

Aluminum Nitride as an Alternative Local Power Range Monitoring Technology

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Abstract—This paper provides an overview of the performance of an aluminum-nitride ceramic-compact based solid-state flux monitor (SSFM). The sensor is intended to operate as a local power range monitor in-core and provide immediate response as well as a larger signal level compared to self-powered neutron detectors. The sensor was developed as part of an International Nuclear Energy Research Project between the United States and the Republic of Korea. The sensor essentially consists of polycrystalline aluminum-nitride ceramic compact configured as a flux sensitive resistor. Neutrons interact through the $^{14}\text{N}(n,p)^{14}\text{C}$, producing free electrical carriers as the daughter particles slow down within the sensor. Bias voltage is applied to sweep the free carriers through the compact, resulting in a current proportional to the ionizing dose deposited in the sensor body. The ceramic compact is a small (3-mm² by ~0.5-mm) high electrical resistance chip with direct-bonded copper contacts on opposing sides, resembling a parallel plate capacitor. The device sensitivity is designed to be low enough to endure in-core, at-power conditions yet high enough to readily observe power level changes. The sensor's measured performance is similar to that expected theoretically. However, the current generation of devices exhibits a significant polarization under bias. While the sensor is designed to function in-core, at power for years, this has not yet been demonstrated. Significant additional development remains before AlN flux monitors can be considered for deployment in operating power reactors.

I. BACKGROUND

Local power-range flux monitoring is desirable for any power reactor both to monitor normal flux profile as well as to observe undesirable phenomena, ranging from boric acid plate out to axial power offset anomalies. Solid-phase semiconducting materials have not generally been considered as candidates for transducers in-core because the electronic transport properties of crystalline solids are much more susceptible to radiation damage than the gases employed in fission counters. Highly polycrystalline, refractory solids, in contrast, are (in terms of electronic transport properties) much more radiation tolerant than single crystal devices. Small grain size, refractory polycrystalline solids begin in a disordered state on a long-range scale and so are not strongly vulnerable to radiation-induced disordering. Refractory ceramics are also very chemically stable. Thus, essentially identical compounds reform

following radiation-produced bond breakage. Consequently, both ionizing and displacement doses are highly likely to result in functionally identical material.

The $^{14}\text{N}(n,p)^{14}\text{C}$ reaction has previously been employed in a pancake type ion chamber in a high-flux neutron beam scattering beam line.ⁱ Also, the concept of using the change in the electrical resistance of a neutron converter-containing ceramic compact as a flux monitor was initially explored 30 years ago.ⁱⁱ However, it is thought that the project and results outlined here represent the first attempt to combine the two concepts.

II. DESIGN AND OPERATING PRINCIPLES

The central idea of the solid-state flux monitor is to use an aluminum-nitride-based ceramic compact as a flux sensitive resistor. Simply put, a piece of aluminum-nitride compact has electrical contacts

made to its surface, voltage is applied to the contacts, the device is placed into a neutron field, and the measured electrical current is the signal. Figure 1 illustrates the situation.

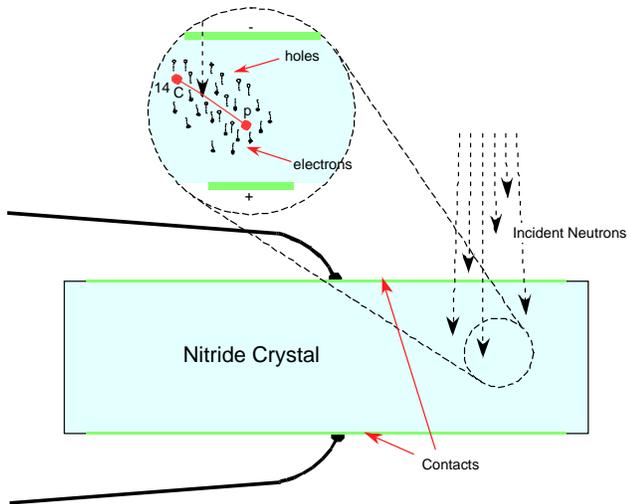
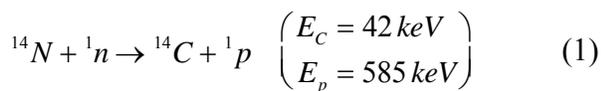


Figure 1—Conceptual layout and physical arrangement of a solid-state flux monitor

The sensor body is formed from sintered AlN powder with a small amount of yttrium oxide added as a binder and oxygen getter. AlN has a high electrical resistivity at light-water-reactor relevant temperatures, resulting in negligible thermally induced current. Also, all of the Group III nitrides exhibit high free carrier mobilities, allowing the charges to move under the applied bias resulting in a current pulse.

Nitrogen-14, which composes 99.6% of naturally occurring nitrogen, has a significant (1.769 b at 25.3 meV) inverse neutron velocity proportional energetic proton production cross-section. The reaction proceeds as shown in equation (1).



The ${}^{14}\text{N}$ capture reaction has a Q-value of 627 keV. Aluminum nitride has a band-gap of 6.28 eV. A useful rule-of-thumb is that a free carrier is produced for every three fold of the band-gap deposited into semiconductors by an energetic electron. Protons, because of both their greater ionization density along the slowing down track as well as the increased fractional expenditure of energy in atomic rearrangement, are generally considered to be roughly one-third as efficient in

generating free carriers as electrons. Roughly speaking, therefore, each neutron interaction within aluminum nitride can be expected to generate 10,000 free carriers. For a neutron flux of $10^{14}/\text{cm}^2\text{-s}$, a 1 mm x 1 mm x 1- μm piece of aluminum nitride with a density of 3.23 g/cm^3 would be expected to generate $8.7 \times 10^{10} \text{ e}^-/\text{s}$. Provided that these generated carriers move readily across the sample, which appears likely due to the high electron mobility ($\sim 100 \text{ cm}^2/\text{V-s}$), this would produce a current of $3.5 \mu\text{A}$ for a 250- μm thick sample. The annual burn-up at power for such a device would be roughly six parts per thousand.

One potential concern about this device is that the intense gamma flux within the reactor will also produce a response that may be of the same magnitude as the neutron response. While in general the gamma flux at power in a nuclear reactor core is itself considered a measure of overall reactor power and hence may not be considered parasitic, it may be necessary to compensate for the gamma response of the device at neutron fluxes. This is technologically possible in much the same fashion as in a compensated ion chamber. Nitrogen-15 does not possess the thermal neutron absorption cross section of ${}^{14}\text{N}$. Hence, an otherwise identical device composed of ${}^{15}\text{N}$ -based aluminum-nitride would be expected to respond just as a ${}^{14}\text{N}$ aluminum-nitride sensor, but without the low-energy neutron sensitivity. Therefore subtracting the currents from the two devices would yield the sensor current due to the thermal neutron flux.

III. DEVICE FABRICATION AND PACKAGING

The transducer bodies were obtained in sheet form (381 μm and 635 μm thick) from Curamik[®] Electronics, Inc., (Toshiba Corporation manufactured substrate.) The AlN sheets were laser scribed and then mechanically broken into 3-mm x 3-mm squares. The electrical contact pads were applied to opposite side of the chips by direct copper bonding. Direct bonded copper (DBC) contacting technology for metallizing aluminum-nitride ceramic surfaces has been developed over the past decade in support of the semiconductor industry.ⁱⁱⁱ The direct copper bonding process starts with applying a copper sheet of the desired thickness to the surface of the aluminum-nitride ceramic. A liquid shell is then formed around the copper sheet by heating under a reducing atmosphere-resulting in

a copper-oxygen eutectic melt on the surface of the aluminum nitride that wets and bonds to the ceramic. Patterning of the resultant copper layer is accomplished through standard semiconductor mask and etching techniques. To limit copper oxidation and ease wire attachment, nickel and gold overlayers (a few microns thick) were applied over the copper (127- μm thick) contact spots.

Ultrasonic wire bonding of 127- μm diameter, soft copper wire proved a successful technique for

attaching lead wires to the contact pads. Ultrasonic wire bonding uses a combination of pressure and ultrasonic heating of the wire and substrate to create a joint and can rapidly be performed in air at room temperature. Figure 2 shows the final generation prototype transducer pieces employing a 381- μm thick AlN substrate with 127- μm -thick copper contact pads overcoated with thin layers of nickel and gold with 127- μm -diameter soft copper leads ultrasonically bonded to the electrical contact pads.

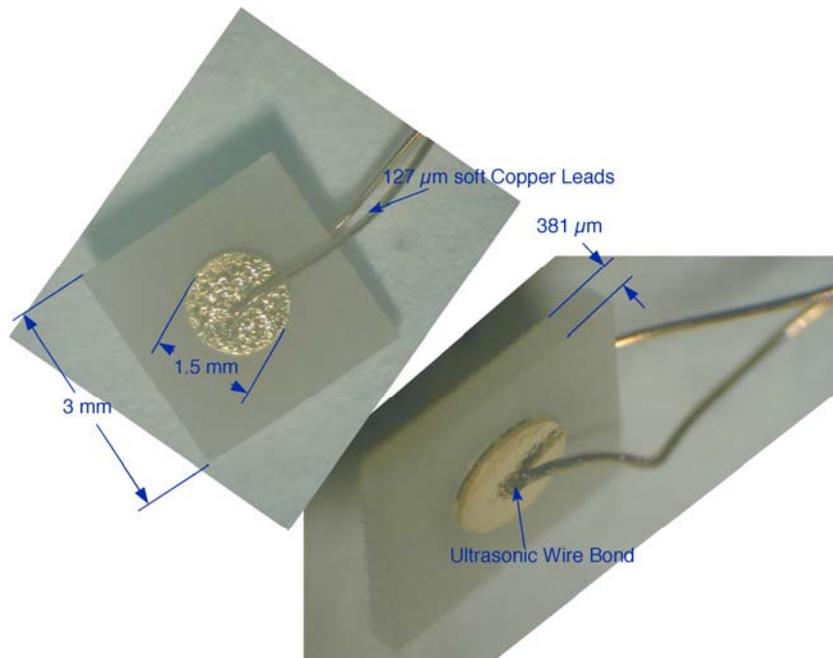


Figure 2—Bare final generation prototype SSFMs using ultrasonic wire bonding

To deploy the sensors in more aggressive environments, the bare devices needed to be encapsulated within a protective housing. Because the SSFMs are small signal devices, the housing is required to provide electromagnetic interference shielding and physical isolation from the environment in addition to mechanical protection. The sensors leads were threaded onto a short piece of dual-bore ceramic tubing (thermocouple bead) and placed within a 1-cm OD Inconel™-600 sheath that was then backfilled with MgO powder. The leads were then brazed to a two-conductor 16-m-long piece of MIMS cable from Thermocoax. Woojin, Inc., performed the encapsulation.

IV. MEASUREMENT RESULTS

Gamma Performance

A wide range of gamma doses delivered by various sources were measured as currents at the Korean Atomic Energy Research Institute (KAERI). The lowest total gamma dose rate measured was for the 59.5-keV gamma rays from a 925-MBq (25-mCi) ^{241}Am source located about 3.2 cm from the sensor (approximately 10^7 gamma rays/cm²-s) with a measured current of 1.8 pA with a leakage current of 0.5 pA. For small source measurements short cables were employed; for the larger sources ~20-m coaxial cables were used. Because no guard conductor was employed (coaxial as opposed to triaxial cables), measurements were also made

without the sensor in place, and the cable contribution to the measured signal was subtracted manually. High dose rate, gamma-induced current measurements of the SSFM were made with two different sources (6.9 TBq ^{60}Co and a 7.4 PBq ^{60}Co) and at three separate distances for each source.

The results of the high- and low-dose measurement sets are combined and plotted as Figure 3 along with a logarithmically linear fit.

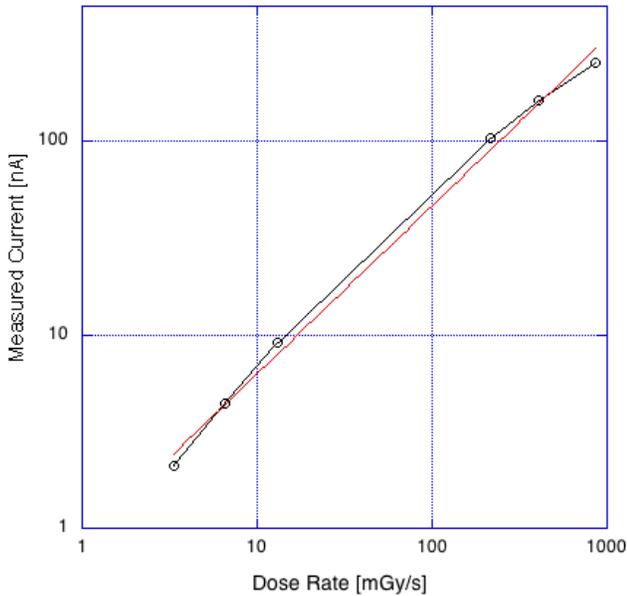


Figure 3—Measured SSFM ^{60}Co induced current and a logarithmically linear fit

Reactor Performance

The new sensors were tested both at the Ohio State University Research Reactor (OSURR) and KAERI’s Hi-Flux Advanced Neutron Application Reactor (HANARO). The OSURR is a 19.5% enriched U_3Si_2 -fueled, light-water-moderated, pool type research reactor licensed for operation up to

500 kW. Testing was performed in the OSURR main thermal column, which consists of 1.524-m-long graphite stringers aligned to point away from the core face. The nuclear-grade graphite provides a highly thermalized neutron environment. At full operating power, the total neutron flux at the test location is about 1×10^{12} nv, with temperatures at or near ambient.

HANARO is a 30-MW(t) open-tank-in-pool type reactor. It uses light-water coolant and heavy water as a reflector. It uses U_3Si_2 in an aluminum matrix fuel (19.75% enriched) with hafnium absorbers. The HANARO cold neutron source (CNS) hole was used for the high flux measurements. The neutron flux in the HANARO CNS at full power is 5.72×10^{13} nv, and the temperature is $\sim 35^\circ\text{C}$.

At the OSURR, the reactor power was first increased to full power, then reduced in increments to check linearity of device response, followed by a return to full power and a test of response to a sudden drop in power (scram). The power increments for the OSURR are 450 kW (indicated nuclear power), 250 kW, 100 kW, 50 kW, and 10 kW. The rod insertion time for the scram response is about 350 ms.

Figure 4 shows the measured sensor current. The power increments can be observed as well as the initial “ramp” increase in power, and the scram shutdown, which ended the operation. Subsequent power decay following the scram can be observed in the sensor output. The dynamic range of the sensor indicates that it would perform reasonably well as a power range monitor. The sensitivity of the SSFM operated under these conditions is about 7.9×10^{-20} A/nv.



Figure 4—Measured SSFM current in G-5 location of the thermal column of OSURR as reactor power is changed

A sealed SSFM unit supplied by Woojin, Inc., was tested for transient response to a reactor scram from full power. This response was compared to that of a conventional compensated ion chamber (CIC), which is a normal part of the OSURR reactor control system.

The OSURR was operated at full power for several minutes before the initiation of the scram. At the time of the scram, all three shim safety rods were fully inserted to drive the reactor power to shutdown levels as quickly as possible. The detector was placed in the OSURR Central Irradiation Facility (CIF) to assure that the maximum possible flux was incident upon the SSFM. Under these conditions, the OSURR thermal power is 450 kW, and the total neutron flux incident upon the SSFM before the scram was about 2.3×10^{13} nv. The total time for control rod insertion is about 300 ms.

The detector outputs were recorded using a computer-based data acquisition system. The data

capture rate was one sample per second. The computer provides a time stamp for recorded data so data capture channels can be aligned in time. Once recorded, the detector outputs were normalized to their maximum value during the data capture to allow comparison of the dynamic response of the sensors.

Figure 5 shows the results of this test. The SSFM tracks the CIC response for the initial portion of the transient, with a slightly slower response time. As the power drops to the shutdown range, the SSFM shows a higher normalized output. This likely indicates that the SSFM output includes a significant gamma component following shutdown compared to that of the CIC. This is not unexpected as the CIC includes electrical subtraction of the gamma component of the signal. The SSFM, at this point in its development, does not have this feature.

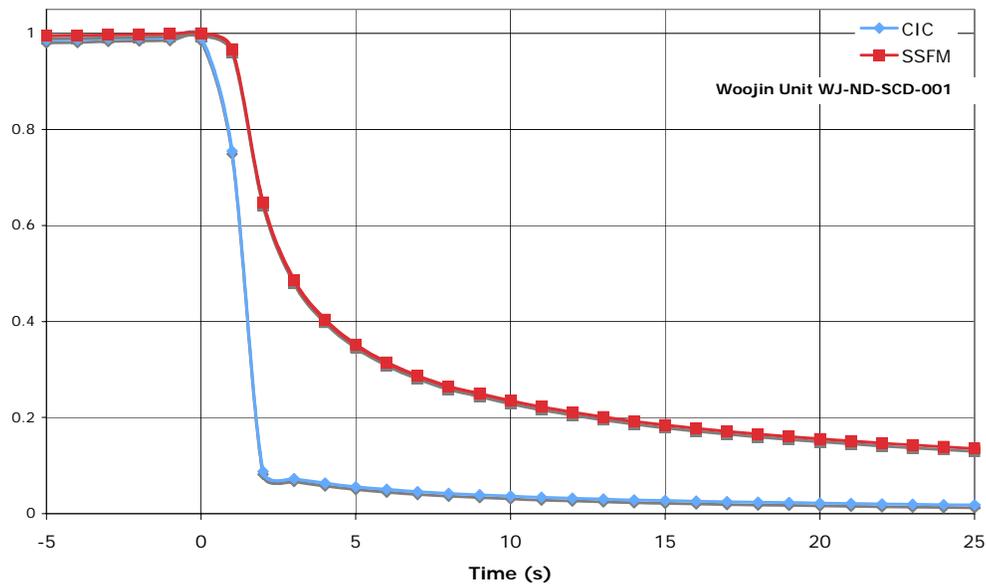


Figure 5—SSFM SCRAM response compared to OSURR CIC

Temperature vs resistance measurements were made outside of a radiation environment to ascertain what fraction of the measured SSFM signal would be due to thermal leakage current. Below 475 °C the device maintains very high resistivity, and consequently little thermal response was anticipated.

The temperature response of the SSFM was measured in-core using an attached thermocouple under steady state conditions at the OSURR CIC. The OSURR was operated at full power for several minutes to observe the stability of the SSFM output signal as the detector experienced heating by radiation absorption.

The CIC, SSFM, and thermocouple detector outputs were captured at 1-s intervals for several

minutes. Figure 6 shows the results of this test. The SSFM output drifts upward over time, resulting in an artifact in the measured current. The apparent reactor power trends upward as measured by the SSFM, although the CIC shows the reactor power to be steady. The increase in the SSFM measured current ceases at roughly the same time that the temperature rise stops. It remains unclear whether the SSFM is responding to its internal temperature rise or another time or external temperature-dependent phenomenon such as increased leakage current in the device packaging with increased temperature. It is recommended to pursue further testing employing nonnuclear heating of packaged and unpackaged devices to isolate the cause of the drift.

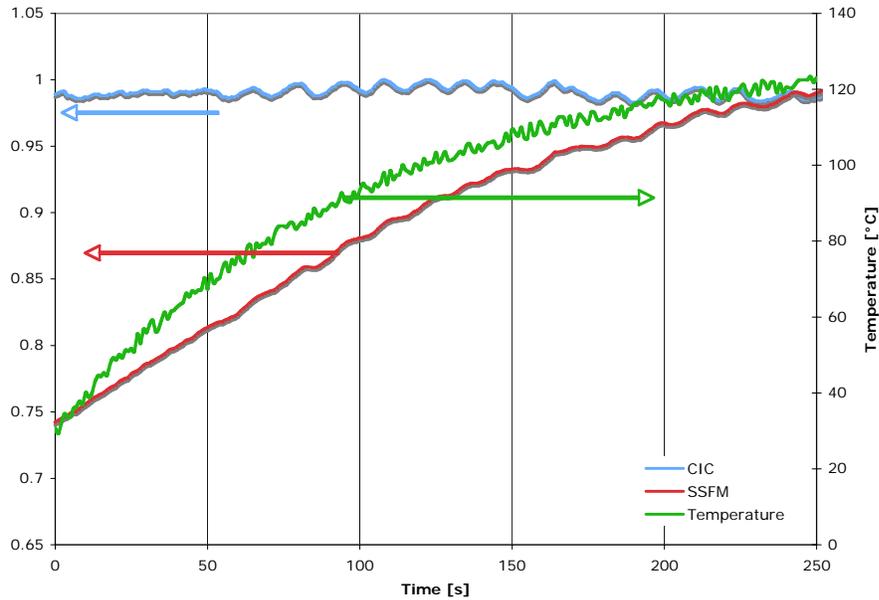


Figure 6—CIC and SSFM response changes and SSFM temperature change

A packaged 381- μm thick SSFM was deployed in the HANARO CNS, and its generated current was recorded as the reactor maneuvered in power. The SSFM was packaged in a capsule with a pair of 16-m-long mineral-insulated signal cable attached and inserted into an aluminum guide pipe and sleeve. The deployed sensor resistance, including cable leakage, was greater than 5 T Ω .

The detector current was measured while the reactor power is being increased from 1 MW to 8 MW, held steady at 8 MW for an hour, increased from 8 MW to 13 MW, and finally the reactor was tripped. The measurement results are shown as Figure 7. The current produced by the SSFM varied linearly with the reactor power over the reactor power range used (0% to 43% full power).

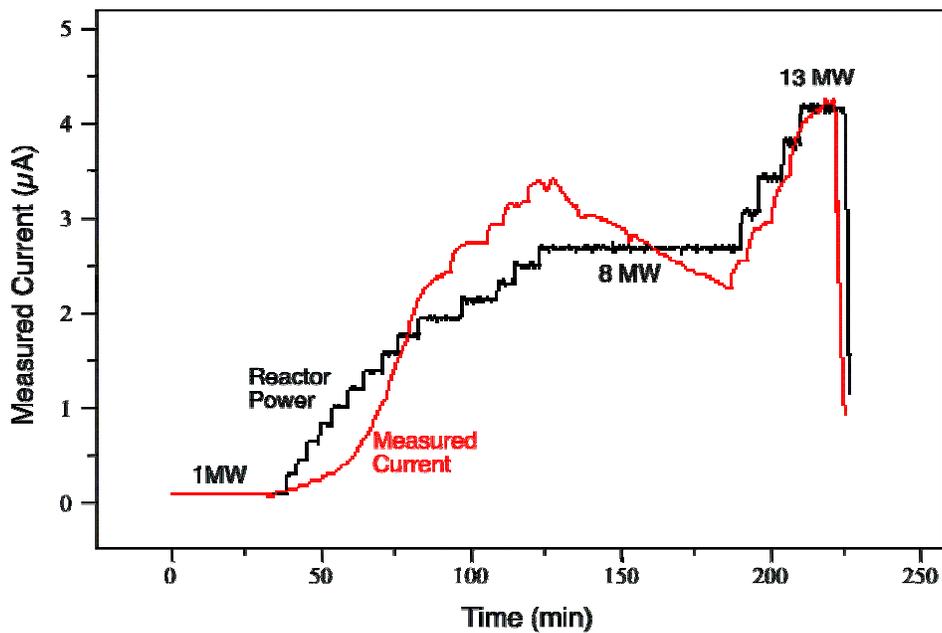


Figure 7—Correspondence of SSFM current measurement with HANARO power

The reason for the delayed increase in the measured current as the reactor begins to ascend in power may be the low voltage applied to the detector. The devices respond more rapidly when higher voltages (3000 V) are applied to them. However, only 1000 V could be applied using the cabling available in the reactor. It is speculated that the decrease in the measured current while the reactor power held constant at 8 MW may reflect device polarization; impurity ions inside the detector body migrate under the applied field and form a dead region whose size increases with time.

V. CONCLUSIONS

A novel solid-state sensor for measuring dose and neutron flux in-core, at power in nuclear reactors was conceived, designed, fabricated, and tested. The basic performance characteristics of the sensor have been established. The transducer essentially consists of an AlN sintered compact with DBC contact pads and ultrasonically bonded copper lead connections. This is the first known use of a solid-state resistive type device as a flux/dose monitor under in-core nuclear reactor type environments.

The measured device performance parameters are similar to those expected theoretically. However, several undesirable device characteristics remain and further development would be required to implement this device as intended into nuclear power plants. The stability of the device requires considerable further investigation as device polarization appears to be significant using the current biasing scheme. Also, an improved high-temperature design, reducing the leakage current, is required to implement the device at core temperatures above those of water reactors. Further, the long-term survival of the devices under in-core environments remains unknown. Transmutation doping of the AlN with the ^{14}C resulting from the neutron capture requires particular attention. In general, device repeatability and consistency needs to be further investigated because this project has not thoroughly evaluated the myriad possible device and cabling configuration variations. Finally, gamma compensation, substituting ^{15}N (lacking the neutron capture cross section) for ^{14}N in the sensor body should be demonstrated. This would establish a purely neutron flux measurement technique analogous to a compensated ion chamber.

VI. REFERENCES

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