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ABSTRACT

Ultrasonic technologies offer the potential for high accuracy and resolution in-pile measurement of a range of parameters, including geometry changes, temperature, crack initiation and growth, gas pressure and composition, and microstructural changes. Many Department of Energy-Office of Nuclear Energy (DOE-NE) programs are exploring the use of ultrasonic technologies to provide enhanced sensors for in-pile instrumentation during irradiation testing. For example, the ability of

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small diameter ultrasonic thermometers (UTs) to provide a temperature profile in candidate metallic and oxide fuel would provide much needed data for validating new fuel performance models. These efforts are limited by the lack of identified ultrasonic transducer materials capable of long term performance under irradiation test conditions. To address this need, the Pennsylvania State University (PSU) was awarded an Advanced Test Reactor National Scientific User Facility (ATR NSUF) project to evaluate the performance of promising magnetostrictive and piezoelectric transducers in the Massachusetts Institute of Technology Research Reactor (MITR) up to a fast fluence of at least 10^{21} n/cm^2. A multi-National Laboratory collaboration funded by the Nuclear Energy Enabling Technologies Advanced Sensors and Instrumentation (NEET ASI) program also provided initial support for this effort. This irradiation, which started in February 2014, is an instrumented lead test and real-time transducer performance data are collected along with temperature and neutron and gamma flux data. The irradiation is ongoing and will continue to approximately mid-2015. To date, very encouraging results have been attained as several transducers continue to operate under irradiation.

Key Words: In-Core, Ultrasonic Transducers, Piezoelectricity, Magnetostriction

1 INTRODUCTION

An effort has been initiated by the Department of Energy-Office of Nuclear Energy (DOE-NE) to characterize the performance of candidate nuclear fuels during irradiation; especially in Material Test Reactor (MTR) tests used to qualify candidate new fuels. Ultrasonic measurements have a long and successful history of use for materials characterization, including detection and characterization of degradation and damage [1] as well as measurement of various physical parameters used for process control, such as temperature and fluid flow rate [2] and are used extensively in non-destructive evaluation (NDE). Although there are numerous types of ultrasonic sensors for measuring different properties of interest, all of these ultrasonic sensors incorporate a transducer; which can limit the survivability of such sensors in an irradiation test. The development of ultrasonic tools to perform a variety of in-pile measurements requires a fundamental understanding of the behavior of ultrasonic transducer materials in high-radiation environments. While a number of irradiation studies of ultrasonic transducers have been described in the literature, a one-to-one comparison of these studies is difficult, as the materials and test conditions often differ. In addition, the tests to date are generally at lower flux/fluences than what might be seen in US Material Testing Reactors (MTRs).

A Pennsylvania State University (PSU) -led effort for an ultrasonic transducer irradiation was selected by the Advanced Test Reactor National Scientific User Facility (ATR NSUF) for an irradiation in the Massachusetts Institute of Technology Nuclear Research Reactor (MITR). This test is an instrumented lead test, allowing real time signals to be received from the transducers. The test is unique because is the first irradiation to include both piezoelectric and magnetostrictive transducers and because exposes transducers to higher fluences than prior irradiations. This test enables accurate measurement of the degradation of candidate transducer materials under irradiation. As discussed in this paper, the test has also been designed to provide fundamental data on piezoelectric and magnetostrictive material performance in irradiation environments; hence, this data can be more easily compared to results of prior irradiations.

2 BACKGROUND

Several US DOE-NE programs are investigating new fuels and materials for advanced and existing reactors. A primary objective of such programs is to characterize the irradiation performance of these fuels and materials. The key parameters needed to evaluate performance which could potentially be measured ultrasonically, as well as the desired accuracies and resolutions, are shown in Table I [3]. Similar measurement parameters exist for structural material tests.

Table I. Summary of desired fuel measurement parameters for irradiation testing.

Parameter	Representative Peak Value	Desired	
		Accuracy	Spatial Resolution
Fuel Temperature	Ceramic Light Water Reactor (LWR): 1400^{o}C	2%	1-2 cm (axially) 0.5 cm (radially)
	Ceramic Sodium Fast Reactor (SFR): 2600^{o}C		
	Metallic SFR: 1100^{o}C		
	Tristructural-isotropic (TRISO) High Temperature Gas Reactor (HTGR): 1250^{o}C		
Cladding Temperature	Ceramic LWR: <400^{o}C	2%	1-2 cm (axially)
	Ceramic SFR: 650^{o}C		
	Metallic SFR: 650^{o}C		
Fuel Rod Pressure	Ceramic LWR: 5.5 MPa	5%	NA
	Ceramic SFR: 8.6 MPa		
	Metallic SFR: 8.6 MPa		
Fission Gas Release	0-100% of Inventory	10%	NA
Fuel and Cladding Dimensions and Density	Initial Length: 1 cm	1%	NA
	Outer Diameter/Strain: 0.5 cm/5-10%	0.1%	NA
	Fuel-Cladding Gap: 0-0.1 mm	0.1%	NA
	Density: Ceramic: < 11 g/cm^3; Metallic: < 50 g/cm^3; TRISO pebble/compact: 2.25 g/cm^3	2%	NA
Fuel Microstructure	Grain size,10 μm	5%	1-10 μm
	Swelling/Porosity: 5-20%	2%	NA
	Crack formation and growth	2%	10-100 μm

Table II lists selected instrumentation available for irradiation tests in various international MTRs 4, which could potentially be replaced by ultrasonic transducer based sensors. It should be noted that many of the sensors used at foreign MTRs often require enhancement before they can be successfully deployed in the higher flux, harsher test conditions typical of US MTRs. If enhanced, these sensors can provide insights with respect to parameters, such as temperature, thermal conductivity, and crack growth. However, in general, the spatial resolution available with such sensors is limited due to the limited size of the irradiation test and the desire to minimize the impact of the sensor on test results. It should also be noted that existing and near-term sensor technologies do not provide any capability for detecting changes in fuel microstructure or constituent migration.

Ultrasonic measurements have a long and successful history of out-of-pile use for measurement of various process control parameters and non-destructive evaluation, including materials characterization and flaw detection. If it can be shown that the transducers used to make these measurements can survive irradiation test conditions, all of the parameters listed in Tables I and II could potentially be monitored ultrasonically with higher fidelity than possible with currently available sensors. For high accuracy measurements, most of these applications are likely to require the high frequency operation of piezoelectric transducers, but some measurements can be made with magnetostrictive transducers as well. For example, Post Irradiation Examinations (PIEs) have shown that fuel microstructural parameters, such as porosity and grain size, can be correlated to ultrasonic velocity 5. As noted by Villard 6, frequency requirements for such measurements are typically restricted to greater than 10 MHz. However, lower frequencies can be used for some applications, such as ultrasonic thermometry, where frequency requirements may be 100 – 150 kHz or lower (such as magnetostrictive transducer based ultrasonic thermometry).

Table II. Selected Instrumentation Available in MTRs

Parameter	Sensor	Comments
Temperature	Melt Wires	Peak value, resolution limited by number of wires, Post Irradiation Examination (PIE) required
	SiC Monitor	Peak value, 100-800 ^{o}C temperature range, PIE required
	Thermocouples (Types N,K)	1100 ^{o}C maximum operating temperature, constituent migration
	Thermocouples [Doped Mo/Nb-alloy High Temperature Irradiation Resistant Thermocouples (HTIR-TC)]	1800 ^{o}C maximum operating temperature, electrical insulation degradation
	Thermocouples (Type C)	Decalibration due to transmutation caused by thermal neutron flux
Density/Displacement	Linear Variable Differential Transformer (LVDT)	Qualified to 500 ^{o}C
	Diameter Gauge	Qualified to 500 ^{o}C
Crack Initiation/Growth	Direct Current Potential Drop (DCPD) Method	Sensitive to water chemistry, accuracy limited to ~20%
Young's Modulus	Loaded Creep Specimen	LVDT based measurement, accuracy limited to ~10%

3 ULTRASONIC TRANSDUCERS

To generate and receive ultrasonic pulses and signals, two of the most commonly used technologies are piezoelectric and magnetostrictive transducers. Ultrasonic measurements using piezoelectric transducers have been demonstrated over a wide frequency range from kHz and up to GHz; however, most non-destructive examination (NDE), materials characterization, and process monitoring are performed in the range from 1-20 MHz, making piezoelectric transduction ideal. The current capabilities of magnetostrictive transducers are typically limited to operation at frequencies up to the order of 100 kHz, although recent research suggests that higher frequencies may be possible for small magnetostrictive transducers [7]. However, mechanical coupling as well as enhanced guided wave mode generation makes magnetostrictive transduction ideal for low frequency measurements, such as ultrasonic thermometry [8]. Therefore, radiation tolerant sensors which utilize piezoelectric or magnetostrictive materials are being considered as candidates for instrumentation for use in US MTR testing. As noted above, the PSU-led MITR irradiation test is unique because it will include both piezoelectric and magnetostrictive transducers.

3.1 Piezoelectric Transducers

The piezoelectric transducer design included in this irradiation test were based on research by Parks and Tittmann [9] and from early ultrasonic sensors developed at the Hanford Engineering Development Laboratory (HEDL) [10] for under-sodium viewing which shared many similar constraints with respect to thermal and neutron radiation tolerance. The transducers rely on pressure for coupling the piezoelectric element to the waveguide. Electrical contact with the piezoelectric element is also achieved through application of pressure. A backing layer behind the piezoelectric sensor provides damping and prevents excessive “ringing” of the transducer. In the current design, the backing layer material is a carbon/carbon composite. A schematic of the piezoelectric transducer design is shown in Figure 1.

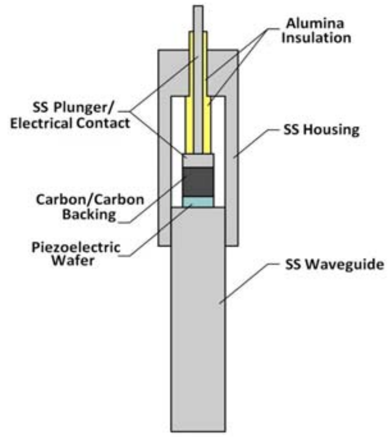

Figure 1. Schematic drawing of piezoelectric test

Due to volume limitations in this irradiation test, a limited number of piezoelectric transducer materials can be included. Piezoelectric transducer materials were selected based on prior irradiation test results, anticipated radiation tolerance, transition temperature, and ease of incorporation into sensor designs. The piezoelectric materials selected for inclusion in this test are described below.

3.1.1 Bismuth Titanate

A literature review revealed bismuth titanate as the most promising material extensively tested to date [11]. However, this material lost roughly 60% of its one way piezoelectric response at a fast neutron fluence of 10^{20} n/cm^2; suggesting that it is not an ideal candidate. The decrease in the signal response is in agreement with the statements provided in Reference [12], which indicates that disordered Ti-O-Ti bridges of highly covalent character form in titanates when subjected to neutron radiation effects. Given that this material has shown the greatest promise in prior testing, it was selected for inclusion as a baseline for comparison in this irradiation test.

3.1.2 Aluminum Nitride

Aluminum nitride is a relatively new bulk single crystal material. In fact, the work of Parks and Tittmann with this material is the first of its sort. In the past, thin film AlN has been shown to be unaffected by gamma irradiation up to 18.7 MGy [13] and temperatures of 1000 ^{o}C. [14,15] Moreover, this material has been explicitly cited in numerous independent studies as a highly radiation tolerant ceramic [12]. This is thought to be at least partially due to its wurzite crystal structure. Further, tests of bulk single crystal AlN in a TRIGA Training, Research, Isotopes, General Atomics (TRIGA) nuclear reactor core showed this material to be completely unaffected by fast and thermal neutron fluences of 1.85 x 10^{18} n/cm^2 and 5.8 x 10^{18} n/cm^2, respectively, and a gamma dose of 26.8 MGy [9]. This work, along with that of Yano [16] and Ito [17], have indicated that the 14N(n,p)14C is not of concern; and AlN was selected for inclusion.

3.1.3 Zinc Oxide

Zinc oxide, like AlN, has a wurzite crystal structure and has been cited as a highly radiation tolerant material [12]. The evaluated nuclear data files (ENDF) do not show any detrimental nuclear cross sections, and this material possesses a high transition temperature and moderate piezoelectric coupling (a measure of the efficiency in converting electrical energy to mechanical energy).

3.2 Magnetostrictive Transducers

The magnetostrictive transducer selected for this test is based on research by Lynnworth [18] and Daw [19]. The magnetostrictive transducers consist of a small driving/sensing coil, a biasing magnet, and a magnetostrictive waveguide. The ultrasonic signal is generated when a high frequency alternating current pulse is driven through the coil. The induced magnetic field causes magnetic domains within the material to oscillate. The domains are pre-biased by the magnet to maximize the response. Received echoes are detected through the reciprocal effect. A schematic of the magnetostrictive transducer design is shown in

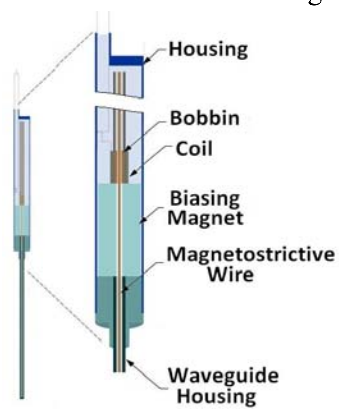

Figure 2. Schematic drawing of magnetostrictive test transducer.

Figure 2.

The magnetostrictive transducer materials were selected based on previous use in radiation environments, amounts of neutron sensitive materials, Curie temperature, and saturation magnetostriction.

3.2.1 Remendur

Remendur has the most history of use in nuclear applications of all the magnetostrictive alloys, having been used previously for short duration thermometry applications. Remendur has a high Curie temperature (950 ^{o}C) and relatively high saturation magnetostriction (~70 µstrains). Remendur is an alloy composed of approximately 49% iron, 49% cobalt, and 2% vanadium. Because of its cobalt content, Remendur was not considered to be an ideal choice (due to concerns about the production of Cobalt-60 during irradiation). However, its successful prior use was deemed sufficient reason to warrant inclusion.

3.2.2 Galfenol [20]

Galfenol is a relatively new alloy of iron and gallium (approximately 13% gallium). Galfenol is a member of the "giant" magnetostrictive alloys and has a very large saturation magnetostriction (100-400 µstrains). It also has an appropriately high Curie temperature (700 ^{o}C). Neither of its constituent elements react strongly with neutron radiation. These factors made Galfenol the most appealing magnetostrictive material candidate.

3.2.3 Arnokrome [21-23]

Arnold Magnetics produces several magnetostrictive alloys, Arnokrome 3, Arnokrome 4, and Arnokrome 5. Arnokrome 3 contains cobalt has much lower magnetostriction than Remendur and is therefore not of interest in the current study. Arnokrome 4 and 5 have similar magnetostriction to Arnokrome 3, but without the presence of cobalt. Due to space limitations and the availability of these alloys only as sheets/strips, Arnokrome was included as stand-alone samples in the test capsule but not incorporated into transducers. These samples will be evaluated in the Post Irradiation Examinations (PIE) program.

4 EXPERIMENT DESIGN

4.1 MITR

The MITR is a tank-type research reactor 24 operating at atmospheric pressure. It began operation in 1958; and its current license, issued in November 2012, authorizes steady-state 6 MW operation. The reactor has two tanks: an inner tank for light water coolant/moderator and an outer tank for the heavy water reflector. A graphite reflector surrounds the heavy water tank. The MITR is equipped with a wide variety of sample irradiation facilities, with fast and thermal neutron fluxes up to $3.6\text{x } 10^{13}$ and $1.2\text{x}10^{14}$ $n/cm^2{\cdot}s$ respectively. The ULTRA test position within the MITR core is shown in Figure 3.

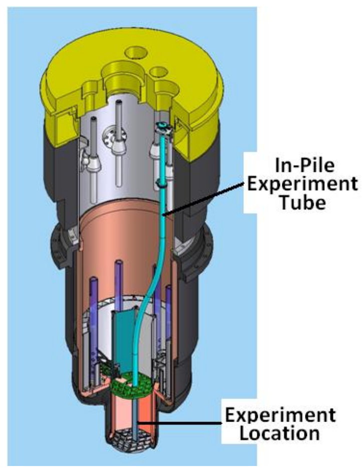

Figure 3. Schematic cutaway view of the MITR reactor showing the locations of the in-pile experiment tube and the experiment location within the core.

4.2 Irradiation Conditions

Temperature is controlled by a helium/neon gas gap with adjustable gas composition. The test temperature has ranged between approximately 400 and 450 ^{o}C. The test is designed to exceed fast neutron fluences of prior piezoelectric transducer irradiations (e.g., > $1\text{x}10^{21}$ n/cm^2 25). In order to observe rapid changes at relatively low fluences, the test was started with the reactor slowly ascending to power. Hence, it is anticipated that the capsule will be irradiated for at least 310 days. The identified irradiation position and flux conditions at the MITR are summarized in Table III.

Table III. Test conditions for MITR irradiation

MITR In-Core Experimental Facility
Capsule dimensions: 42 mm OD x 152.4 mm long
Thermal Flux: $3.6\text{x}10^{13}$ $n/cm^2{\cdot}sec$ Fast Flux (>1 MeV): $1.2\text{x}10^{14}$ $n/cm^2{\cdot}sec$ Gamma dose rate: $1\text{x}10^9$ r/hr
Temperature: 350 ^{o}C - 400 ^{o}C
Testing Period: 310 Effective Full Power Days requiring approximately 540 calendar days (18 months)

4.3 Capsule

The MITR configuration restricts the test capsule to a cylinder 42 mm in diameter and 152.4 mm in length (see Figure 4). The capsule uses structural graphite as a holder material. Graphite is an ideal material as it has low density (for reduced gamma heating). In addition, graphite is thermally conductive (to produce a uniform predictable temperature), exhibits low neutron activation, and can be used at very high temperatures. During the irradiation, the graphite holds the test specimens in place while also efficiently conducting heat generated to the coolant.

Based on space requirements for each transducer and thermal considerations, four piezoelectric samples and two magnetostrictive samples are included in the test. Additionally, stand-alone samples of each test material have been included. This will allow for PIE of samples even if the transducers cannot be dismantled after the irradiation.

An array of sensors was included in the irradiation capsule to ensure that test conditions are well-characterized. Two type-K thermocouples and melt wires are used to monitor temperatures online and verify maximum test temperature. Thermal neutron flux is monitored online using a vanadium emitter Self-Powered Neutron Detector (SPND), and thermal and fast flux will be verified using flux wires. Gamma flux is monitored online using a platinum emitter Self-Powered Gamma Detector (SPGD).

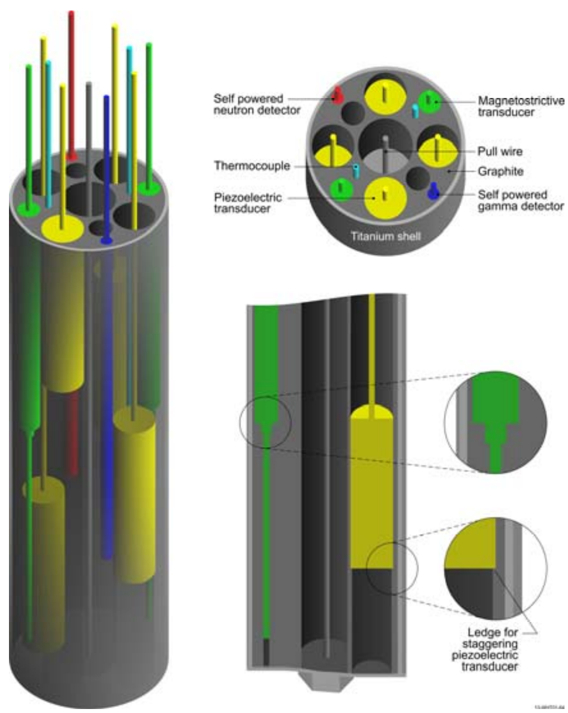

Figure 4. Test capsule showing locations of transducers and sensors.

5 RESULTS TO DATE

Two of the piezoelectric test transducers (the zinc oxide transducer and one of the aluminum nitride transducers) failed due to electrical connection issues during or just after reactor startup. Hence, real time data for these transducers was not available for analysis. The materials will be evaluated during PIE, but this may not reveal useful information if the transducer materials have degraded significantly during irradiation.

The bismuth titanate performance was characterized by measuring the change, with respect to accumulated fluence, of the normalized magnitude of the first resonant frequency of the Fourier transform of the recorded A-scan (signal amplitude as a function of time) signal. The signal was observed to decrease steadily during the early reactor cycles, up to a fast fluence of approximately $5.0*10^{19}$ n/cm^2. After this point, the transducer has shown a slow recovery, with sharp increases in amplitude during shutdowns, as seen in Figure 5a. The cause of this behavior is unclear, but may be a result of changes to the coupling between the piezoelectric element and the metallic waveguide. PIE may reveal more about whether this is a function of mechanical changes to the transducer or changes in the crystal itself.

The performance of the aluminum nitride transducer was characterized by measuring the change, with respect to accumulated fluence, of the normalized magnitude of the resonant frequency of the Fast Fourier Transform (FFT) of the first waveguide reflection observed in the recorded A-scan signal. The aluminum nitride response has been stable, with the exception of rapid drops in signal strength accompanying rapid changes in the reactor condition (i.e., rapid flux and temperature changes), as shown in Figure 5b. As with the bismuth titanate transducer, these rapid changes are likely due to sharp changes in coupling efficiency caused by the temperature changes. To this point in the irradiation, aluminum nitride appears to be a viable candidate for in-core use.

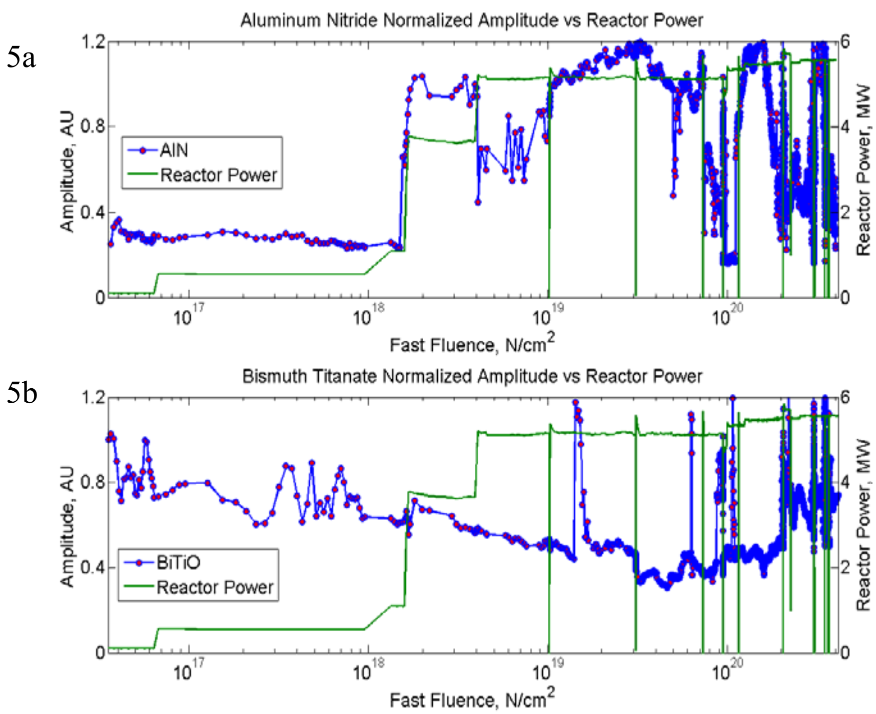

Figure 5. Piezoelectric transducer signal amplitude as a function of accumulated fast fluence.

Performance of the magnetostrictive transducers is characterized using the same method as was used for the aluminum nitride transducer. The normalized magnitude of the FFT of the first waveguide reflection signal was used to track the change in signal strength (normalized to the signal when the reactor reached full power). The frequency transformed signal is used because it is less sensitive to the interference effects of noise and signal transients. The same method was used for the Galfenol transducer.

The Galfenol transducer has shown very stable operation over the course of the irradiation, though the total peak to peak signal amplitude is typically on the order of one third of that observed for Remendur. Figure 6a shows the normalized peak to peak amplitude for the Galfenol transducer as a function of accumulated fluence. The green trace shows the reactor power history. The Galfenol transducer shows steady operation during periods when the reactor power level was stable. There is little decrease in the signal strength over these periods. The decreases in signal strength observed when reactor power is increased appear to be due to increases in operating temperature, as the signal strength stabilizes shortly after each power increase. Increased noise after the reactor was restarted after refueling may indicate an intermittent short in the drive/sense coil, likely caused during removal of the in-core tube during refueling.

Figure 6b shows the normalized amplitude for the Remendur transducer as a function of accumulated fluence. The green trace corresponds to the reactor power history. There is a general decreasing trend, but signal recovery after temperature transients indicate that some of the signal attenuation is due to temperature effects, in this case binding of the wire against the coil bobbin (see Figure 2 for transducer component diagram). As with the Galfenol transducer, increased noise after the first reactor restart after refueling may indicate an intermittent short in the drive/sense coil.

6a

6b

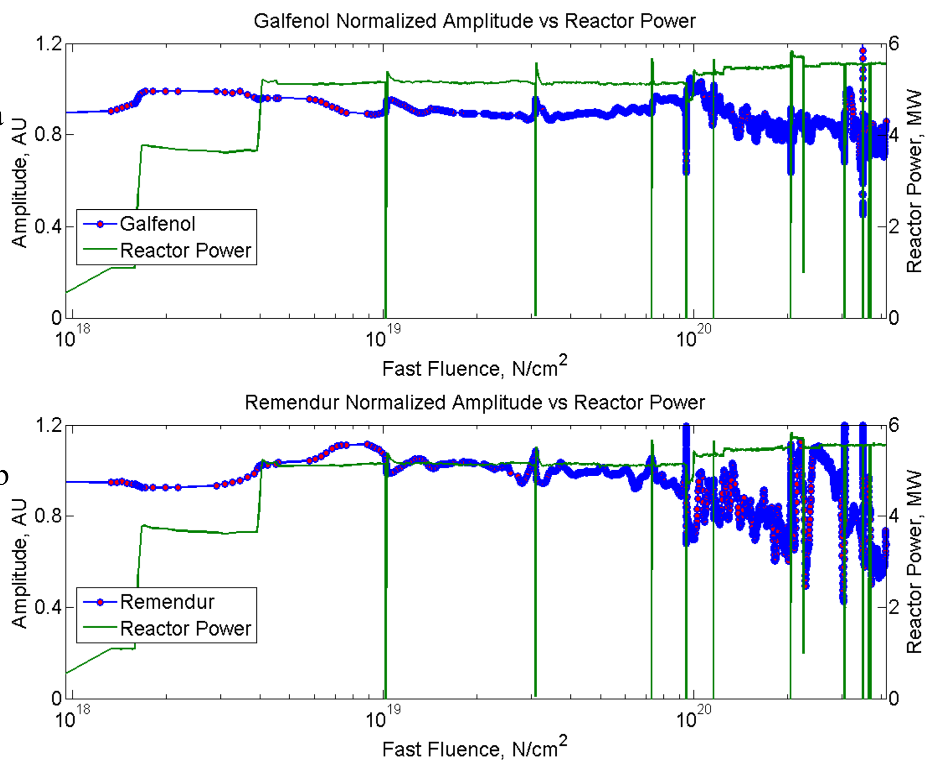

Figure 6. Magnetostrictive transducer signal amplitude as a function of accumulated fast fluence.

6 CONCLUSIONS

Results to date of this ongoing irradiation test indicate that transducers fabricated using magnetostrictive alloys (Galfenol, in particular) are very tolerant to neutron and gamma radiation effects and would be good candidates for in-core use. Piezoelectric transducer performance has been less steady, but this may be due to inconsistencies in coupling between the piezoelectric element and the waveguide (as the coupling is affected by mechanical pressure and can vary greatly during rapid temperature changes). Nonetheless, aluminum nitride has performed well and should be considered a good choice for in-core use. As the irradiation is ongoing, final conclusions cannot be made until the irradiation has finished and PIE of the test transducers and material samples is completed.

7 ACKNOWLEDGEMENT

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