

Progress Towards Developing Neutron Tolerant Magnetostrictive and Piezoelectric Transducers

Review of Progress in Quantitative Nondestructive Evaluation (QNDE)

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Progress Towards Developing Neutron Tolerant Magnetostrictive and Piezoelectric Transducers

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Abstract. Current generation light water reactors (LWRs), sodium cooled fast reactors (SFRs), small modular reactors (SMRs), and next generation nuclear plants (NGNPs) produce harsh environments in and near the reactor core that can severely tax material performance and limit component operational life. To address this issue, several Department of Energy Office of Nuclear Energy (DOE-NE) research programs are evaluating the long duration irradiation performance of fuel and structural materials used in existing and new reactors. In order to maximize the amount of information obtained from Material Testing Reactor (MTR) irradiations, DOE is also funding development of enhanced instrumentation that will be able to obtain in-situ, real-time data on key material characteristics and properties, with unprecedented accuracy and resolution. Such data are required to validate new multi-scale, multi-physics modeling tools under development as part of a science-based, engineering driven approach to reactor development. It is not feasible to obtain high resolution/microscale data with the current state of instrumentation technology. However, ultrasound-based sensors offer the ability to obtain such data if it is demonstrated that these sensors and their associated transducers are resistant to high neutron flux, high gamma radiation, and high temperature. To address this need, the Advanced Test Reactor National Scientific User Facility (ATR-NSUF) is funding an irradiation, led by PSU, at the Massachusetts Institute of Technology Research Reactor to test the survivability of ultrasound transducers. As part of this effort, PSU and collaborators have designed, fabricated, and provided piezoelectric and magnetostrictive transducers that are optimized to perform in harsh, high flux, environments. Four piezoelectric transducers were fabricated with either aluminum nitride, zinc oxide, or bismuth titanate as the active element that were coupled to either Kovar or aluminum waveguides and two magnetostrictive transducers were fabricated with Remendur or Galfenol as the active elements. Pulse-echo ultrasonic measurements of these transducers are made in-situ. This paper will present an overview of the test design including selection criteria for candidate materials and optimization of test assembly parameters, data obtained from both out-of-pile and in-pile testing at elevated temperatures, and an assessment based on initial data of the expected performance of ultrasonic devices in irradiation conditions.

BACKGROUND

Current generation light water reactors (LWRs), sodium cooled fast reactors (SFRs), small modular reactors (SMRs), and next generation nuclear plants (NGNPs) produce harsh environments in and near the reactor core that can severely tax material performance and limit component operational life. To address this issue, several

Department of Energy Office of Nuclear Energy (DOE-NE) research programs are evaluating the long duration irradiation performance of fuel and structural materials used in existing and new reactors. In order to maximize the amount of information obtained from Material Testing Reactor (MTR) irradiations, DOE is also funding development of enhanced instrumentation that will be able to obtain in-situ, real-time data on key material characteristics and properties, with unprecedented accuracy and resolution. Such data are required to validate new multi-scale, multi-physics modeling tools under development as part of a science-based, engineering driven approach to reactor development. It is not feasible to obtain high resolution/microscale data with the current state of instrumentation technology (1).

Ultrasonic measurements have a long and successful history of use for materials characterization, including detection and characterization of degradation and damage, measurement of various physical parameters used for process control, such as temperature and fluid flow rate, and in non-destructive evaluation (NDE) (2). Ultrasonic measurements have been demonstrated to successfully measure fission gas release (3), for under sodium viewing (4) for fuel porosity measurements (5,6), and for thermometry (7). However, application of ultrasonic sensors in nuclear reactors has been limited to low neutron flux environments (1). The development of ultrasonic tools to perform different in-pile measurements requires a fundamental understanding of the behavior of ultrasonic-transducer materials in these high neutron flux environments.

Ultrasound-based sensors offer the ability to obtain such data if it is demonstrated that these sensors and their associated transducers are resistant to high neutron flux, high gamma radiation, and high temperature. To address this need, the Advanced Test Reactor National Scientific User Facility (ATR-NSUF) is funding an irradiation, led by PSU, at the Massachusetts Institute of Technology Research Reactor to test the survivability of ultrasound transducers. As part of this effort, PSU and collaborators have designed, fabricated, and provided piezoelectric and magnetostrictive transducers that are optimized to perform in harsh, high flux, environments. Four piezoelectric transducers were fabricated with either aluminum nitride, zinc oxide, or bismuth titanate as the active elements where the transducers are coupled to either Kovar or aluminum waveguides. Two magnetostrictive transducers were fabricated with either Remendur or Galfenol as the active elements. Pulse-echo ultrasonic measurements of these devices are made in-situ. This paper will present an overview of the test design including selection criteria for candidate materials and optimization of test assembly parameters, data obtained from in-pile testing at elevated temperatures, and an assessment based on initial data of the expected performance of ultrasonic devices in irradiation conditions.

PIEZOELECTRIC TRANSDUCER

Much research has already been conducted regarding the performance of piezoelectric transducers. Ferroelectric materials such as PZT (8) and Barium Titanate, (9,10), have been exposed to fluences of up to 10^{18} n/cm² ($n > 0.1$ MeV). In both cases the ferroelectric materials lost half of their remnant polarization with PZT tending towards an anti-ferroelectric state. Lithium niobate was exposed up to 10^{19} n/cm² (thermal) and was measured to have a 10 fold decrease in piezoelectric sensitivity (11).

It has been noted that “hard” piezoelectric materials are the most radiation resistant (12). Hobbs (13) and Trachenko (14) have compiled considerable data on ceramics, indicating that radiation tolerance is in large part based on amorphization resistance. A down selection process (15) was applied taking into consideration phase transformations, transition temperatures, efficiency, and amorphization resistance. As such, three piezoelectric sensor materials were chosen: Aluminum Nitride (AlN), Zinc Oxide (ZnO), and Bismuth Titanate (BiT). AlN and ZnO were chosen because of their high transition temperature, see Table 1, and the Wurtzite crystal structure. Further, AlN was successfully irradiated by Parks and Tittmann at the Pennsylvania State University’s Breazeale Reactor up to 10^{18} n/cm² ($n > 1$ MeV) without loss in transduction efficiency (16). Although BiT has a lower amorphization resistance, it has higher efficiency, see Table 1.

TABLE 1. Candidate piezoelectric materials.

Piezoelectric Sensors			
Material	Transition Temperature, °C	Transition Type	d_{33} , (m/V)* 10^{-12}
AlN (Aluminum Nitride)	2200x	Melt	0.3
Bi ₄ Ti ₃ O ₁₂ (Bismuth Titanate)	680	Curie Temperature	18
ZnO (Zinc Oxide)	>1500	Melt	10

In order to access the performance of the sensors during irradiation, a test capsule was designed to enable pulse-echo measurements. The design came from Parks and Tittmann's work with high temperature transducers (16,17). Briefly, the sensor (AlN, BiT, or ZnO) was coupled to a waveguide (Kovar or Aluminum 6061) using high purity gold or aluminum foil. A carbon-carbon backing was used to dampen the sensor vibrations, while a nickel plunger was used to increase pressure between the waveguide and the sensor material as well as to connect to the lead electrode. A stainless 304 casing was used as ground while a stainless 204 cap screwed into the casing was used to exert pressure on the nickel plunger increasing the pressure between the sensor and the waveguide. A high temperature nickel-iron-cobalt spring was used to help maintain coupling pressure as temperature varied. See Figure 1 for pictures during various stages of the assembly.



FIGURE 1. (a) The AlN transducer bonded to the Kovar waveguide. (b) The assembled sensor and capsule. The lead electrode is contacting the nickel plunger through direct contact and covered with an alumina sleeve. The alumina sleeve is fixed in place using Sauereisen. (c) A strain relief was design to ad support to the cable connections. (d) The full assembly laid out. (e) The assembled sensor.

MAGNETOSTRICTIVE TRANSDUCER

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 relatively high Curie temperature and magnetostriction. Although Remendur is no longer commercially available, several identical alloys are available under different names. INL also has a supply of Remendur which is being used for this test. Galfenol is a relatively new alloy of iron and gallium. Galfenol is a member of the “giant” magnetostrictive alloys and has a very large saturation magnetostriction. It also has an appropriately high Curie temperature, see Table 2. Neither constituent element reacts strongly with neutron radiation.

TABLE 2. Candidate magnetostrictive materials.

Magnetostrictive Transducers		
Material	Composition	Key Properties
Remendur	49%Fe-49%Co-2%V	10-100 ppm strain max, magnetostriction, Curie temperature 950 °C
Galfenol	Fe-14Ga-Nbc	100-400 ppm strain max. magnetostriction, Curie temperature 700 °C

The magnetostrictive transducer design was selected based on research by Daw (18). The magnetostrictive transducers consist of a small driving/sensing coil made of silver palladium by wrapping it around an alumina bobbin. The coil is coated in a standoff insulation and heat treated. The coil is then coated in an alumina cement and heat treated until cured. The lead electrodes are then laser welded to the ends of the coil. The magnetostrictive element is inserted into the alumina bobbin. The assembly is then placed in a pre-fabricated stainless steel 304 casing. See Figure 2 for images at different steps in the fabrication process.

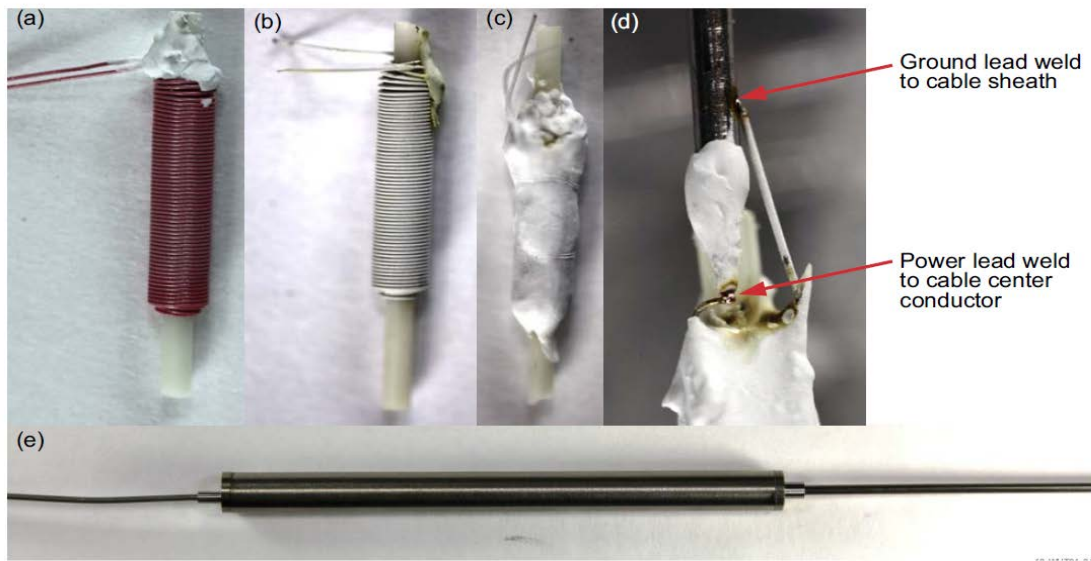


FIGURE 2. Silver-palladium is wrapped around an alumina bobbin (b) the wire is coated in a standoff insulation and heat treated, (c) coated with alumina cement, and heat treated a second time, (d) laser welded coax cable, and (e) placed into a pre-fabricated housing.

TEST CAPSULE

In this experiment, two magnetostrictive transducers, each consisting of either a Galfenol or Remendur active element, four piezoelectric transducers, each consisting of either AlN, ZnO, or BiT as sensor materials were constructed. Along with these transducers were: two K-Type thermocouples, a self-powered neutron detector, a self-powered gamma detector, melt wires, and spare sensor material wafers to ride along with the experiment. The Massachusetts Institute of Technology Reactor is characterized by the following features:

- Total flux = $1.89\text{E}+14$ n/cm²/s
- Thermal flux (<0.4 eV) = $2.12\text{E}+13$ n/cm²/s
- Epi-thermal flux (0.4 eV - 0.1 MeV) = $8.03\text{E}+13$ n/cm²/s
- Fast flux 1 (>0.1 MeV) = $8.78\text{E}+13$ n/cm²/s
- Fast flux 2 (>1.0 MeV) = $4.05\text{E}+13$ n/cm²/s
- Gamma dose rate: 1×10^9 r/hr
- Temperature: 350–500°C

The primary means for setting the experiment's operating temperature is to balance the gamma and neutron heating of the components with heat rejection to the reactor primary water (50°C). The in-core facility used for this irradiation, the In-Core Sample Assembly (ICSA), consists of a titanium tube in contact with the reactor primary water on its outside surface. The internal space (about 5cm in diameter) is filled with an inert gas that is injected at a set mass flow rate at the bottom of the core and removed at the top. Figure 3 (A) shows the test capsule and sensor layout. Figure 3 (B) shows the thermal profile of the sensors during irradiation.

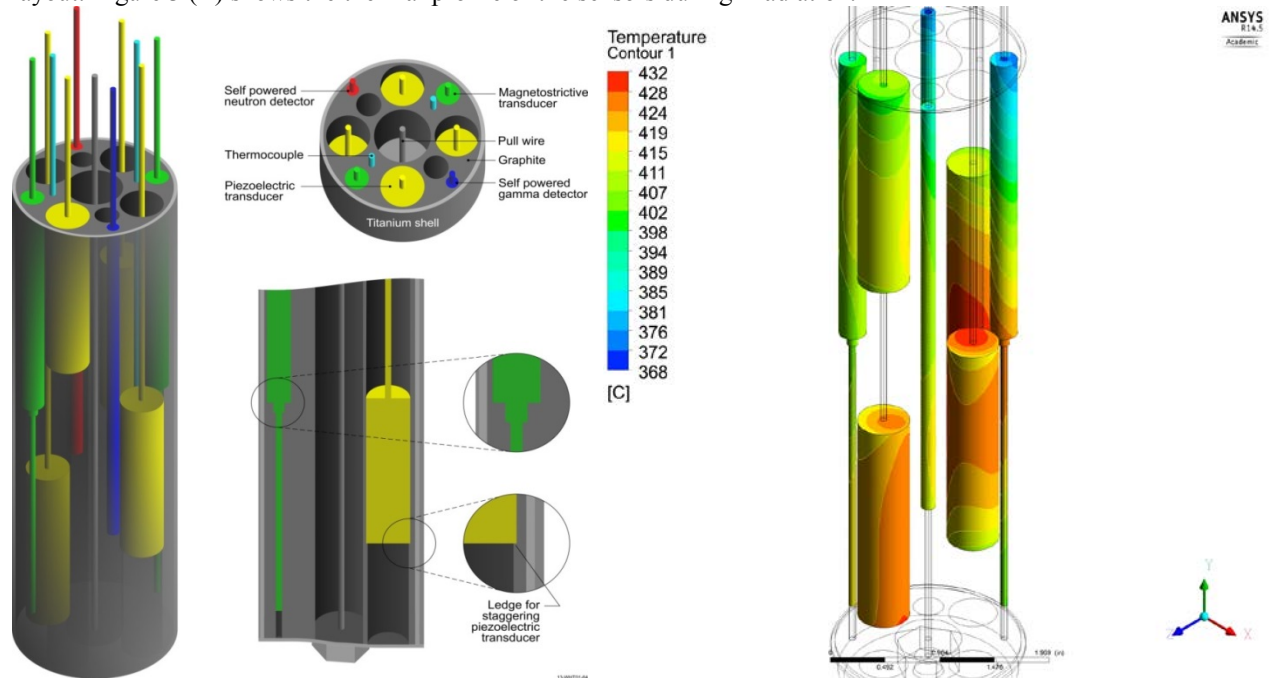


FIGURE 3. (A) Test capsule and sensor layout. (B) Thermal profile of sensors during irradiation.

RESULTS

The experiment is still on going and here we present results from the first power cycle (180–980 hours) when the reactor was operating at 5 MW. The reactor power was incremented to 5 MW over a period of one week to allow the sensors to slowly increase in temperature, see Figure 4. The zinc oxide transducer had an electrical malfunction during the insertion process. One of the AlN transducers developed an electrical short when the temperature reached 400 °C.

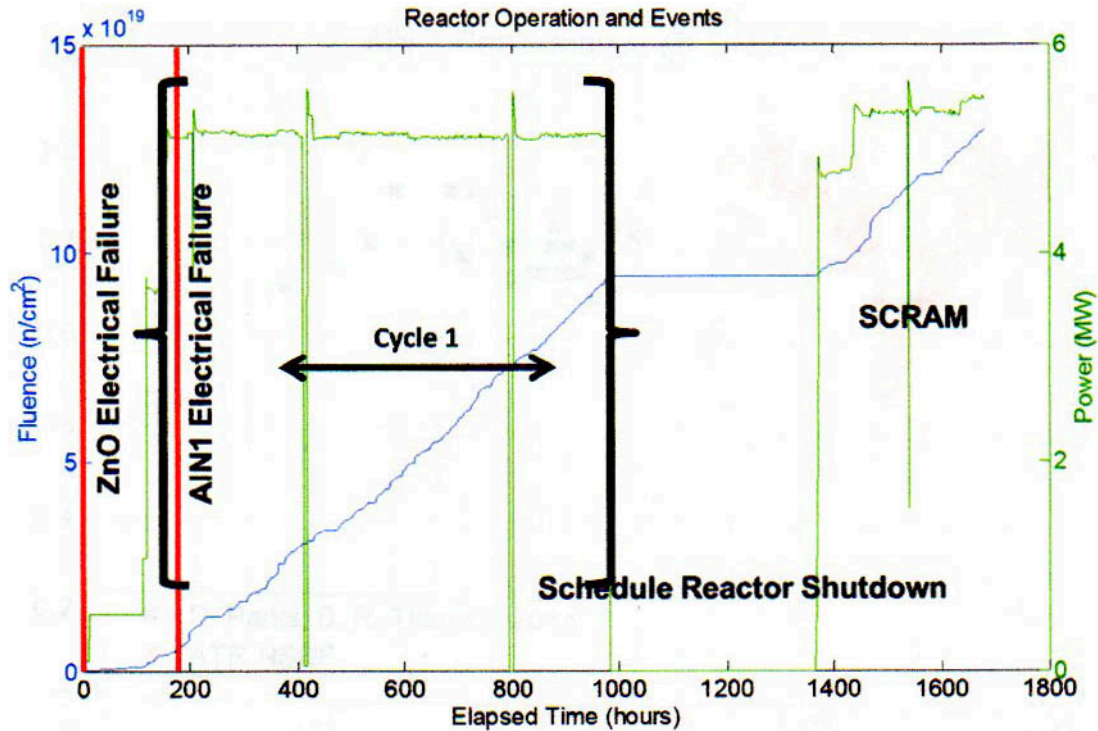


FIGURE 4. The blue curve indicates the integrated neutron flux (fluence) for the experiment for neutrons with energy greater than 1 MeV. The green curve represents the reactor power.

During the first power cycle, A-scans were periodically collected from the remaining AlN and BiT transducers as well as from the two magnetostrictive transducers. The pulse echo-amplitude was determined from windowing the first returned echo and measuring the amplitude of the fundamental frequency component on an FFT. The results of this analysis are shown in Figures 5–7. Figure 5 shows the pulse-echo amplitude measured from the surviving aluminum nitride sensor. The plot was normalized to the amplitude pre-irradiation. The pulse-echo amplitude does vary by $\pm 20\%$ during irradiation. Figure 6 shows the pulse-echo amplitude measured from the BiT sensor. During the first cycle the pulse-echo amplitude decreased by approximately 65%. Figure 7 shows the pulse-echo amplitude of the magnetostrictive transducers. These plots were also normalized to the pre-irradiated pulse-echo amplitude. The transient behavior in these sensors seems to be less pronounced as the pulse-echo amplitude only varies by about $\pm 10\%$.

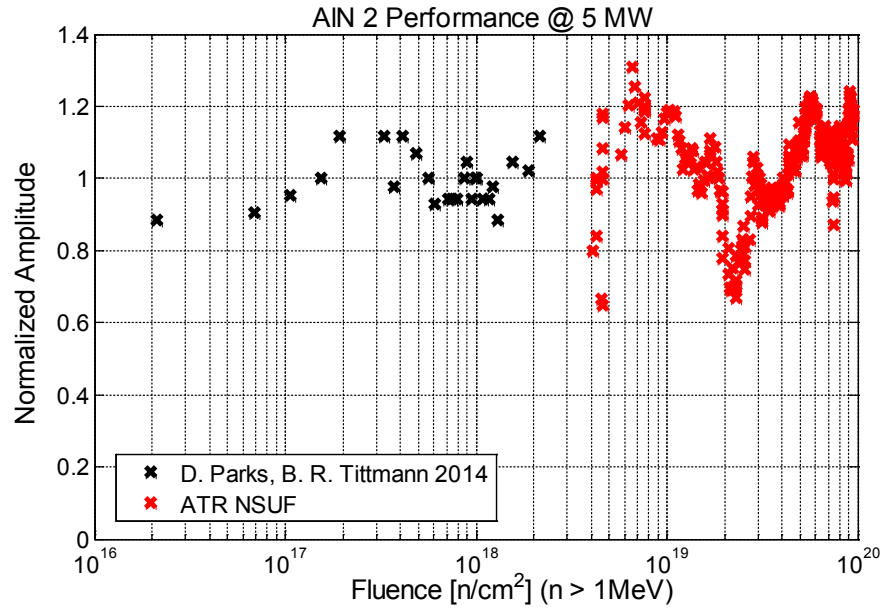


FIGURE 5. Plot of measured pulse-echo amplitude measured by the remaining AIN transducer. The amplitude was normalized to the first measurement made at 0 fluence. The data is plotted only for the first power cycle and compared to the data collected by Parks and Tittmann (15,16).

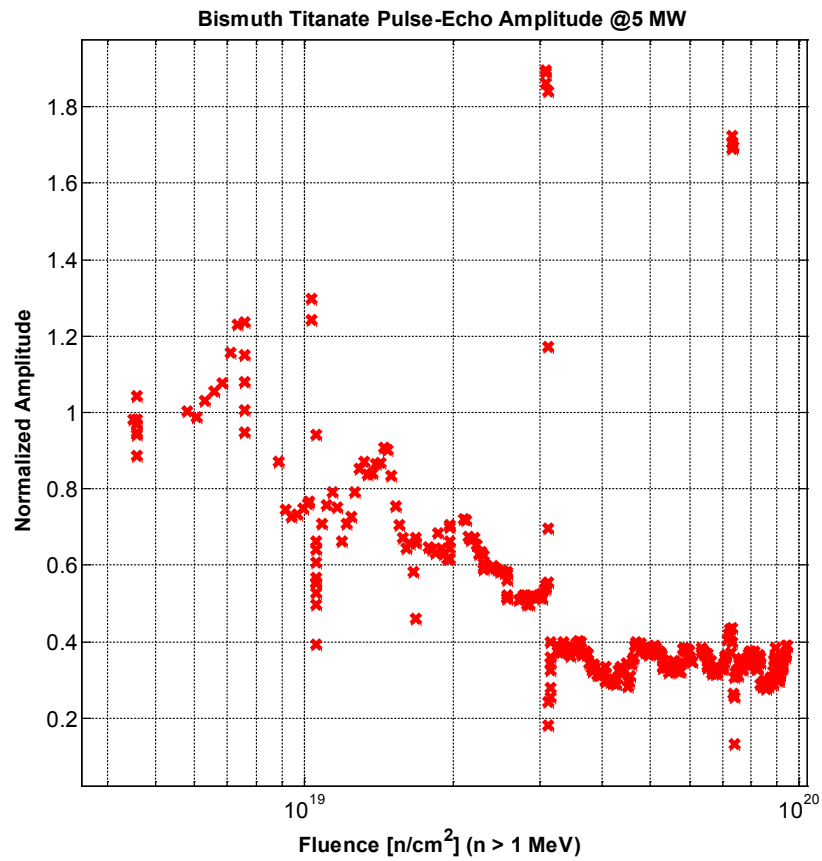


FIGURE 6. Pulse-echo amplitude measured for the Bismuth Titanate transducer during the first power cycle.

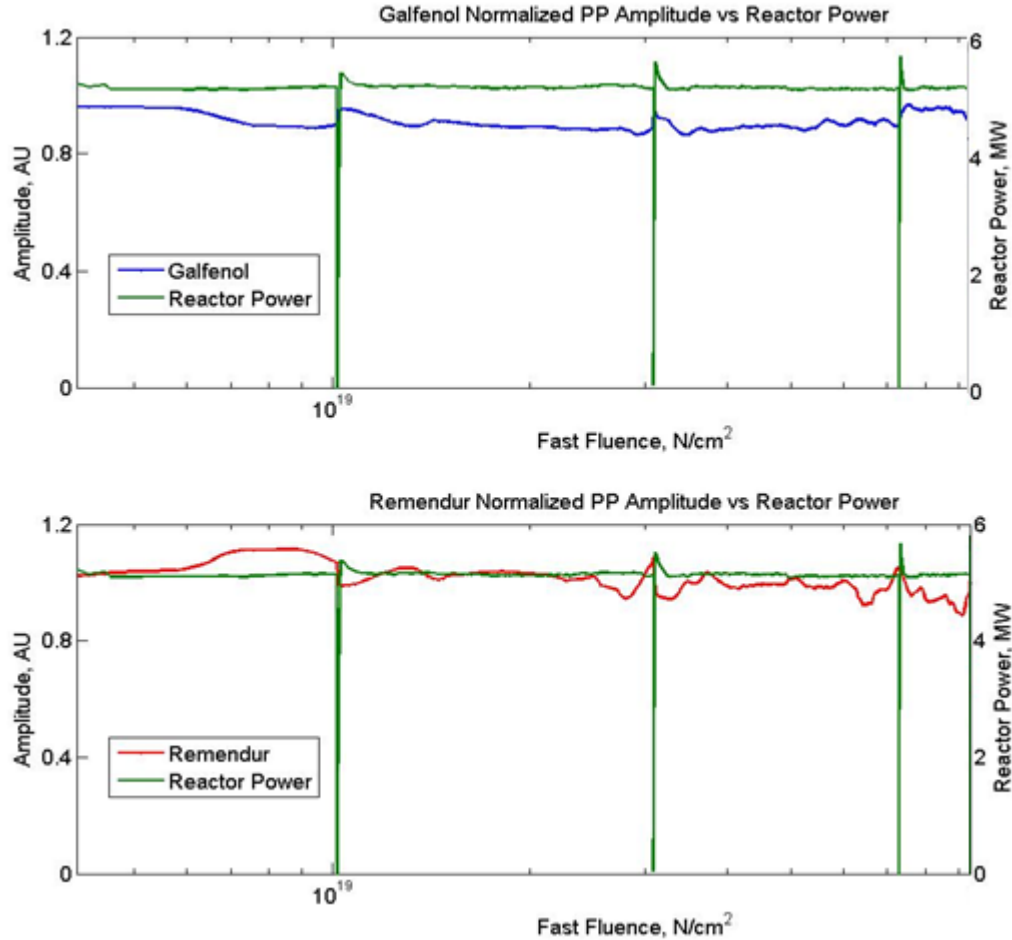


FIGURE 7. Pulse-echo amplitude as measured by the Galfenol and Remendur magnetostrictive transducers.

CONCLUSIONS

For practical use in harsh radiation environments the selection criteria for piezoelectric materials for NDE and material characterization were summarized. Using these criteria piezoelectric Aluminum Nitride was shown to be a viable candidate. The results of tests on an Aluminum Nitride, Bismuth Titanate, Remendur, and Galfenol based transducers operating in a nuclear reactor during a window of forty days at a fast neutron flux of $4.05\text{E}+13\text{ n/cm}^2$ and a gamma dose rate of $1 \times 10^9\text{ r/hr}$ were presented. In all cases clear A-Scan measurements were made at the end of the power cycle. Remendur, Galfenol, and AlN seemed to maintain the initial transduction efficiency. The Aluminum nitride sensors pulse-echo amplitude varied by $\pm 20\%$ while the Remendur and Galfenol varied by $\pm 10\%$. The BiT pulse-echo amplitude had decreased by 65% of its initial value at the end of the first power cycle. The results show promise for utilizing both piezoelectric and magnetostrictive transducers in high neutron flux environments. The results offer potential for improving reactor safety and furthering the understanding of radiation effects on materials by enabling structural health monitoring and NDE in spite of the high levels of radiation and high temperatures known to destroy typical commercial ultrasonic transducers.

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