors (MPFDs) for in-core neutron detection

Michael A. Reichenberger [a,*], Troy C. Unruh [b], Philip B. Ugorowski [a], Takashi Ito [c], Jeremy A. Roberts [a], Sarah R. Stevenson [a], Daniel M. Nichols [a], Douglas S. McGregor [a]

[a] S.M.A.R.T. Laboratory, Mechanical and Nuclear Engineering Dept., Kansas State University, Manhattan, KS 66506, USA
[b] Idaho National Laboratory, Idaho Falls, ID 83415-3531, USA
[c] Department of Chemistry, Kansas State University, 213 CBC Building, Manhattan, KS 66506-0401, USA

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ABSTRACT

Neutron sensors capable of real-time measurement of neutrons in high-flux environments are necessary for tests aimed at demonstrating the performance of experimental nuclear reactor fuels and materials in material test reactors (MTRs). In-core Micro-Pocket Fission Detectors (MPFDs) have been studied at Kansas State University for many years. Previous MPFD prototypes were successfully built and tested with promising results. Efforts are now underway to develop advanced MPFDs with radiation-resistant, high-temperature materials capable of withstanding irradiation test conditions in high performance material and test reactors. Stackable MPFDs have been designed, built, and successfully demonstrated as in-core neutron sensors. Advances in the electrodeposition and measurement of neutron reactive material, along with refinements to composition optimization simulations, have enhanced the capabilities of contemporary MPFDs.

1. Research motivation

The development of small, accurate, and robust neutron flux measurement system is an important enhancement for advancing nuclear technology. Ionization chambers and fission chambers are commonly used outside a reactor core to measure the neutron flux (Knoll, 2010). Recent interest in the development of advanced fuel and material testing in the United States has exposed the necessity for neutron sensors which can be deployed for long-lifetime, in-core and near-core measurements (Unruh et al., 2013). The physical requirements for in-core neutron sensors limit material selection and device geometry. Several technologies exist that are used to measure neutron flux for in-core and near-core environments.

Ionization chambers and fission chambers are commonly used for near-core neutron measurements (Knoll, 2010). Typical ionization and fission chambers are necessarily large, and are only capable of monitoring neutrons which have escaped the reactor core. Such devices are impractical for in-core measurements because of their large size, fragile construction, and large flux perturbation.

Alternatively, iron or gold activation analysis can be used to determine the neutron fluence within a reactor core during an operational period. However the neutron fluence is not as useful for experiments in high-performance reactors, transient test reactors, and critical mock-ups, which distinctly benefit from real-time flux measurement.

Self-powered neutron detectors (SPND) incorporate neutron-sensitive materials that decay by beta or gamma-ray emission. The simplest versions of SPNDs rely on the direct measurement of the beta decay current following a neutron absorption (Knoll, 2010). In contrast to typical fission chambers, SPN detectors are typically very small, and require no applied bias. However, the output current from SPN detectors is very small, and many types suffer a time delay limited by the half-life of the activated material (Knoll, 2010).

Miniature fission chambers are commonly used for in-core neutron measurements. Miniature fission chambers are typically lined with highly enriched uranium, and are typically made in a cylindrical geometry (Knoll, 2010). Device dimensions for miniature fission chambers are usually in the mm to cm range. The burnup of fissile material, and buildup of fission fragments in sealed miniature fission chambers greatly limits the application of such devices for extended periods of time. A fission chamber using enriched ^{235}U will decrease in sensitivity by 10% after an integrated neutron fluence of 10^{20} n/cm^2 in a typical power nuclear reactor (Böck and Balcar, 1974). In order to extend stable device lifetime, fertile isotopes can be added to the neutron-sensitive coating (Böck and Balcar, 1974). The buildup of fission fragments in the sealed gas

* Corresponding author.

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chamber of typical miniature fission chambers also produces a 'memory effect', where the radioactive decay of fission fragments in the detection gas produces a residual current, thereby reducing device accuracy (Knoll, 2010).

Finally, sub-miniature fission chambers utilize highly-enriched uranium (97% ^{235}U) coatings upon the cathode wall of small, sealed, proportional gas-filled detectors (Blandin et al., 2003). Current mode operation is required for in-core operation of sub-miniature fission chambers due to the high neutron sensitivity of the fissile coating. The stable device lifetime also suffers from the high sensitivity, limiting the effective use of such detectors to fluences below 2×10^{21} n cm^{-2} (Blandin et al., 2003). Such devices are impractical for use in critical mock-ups, high performance material test reactors (MTRs), and transient test reactors because of low-fluence design and large flux perturbation when installed in-core or near-core.

Micro-Pocket Fission Detectors (MPFDs) are under development, which are constructed of materials resistant to the high-neutron flux and high-temperature environment present in the core of many material test reactors (MTRs). In addition, MPFDs do not significantly perturb the local neutron flux, while providing a real-time measurement of critical reactor parameters (such as neutron flux and temperature), and are capable of continuous operation during extended testing periods.

The present status of MPFD sensor development is discussed here. Previous studies have shown potential for the development of advanced sensors (McGregor et al., 2005). The present work has enhanced the development of MPFDs by using a controlled deposition process for neutron-conversion material, by developing a non-destructive means to measure the amount of neutron-conversion material deposited, and by numerical optimization of the device lifetime for high-fluence applications.

2. Previous developments

The preliminary development of MPFDs has shown by simulation that the use of such devices for in-core neutron-flux measurement can be used to reconstruct the neutron-flux profile (and subsequently the power density profile) in the KSU TRIGA MkII nuclear reactor (Ohmes et al., 2004). Additionally, a large (3 mm diameter electrode) prototype utilizing enriched ^{235}U was tested in a neutron beam-port (neutron flux of $\sim 10^8$ n cm^{-2} s^{-1}) (Ohmes et al., 2004). The results of previous studies have shown that MPFDs can be built that are neutron-sensitive, and have very high gamma-ray discrimination (McGregor et al., 2005). The miniature size of MPFDs presents several distinct advantages for in-core neutron monitoring, including reduced gamma-ray sensitivity, uniform charge collection, and minimal neutron perturbation (McGregor et al., 2005). The highly energetic fission fragments resulting from the fission of heavy isotopes deposit significantly more energy in a small (<1 mm thick) gas pocket than gamma rays and/or electrons, as shown in Fig. 1 (McGregor et al., 2005). Fission fragments, although varied in energy (commonly ranging between 60 MeV and 100 MeV) and size (commonly ranging between $Z \approx 90$ and $Z \approx 140$), all deposit roughly the same amount of energy in the form of ionization through the first 500 μm of travel through a gas medium (McGregor et al., 2005). Although fission fragments deposit only a small portion of their energy in the first 500 μm of travel, this energy can be easily distinguished above background signals in a high gamma ray exposure (McGregor et al., 2005). Shown in Fig. 2 are the ionization curves of two common fission fragments over their first 500 μm of travel. The total integration of the energy deposited by the 60 MeV iodine ion, and the 95 MeV bromine ion are approximately 3.0 MeV and 2.9 MeV respectively within the 500 μm cavity (McGregor et al., 2005).

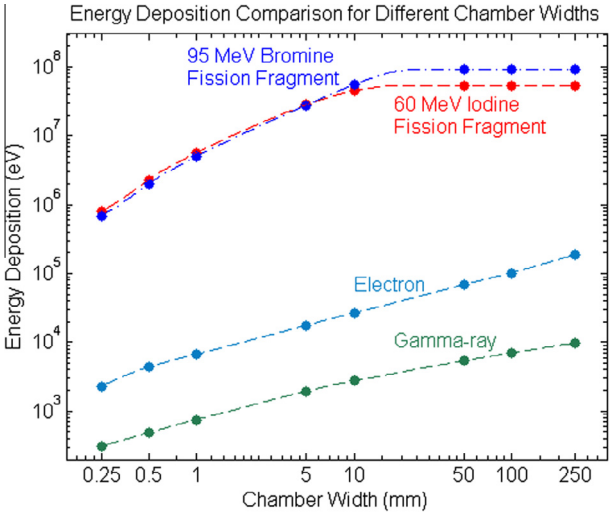


Fig. 1. Fission fragment ionization in small gas chambers. The ionization from fission fragments is >100 times greater than electron or gamma-ray ionization for the small MPFD gas chambers (Ohmes et al., 2006).

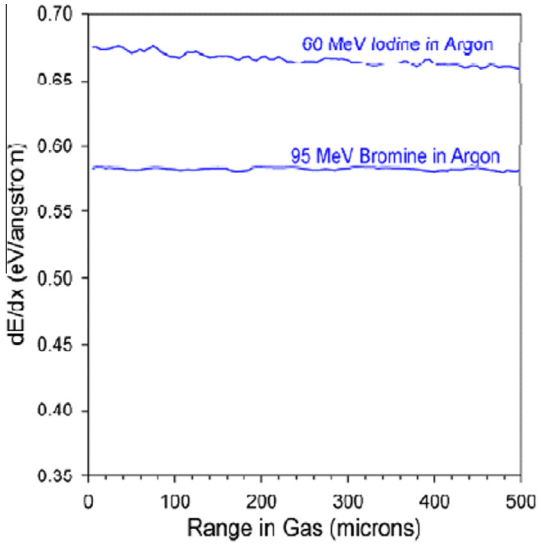


Fig. 2. Simulations using SRIM show the energy deposition of two common fission fragments in argon gas. The total energy deposited in the first 500 μm of travel is represented as the area under each curve.

Prototype MPFDs of various dimensions have been constructed and tested (McGregor et al., 2005; Ohmes et al., 2004). Preliminary results demonstrated excellent gamma-ray discrimination during beam port testing in an average neutron flux of 1.6×10^6 n cm^{-2} s^{-1} and gamma-ray background of 10^2 Rad h^{-1} (McGregor et al., 2005). However, due to the large size of the prototype device (3 mm diameter micro-pocket) and enriched neutron conversion material, the detector suffered from significant dead time (up 24% at a neutron flux near 5×10^{12} n cm^{-2} s^{-1}) (McGregor et al., 2005). Additionally, 'triad MPFDs' capable of distinguishing between thermal-neutron, fast-neutron, and gamma-ray flux were designed and simulated (Ohmes et al., 2004). Finally, by optimizing the neutron-conversion material composition (mixing fissile and fertile materials), stable detector response can be achieved for high neutron fluences (Reichenberger et al., 2014). Although the prototype detectors did show favorable characteristics (Böck and Balcar, 1974; Blandin et al., 2003), and simulations indicated that MPFDs could be used to extract real-time data for in-core testing (Ohmes

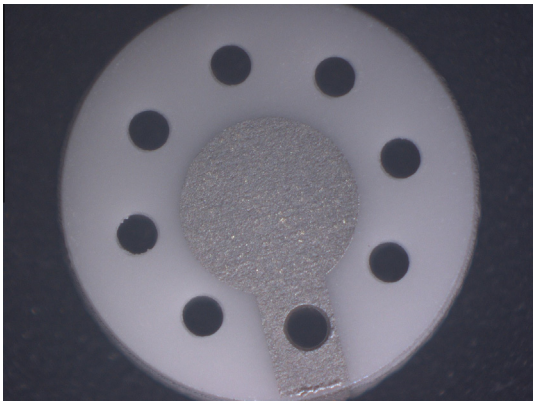


Fig. 3. An optical microscope image of an MPFD substrate with a 2.0 mm diameter Pt electrode. Conductive electrodes are evaporated onto polished alumina substrates to serve as an electrodeposition surface and electrode for charge collection.

et al., 2004), little progress was made towards the repeatable fabrication and characterization of MPFDs after these initial results. Prototype detector dimensions were also too large for in-core deployment. Precise deposition and characterization of neutron conversion material is necessary in order to deploy MPFDs in test reactors (Reichenberger et al., submitted for publication).

A third version of the MPFD, discussed here, has been developed, in which the detector chambers are stacked. A thermocouple is included with the design for temperature measurements (Unruh et al., 2013). Substrate material selection has also become of particular interest, specifically regarding impurities (which can activate during use) and surface roughness (which affects the deposition of the neutron-conversion material). Currently, MPFDs are being developed with substrates fabricated from polished, amorphous, high-purity alumina (Al_2O_3) been machined into circular disks and spacers as shown in Fig. 3 (Unruh et al., 2013). The disks and spacers can be stacked one upon another in order to create a micro-pocket (Unruh et al., 2013). A more extensive description of previous work and current technologies has been reported elsewhere (Unruh et al., 2013).

3. Detector development

The MPFD design has been refined by combining multiple sensors into a single package small enough to deploy in MTRs (Unruh et al., 2013). In order to build such a detector, advances in neutron-conversion material deposition, measurement, and lifetime optimization were all necessary. Material deposition methods must be enhanced in order to accurately and reproducibly deposit a small amount of neutron-conversion material onto the MPFD chamber substrate. In order to predict the response rate for MPFDs, the mass of neutron-conversion material must be well-known. A combination of neutron-reactive materials can be utilized, creating a breeding MPFD chamber which extends device lifetime (Reichenberger et al., 2014). The breeding optimization was made possible by the utilization of ORIGEN-S and NEWT software packages (Laboratory, 2009). By combining accurate electrodeposition and measurement of neutron-reactive materials with a numerical optimization of a mixed neutron-reactive coating, an optimal MPFD was designed which maximizes stable device lifetime.

3.1. Neutron-conversion material electrodeposition

Before a neutron-conversion material is deposited onto the surface of an MPFD chamber, a thin metallic layer must be deposited to provide a conductive surface for electrodeposition. Presently, an electron-beam evaporator is used to evaporate 50 Å of titanium (an adhesion layer) and 500 Å of platinum onto the surface of a high-purity, clean, polished, amorphous alumina sample. Platinum is used because of it high melting point, inert characteristics, and low neutron absorption cross-section. Shadow masks are used to evaporate electrodes of the desired shape onto the alumina substrate. Electrodes as small as 0.33 mm in diameter have been successfully evaporated and coated with neutron-conversion materials (Reichenberger et al., submitted for publication).

Although numerous methods exist for deposition of neutron-conversion material onto a conductive surface, electrodeposition was chosen as the method to deposit the neutron reactive materials (uranium and thorium) onto a detector substrate (Hao et al., 2010). Numerous deposition methods have been reported, but apparently were not well characterized (Ohmes et al., 2004). However, electrodeposition has been determined to be a reliable and repeatable method of deposition (Reichenberger et al., submitted for publication). The cyclic potential sweep process utilizes a platinum working electrode and a silver/silver-chloride reference electrode and has been successfully used to coat small amounts of natural uranium (shown in Fig. 4) and thorium upon the conductive electrodes (Reichenberger et al., submitted for publication).

3.2. Neutron-conversion material measurement

Visual inspection of the electroplated surfaces was used to confirm material deposition, while X-ray fluorescence (XRF) measurements have confirmed the presence of the desired neutron-conversion material. However, neither visual inspection nor XRF measurements can provide an accurate estimate of the mass of the neutron-conversion material deposited on the surface. An alpha particle decay measurement technique was developed to estimate the total mass of the electroplated material by capitalizing on the radioactive emissions from the neutron-conversion materials (natural uranium and thorium) for the thin coatings. The accurate measurement of neutron-conversion material in MPFDs allows the calculations of the device response to different neutron fluxes. Further, knowledge of the mass and composition is necessary when mixing neutron-conversion materials to extend the stable detector response lifetime. Measurement of the neutron-conversion material was first attempted using XRF measurements. XRF was an adequate method of verifying the deposition of uranium and thorium, but did not provide sufficient information on the total mass deposited on the substrate. The desired mass for MPFD coatings varies with the expected neutron flux distribution,

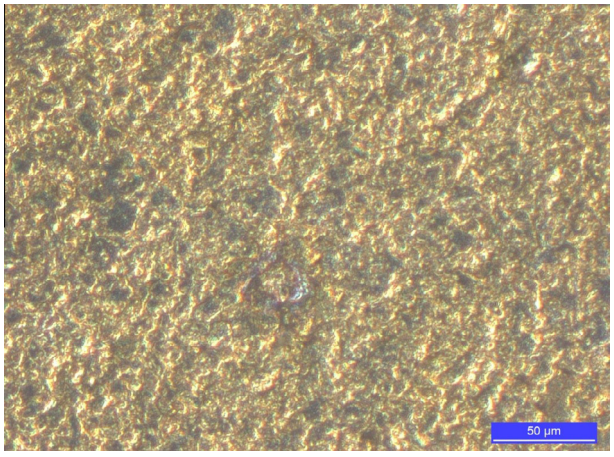


Fig. 4. Optical microscope image of a neutron-conversion material, an uranyl hydroxide layer, electrodeposited on the Pt electrode surface.

and desired device performance. In general however, coating thicknesses of <350 μg/cm^2 are desired for most applications, based on preliminary calculations. Small masses are desired in order to *reduce* the neutron-detection efficiency of the MPFDs. A very low interaction rate is necessary to reduce the effects of dead time in the detector because MPFDs will be used in neutron fluxes exceeding 10^{12} n cm^{-2} s^{-1}. The desired mass of uranium or thorium deposit determines the limitations and error allowable (<10%) imposed upon the measurement system. Imaging techniques have not been successful primarily due to the thin nature of these coatings and the surface roughness of the sample. Most imaging techniques also require destruction of the device to view a cross-section of the surface. However, measurement of the natural radioactivity emitted from the samples has proven to be an effective method of estimating the deposition mass.

Natural uranium and thorium radioactively decay by alpha particle emission. In secular equilibrium, natural uranium has a specific activity of 2.6×10^{-8} TBq g^{-1}, and thorium has a specific activity of 8.14×10^{-9} TBq g^{-1} (Department of Transportation Report, 2011). By measuring the activity of an electrodeposited sample, the mass of the radioactive material can be estimated by dividing the activity by the specific activity of the material which was electrodeposited. Uranium and thorium are likely present as their hydroxides on the surface of the MPFD samples (Reichenberger et al., submitted for publication); however only the mass of uranium or thorium is pertinent to the measurement of neutrons in this instance. These alpha particle measurements have the unexpected advantage of only providing the mass of the neutron-converting material. A low background silicon surface barrier (SSB) alpha particle spectrometry system was used to measure the test samples. Background, solid-angle, and alpha-particle backscatter effects were all considered in the mass calculation. Forty-eight (48) hour background measurements were taken intermittently during the procedure to account for fluctuations in background; however these measurements did not suffer from significant background fluctuations during the measurement periods. Alpha-particle backscatter effects were determined by analog MCNP6 simulation of the test environment, and were found to be negligible (≈0.03%). The alpha-particle emission measurement for the mass estimation also makes possible the characterization of the electrodeposition methods described elsewhere (Reichenberger et al., submitted for publication). The characterization process for electrodeposition of uranium and thorium masses less than 1 μg requires an extensive measurement time to ensure proper counting statistics. Typical samples must be measured for over 48 h in order to achieve 5% measurement accuracy because of the low specific activity and low mass of the deposits. After measuring the mass of neutron-conversion material deposited on numerous samples, the amount of material deposited onto an electrode by electrodeposition has been shown, as expected, to have a strong relationship with surface area of the electrode. Using a similar electrodeposition method described in greater detail elsewhere (Reichenberger et al., submitted for publication), as much as 5.24 ± 0.27 μg of uranium, and 4.92 ± 0.24 μg of thorium have been deposited on samples with 2 mm electrodes. In comparison, only 0.19 ± 0.02 μg of uranium and 0.55 ± 0.05 μg of thorium have been deposited on samples with 0.33 mm electrodes and 750 potential segments at a sweep-rate of 0.01 Vs^{-1}. Efforts are presently underway to characterize thorium electrodeposition from 0.1 μg to 0.3 μg on 0.33 mm diameter electrodes.

3.3. Neutron-conversion material lifetime calculation

Previous studies predicted the response rate of various neutron-conversion materials used in MPFDs (Ohmes et al., 2006) and other fission chamber designs (Jammes et al., 2012; Filiatre et al., 2008; Cabellos et al., 2010). In order to design MPFDs for long-term, in-core deployment, the reactive material must be chosen carefully to ensure a relatively stable response over long periods of time. Although many neutron-reactive materials may be used for an MPFD, a combination of uranium and thorium isotopes has been proposed for use as the reactive material (Reichenberger et al., 2014). Other nuclides have also been studied for fission chambers, particularly for long-term monitoring of the fast flux (Filiatre et al., 2008; Cabellos et al., 2010). By determining an optimum combination of nuclides, the signal deviation over time (for a constant total flux) can be minimized (Reichenberger et al., 2014).

A basic MPFD device has a thin-film of the neutron reactive material. Because the film thickness is small compared to the film surface area, the reactive material can be treated as infinitely dilute. Hence, spatial self-shielding effects are negligible, consistent with previous studies (Filiatre et al., 2008), and each isotope of interest may therefore be treated independently. Once the time-dependent signal (per unit mass) is known for each individual isotope (e.g., ^{235}U or ^{232}Th) subject to a specific flux and spectrum, the combined signal for any initial composition of the isotopes can be found directly using the mass fraction of each isotope and the mass of the device. This simplified approach, made possible by

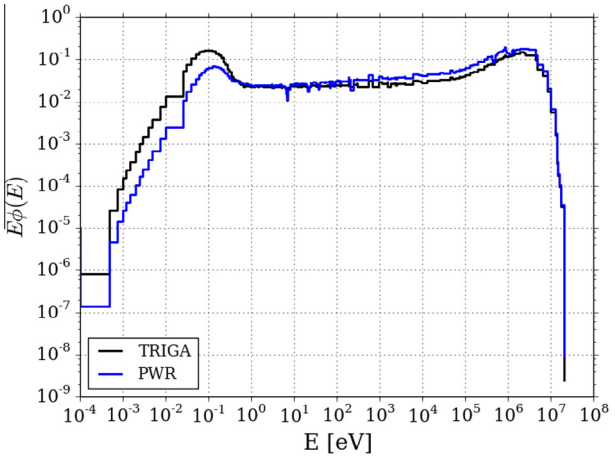


Fig. 5. Energy spectra characteristic of TRIGA and PWR reactors at nominal conditions. The TRIGA spectrum is clearly more strongly thermalized, evidenced by its stronger Maxwellian thermal peak and more nearly constant (i.e., 1/E) epithermal flux.

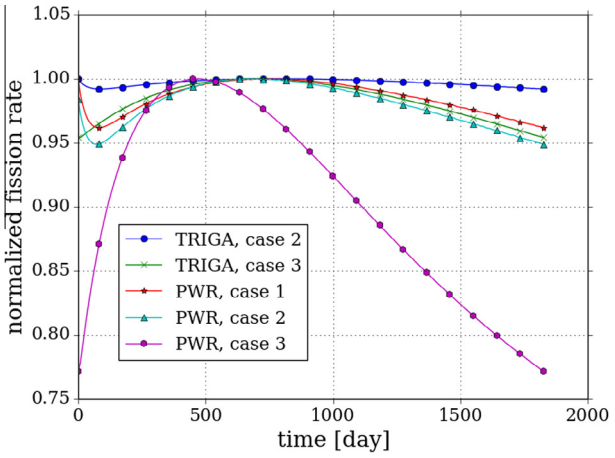


Fig. 6. Signal over 5 years in a TRIGA spectrum (3×10^{13} n cm^{-2} s^{-1} total flux) and a PWR spectrum (3×10^{14} n cm^{-2} s^{-1} total flux). Case 1 for the TRIGA is identical to Case 2.

Table 1
Optimal compositions (by weight%) and corresponding maximum relative deviation from the maximum signal for each spectrum and case.

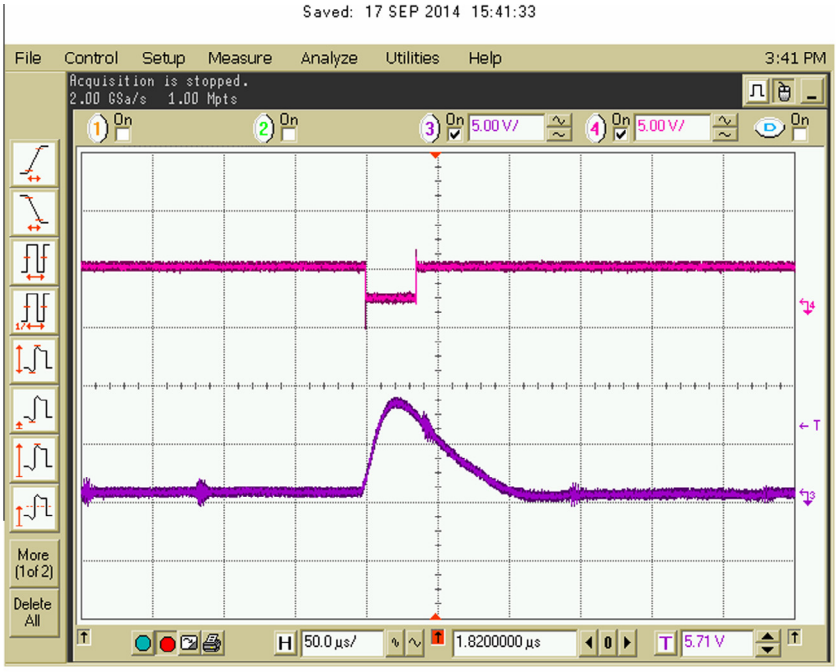
Spectrum	Case	^{232}Th	^{233}U	^{235}U	^{238}U	Max. rel. deviation (%)
TRIGA	1	85.7	0	2.7	11.6	0.8
TRIGA	2	85.7	N/A	2.7	11.6	Same
TRIGA	3	N/A	N/A	4.8	95.2	4.6
PWR	1	91.8	4.3	0	3.9	3.8
PWR	2	94.7	N/A	5.2	0	5.1
PWR	3	N/A	N/A	7.6	92.4	22.8

the extremely small dimension of the device, greatly simplifies the determination of an optimum composition.

ORIGEN-S (Laboratory, 2009) was used to compute the device signal (the fission rate) as a function of time for two different reactor types, those being a TRIGA Mk II nuclear reactor, a pressurized water reactor (PWR), along with different combinations of thorium (^{232}Th) and uranium (^{233}U, ^{235}U, and ^{238}U). ORIGEN-S allows the user to create problem-specific, cross section data by weighting the built-in, unshielded cross sections with an appropriate energy spectrum. Here, the spectra were computed using NEWT (Laboratory, 2009) for representative, square-pitch fuel elements with nominal thermal conditions, as shown in Fig. 5. Notably, the TRIGA Mk II nuclear reactor spectrum as computed here differs substantially from the spectrum assumed in previous scoping studies, which severely under-predicted the epithermal flux and led to different optimum compositions (Reichenberger et al., 2014).

Constant total neutron fluxes of 3×10^{13} and 3×10^{14} cm^{-2} s^{-1} were assumed for the TRIGA Mk II nuclear reactor and PWR configurations, respectively, over a five-year period. For a PWR, the resulting fluence is approximately equal to three cycles, while for a TRIGA Mk II nuclear reactor (often operated only a part of the time), the resulting fluence likely corresponds to an operational period much longer than 5 years. Example signals are shown in Fig. 6 for optimal compositions limited to the following three cases (1) all four isotopes, (2) combinations of ^{232}Th, ^{235}U, and ^{238}U, and (3) only a combination ^{235}U and ^{238}U for each reactor. For the TRIGA Mk II nuclear reactor, cases (1) and (2) led to identical compositions and only case (2) is shown. The optimal compositions are given in Table 1 along with the corresponding maximum relative deviation from the maximum signal. The deviations for the TRIGA Mk II nuclear reactor are smaller than those for the PWR, due in part to the much lower total fluence.

The results indicate that the maximum signal deviation is a strong function of composition (both uranium fraction and enrichment), and that the optimal composition is a strong function of the flux spectrum. Compositions based on ^{235}U and ^{238}U alone are insufficient for long-term stability. The addition of ^{232}Th improves the stability substantially, due to the neutron transmutation into ^{233}U. A slight further improvement is observed for the PWR case if ^{233}U is also allowed in the initial composition, but the commercial availability of ^{233}U is not known.



Acquisition	Sampling mode real time Normal Memory depth automatic 2050000 pts Sampling rate automatic Sampling rate 2.00 GSa/s Averaging off Interpolation on
Channel 3	Scale 5.00 V/ Offset 9.140 V Coupling DC Impedance 1M Ohm
Channel 4	Scale 5.00 V/ Offset -7.200 V Coupling DC Impedance 1M Ohm
Time base	Scale 50.0 µs/ Position 1.8200000 µs Reference center
Trigger	Mode advanced Sweep single Advanced violation Type Pulse Width Direction width > Width 20.0035 µs Sensitivity normal Holdoff time 50 ns Source channel 3 Trigger level 5.71 V Polarity pos

Fig. 7. Ionization within the MPFD chamber was collected and produced the analog signal, the bottom signal. A TTL signal was also produced for fast counting, the top signal.

Efforts are underway to understand the effect of time-varying power levels on the time-dependent signal and to model these and depletion effects to provide corrected, real-time flux maps. As part of that effort, a detailed study of the uncertainty in the detector response due to uncertainties in the underlying cross-section data is necessary. Previous work has shown that such uncertainties can have magnitudes close to the predicted signal deviations and, hence, must be thoroughly assessed (Cabellos et al., 2010).

4. Detector testing

MPFD development and testing are underway. A 2-chamber prototype was constructed using two 2 mm diameter, 1.5 mm tall chambers; one chamber was electroplated with 5.24 ± 0.27 μg of natural uranium, while the other was electroplated with 4.92 ± 0.24 μg of thorium.

4.1. Test MPFD assembly

The 2-chamber MPFD was assembled at Idaho National Laboratory (INL). MPFD construction was completed using equipment at INL's High Temperature Test Laboratory (HTTL). The unique design of the MPFD requires several construction steps typically not required for conventional in-pile sensor fabrication. Specifically, specialized techniques have been developed that emphasize robustness (e.g., minimizing the number of wire splices and component embrittlement associated with welding). The main components of the MPFD and extension cable are specially manufactured hard-fired alumina substrates and crushable alumina insulators, respectively, all housed within leak-tight tubing. A "loose assembly construction" is used with a rigid tube that contains hard-fired alumina substrates for the fission chambers, a thermocouple, and wire contacts. Stainless steel tubing was used for the prototype MPFD, however other materials can be easily adapted using similar construction techniques. The loose assembly is laser welded to a six conductor extension cable fabricated from crushable alumina insulators and wire. The extension assembly is drawn to the desired length and diameter to provide flexibility for installation in the reactor tank. The final step to insert the fill gas, typically argon, is performed by sealing a portion of the loose assembly in specialized inert gas fixture at the HTTL prior to performing the final laser weld.

4.2. Test MPFD results

After insertion into the central thimble of the KSU TRIGA Mk II nuclear reactor, neutron pulses were produced above noise and background as shown in Fig. 7. The fission-fragments emitted as a result of a neutron interaction in the uranium coated MPFD electrode produced ionization in the MPFD chamber. Using a 100 V applied bias across the 1.5 mm chamber, a large (almost 10 V) pulse was observed after filtering and amplification with specialized electronics.

5. Summary

The stackable MPFDs have been designed, built, and successfully demonstrated as in-core neutron sensors. An MPFD with 1.5 mm thick chamber was successfully tested at the KSU TRIGA MkII nuclear reactor. Thin conductive electrodes were evaporated onto insulating MPFD substrates. A cyclic potential sweep process was used to deposit neutron-conversion materials. Alpha-particle counting was used to measure the deposited mass for each MPFD.

Improvements to the MPFD design and manufacturing processes will enhance the producibility and effectiveness of these flux-monitoring neutron detectors. Current progress will characterize the electrodeposition method for uranium and thorium in order to improve the accurate manufacturing of MPFDs. With continued development, MPFDs will prove a valuable data acquisition tool for high-neutron-flux environments in the coming years.

Acknowledgments

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