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Testing Piezoelectric Sensors in a Nuclear Reactor Environment

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Abstract. Several Department of Energy Office of Nuclear Energy (DOE-NE) programs, such as the Fuel Cycle Research and Development (FCRD), Advanced Reactor Concepts (ARC), Light Water Reactor Sustainability, and Next Generation Nuclear Power Plants (NGNP), are investigating new fuels, materials, and inspection paradigms for advanced and existing reactors. A key objective of such programs is to understand the performance of these fuels and materials during irradiation. In DOE-NE's FCRD program, ultrasonic based technology was identified as a key approach that should be pursued to obtain the high-fidelity, high-accuracy data required to characterize the behavior and performance of new candidate fuels and structural materials during irradiation testing. The radiation, high temperatures, and pressure can limit the available tools and characterization methods. In this work piezoelectric transducers capable of making these measurements are developed. Specifically, three piezoelectric sensors (Bismuth Titanate, Aluminum Nitride, and Zinc Oxide) are tested in the Massachusetts Institute of Technology Research reactor to a fast neutron fluence of 8.65×10^{20} n/cm². It is demonstrated that Bismuth Titanate is capable of transduction up to 5×10^{20} n/cm², Zinc Oxide is capable of transduction up to at least 6.27×10^{20} n/cm², and Aluminum Nitride is capable of transduction up to at least 8.65×10^{20} n/cm².

INTRODUCTION

Motivation

Several DOE-NE programs, such as the Fuel Cycle Research and Development (FCRD) [1], Advanced Reactor Concepts (ARC) [2], Light Water Reactor Sustainability (LWRS) [3], and Next Generation Nuclear Power Plants (NGNP) programs [4], [5], are investigating new fuels and materials for advanced and existing reactors. A key objective of such programs is to understand the performance of these fuels and materials during irradiation. The Nuclear Energy Enabling Technology (NEET) Advanced Sensors and Instrumentation (ASI) in-pile instrumentation development activities are focused upon addressing cross-cutting needs for DOE-NE irradiation testing by providing higher fidelity, real-time data, with increased accuracy and resolution from smaller, compact sensors that are less intrusive [6]. The NEET ASI has established the Ultrasonic Transducer Irradiation and Signal Processing Enhancements project because it will enable in-core use of ultrasound-based sensors that are uniquely capable of meeting DOE program enhanced instrumentation needs.

In-situ instrumentation is desired to provide real-time data on fuel performance phenomena. Without wire-less transmission capabilities, drop-in or static capsule experiments only allow data to be obtained at the endpoint of an experiment. Although material property measurements may be made on samples at the end of the test, such measurements are subject to error because of handling and because measurements are not made under prototypic pressures, fluxes, and/or temperatures. Sensors included in such tests can provide only insights about the integral neutron fluence or peak temperatures. In-situ instrumentation in irradiation tests can provide data showing the evolution of particular phenomena over time. However, in-pile instrumentation has proven difficult to develop as piezoelectric materials along with transducers degrade when subjected to high temperatures and radiation.

Many piezoelectric materials have been tested [7, 8, 9, 10]: in PZT, anti-ferroelectric properties develop [8]; in Barium Titanate, a phase transition occurs [7], and in Lithium-Niobate, the process of metamictization destroys the asymmetry necessary for piezoelectric transduction [11] all of which fail under neutron fluences on the order of 10^{18} - 10^{19} n/cm². Although some of the high temperature single crystals such as quartz or gallium phosphate looked like promising materials, amorphization of the bulk material is problematic. For instance, quartz has been shown to degrade at fluences of approximately 9×10^{19} n/cm² due to amorphization of domains, as measured by x-ray spectroscopy [12], [13].

Not all materials are doomed when it comes to piezoelectric transduction in nuclear reactors. Thin film AlN has been shown to be unaffected by gamma irradiation up to 18.7 MGy [14] and temperatures up to 1000° C [15], [16]. Moreover, this material has been explicitly cited in numerous independent studies as a highly radiation tolerant [17]. Tests of bulk single crystal AlN in a TRIGA nuclear reactor core showed this material to be completely unaffected by a fast and thermal neutron fluence of 1.85×10^{18} n/cm² and 5.8×10^{18} n/cm² respectively and a gamma dose of 26.8 MGy [18].

That being said, there has been an insufficient number of studies on the effects of neutron irradiation on piezoelectric materials. A review paper on the radiation endurance of piezoelectric transducers by Sinclair and Chertov only lists six piezoelectric materials, few of which have been exposed to a high enough neutron fluence to replicate commercial power water reactor environments. The purpose of this study is to collect more data on the radiation tolerance of piezoelectric materials.

In this study, three piezoelectric materials are instrumented and placed in the Massachusetts Institute of Technology's (MIT) Nuclear Reactor. The materials chosen were Aluminum Nitride, Zinc Oxide and Bismuth Titanate. Due to the limited nature of the TRIGA reactor testing by Parks and Tittmann, AlN was chosen for this to determine its transduction capabilities at higher fluences.

Zinc oxide has one of the strongest piezoelectric electromechanical coupling factors (k_{33}) of the tetrahedrally bonded semiconductors, such as GaN and AlN, mostly because the clamped-ion and internal strain contributions do not cancel each other out as substantially [19]. It has a coupling coefficient of approximately 0.48 while aluminum nitride has a coupling coefficient of approximately 0.3. This is in-part due to the higher piezoelectric coupling coefficient (d_{33}). Zinc Oxide's d_{33} is approximately 12.4 pC/N, while aluminum nitride's is approximately 5 pC/N [20].

There is little literature on zinc oxide's transduction capabilities in a radiation environment. However, like AlN, the strong ionic bonding of ZnO is thought to provide a high resistance to amorphization [17]. A fast neutron (1 MeV) is capable of delivering 59 keV to a zinc primary knock-on-atom or 221 keV to an oxygen primary knock-on-atom. This is sufficient energy to displace either atom from their lattice position with similar energies to keV ion implantations. However, during an irradiation of approximately 1.0×10^{19} n/cm² only a slight change in lattice parameters was observed ($a=0.03\%$ decrease and $c=0.04\%$ increase) as well as blistering to the specimens surface [21].

Of the pervoskyte type ceramics bismuth titanate is thought of as the likeliest candidate to perform well in a high neutron flux environment. Kazys has exposed Bismuth Titanate to 22.7 MGy dose from a ⁶⁰Co source with only a moderate decrease in pulse-echo amplitude (4%) [14]. Although the phase transition at 650 °C is unfavorable for thermal spike damage, bismuth Titanate has a higher d_{33} than both AlN and ZnO. Although it may not survive as long as single crystal ceramics, it could be utilized for environments with a lower neutron flux, while providing increased sensitivity.

METHOD

The irradiation of the chosen sensor materials was conducted at the MIT nuclear reactor, being inserted in-core. The MIT reactor is a light-water cooled and moderated, heavy-water reflected, nuclear reactor. The reactor uses UAl_x cermet clad with Al 6061 designed in a rhombohedral shape, producing neutron flux rates for the sensors position given in Table 1 [22].

TABLE 1. Overview of neutron flux energy spectrum.

Unit: n/cm²/s	Flux @ 5.8 MW	Flux/MW
Thermal Flux (<0.4 eV)	2.12E+13 (11.2%)	3.66E+12
Epi-thermal flux (0.4 eV, 0.1 MeV)	8.03E+13 (42.4%)	1.38E+13
Fast flux 1 (>0.1 MeV)	8.78E+13 (46.4%)	1.51E+13
Fast flux 2 (>1.0 MeV)	4.05E+13 (21.4%)	6.98E+12
Total (Full Range)	1.89E+14 (100%)	3.26E+13

The in-core sample assembly (ICSA) consisted of a titanium tube in contact with the primary water on its outside surface, originating at the reactor head and extending into the reactor core. The internal space (about 5cm in diameter) was filled with inert gasses, helium and neon, that were injected at variable mass flow rates at the bottom of the core and removed at the top. Flow rates were adjusted to control the temperature of the sensor capsules.

A specimen fixture was machined from graphite and clad with titanium. In it, were four piezoelectric sensors, two magnetostrictive sensors, two K-type thermocouples, a neutron flux detector, and a gamma flux detector. Each of these devices was fitted with a 1mm outer diameter, mineral insulated coaxial cable (Thermocoax Inc., Suresnes, France). The cable was constructed with a stainless steel 304L outer sheath, a copper inner sheath, silicon dioxide insulation, and a copper inner lead wire. There were a total of ten cables each approximately 18 feet long. They were connected to a 50Ω impedance junction box at the top of the reactor and to each of the respective sensors through a strain relief at the top of the capsule.

In addition to the active and passive sensors were five melt wires encapsulated in quartz, with melt temperatures ranging from 327 to 514 °C, Fe-Ni-Cr flux wires in various positions in the capsule, and spare active elements Galfenol, Remendur, AlN, and BiT. The capsule and each of the respective components is shown in figure 2. Although apart of the experimental setup, the magnetostrictive sensors will not be discussed further.

The piezoelectric sensors were constructed from a stainless steel 218 housing, stainless steel 304 cap, alumina insulation, nickel lead electrode, carbon-carbon backing, kovar or aluminum 6061 waveguide, and a high temperature spring. The active element was bonded to the kovar waveguide, in the case of the aluminum nitride sensors, using a high temperature ceramic (Aremco Products, Inc.), and, in the case of the zinc oxide and bismuth titanate sensors, using 99.99% pure aluminum foil, Figure 2.

A 15.24 meter RG 58 low loss coaxial cable connected to the 50Ω junction box and an NI-PXI 1042 chassis through PXI-2593 switch cards. The switch card was connected to the NDT-5800 PR pulser/receiver and a SMX-2064 Digital Multimeter. This enabled either DC impedance measurements or pulser/receiver measurements. The settings used for the pulser/receiver are saved alongside each of the waveforms in an associated file. The pulser/receiver was connected to a ZTEC ZT4211 digital oscilloscope.

A LabVIEW program was designed so that periodic measurements could be made on each of the transducers connected to the switch board. The data output was organized by transducer and tagged with the acquisition date and time so that acoustical data could be correlated with external sensor measurements such as neutron power or temperature. The sample rate was initially one measurement every thirty minutes, but was increased to one measurement every two hours after the reactor reached its first power cycle.



FIGURE 1. 3D Renderings of the ULTRA-capsule as constructed for the irradiation. The piezoelectric sensors are shown in yellow, the magnetostriuctive sensors are shown in green, the piezoelectric drop in specimens are shown as gray disks, while the magnetostriuctive drop-in samples are shown in a peach cylinder.

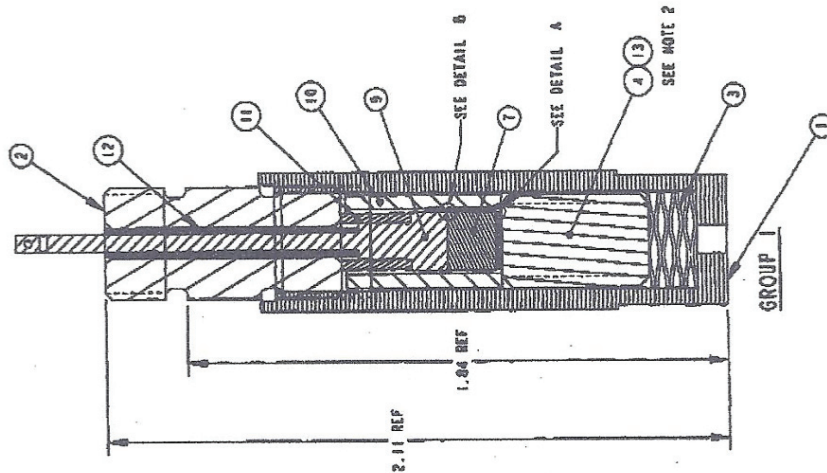


FIGURE 2. Design of piezoelectric capsule. The components are: (1) outer Casing, (2) cap (3) high temperature spring, (4) waveguide, (7) backing, (9) plunger, (10) insulation, (11) Insulation, and (12) insulation. The piezoelectric element was placed between the backing (7) and the waveguide (4) using either foil or a bonding agent.

Overview of Irradiation

The ULTRA capsule was inserted into the reactor on February 18th 2014 and removed on May 12th 2015. The samples were in the reactor for a total of 449 days before they were moved to the hot cell for post irradiation analysis. The capsule rode along the reactors normal operation and as such was subjected to periodic shutdowns. The neutron fluence reached 1.877×10^{21} n/cm², for neutrons with energy greater than 0.1 MeV, and 8.68×10^{20} n/cm², for neutrons with energy greater than 1 MeV. The max gamma exposure was approximately 7.23×10^{21} gamma/cm².

RESULTS

Aluminum Nitride

One of the aluminum nitride sensors survived until the very end of the experiment, while the other failed due to an electrical short. The A-scan from the first day following insertion and the last measurement made during the irradiation is plotted in Figure 3(a). The last measurement clearly shows a longer ring down than the first measurement as well as generally lower signal to noise ratio. Although the first and second echoes of the last A-scan are not resolvable, the third and fourth echoes are distinct.

A comparison of the third echoes is shown in Figure 3(b). There is a reduction in the pulse-echo amplitude, coupled with a 6.7% reduction in the center frequency, and 14% increase in bandwidth. Comparing the fourth echoes before and after the experiment, there was a 57% reduction in the peak amplitude.

After the irradiation the capsule was disassembled and optical inspections revealed that the backing material had delaminated from the surviving AlN sensor, which might account for the change in quality factor. Also, the sensor changed from a milky white before the irradiated to black after the irradiation. Further, d_{33} measurements made on the AlN drop in sample were in approximately 2.9 ± 2 pC/N.

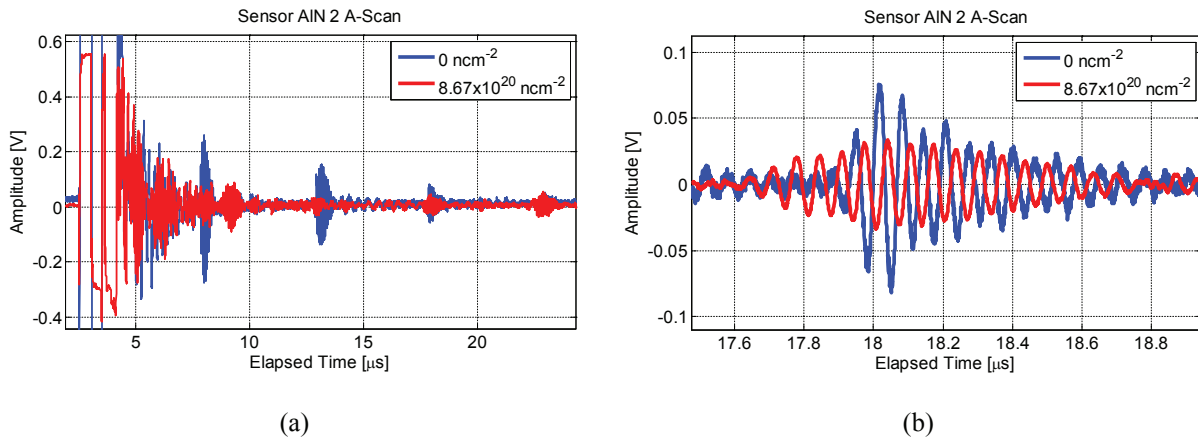


FIGURE 3. (a) Comparison of a-scans before and after irradiation. (b) Comparison of third echo in a-scans before and after irradiation.

Zinc Oxide

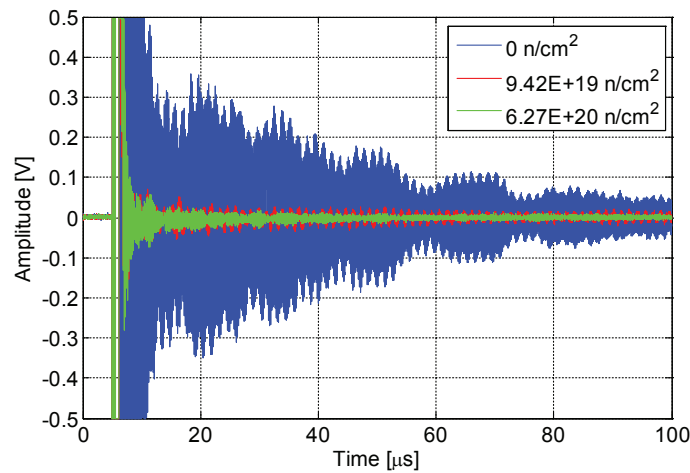


FIGURE 4. A-scans from the ZnO sensors for selected fluence levels during the irradiation. In all cases the reactor was off.

Due to changes in coupling of the active element and the waveguide as a function of reactor power, measurements could only be made while the reactor was off. The change in coupling occurred early in the irradiation, which optical inspection after the test indicates was a delamination of the active element from the waveguide.

Figure 4 shows selected A-scans during the irradiation. It is assumed that the initial decrease in pulse-echo amplitude is a result of the delamination. An echo from the back wall of the waveguide was identified and measured to determine the performance of the ZnO sensor during the irradiation. At the end of the experiment no pulse-echo amplitude was measured. Because the pulse-echo amplitude could only be measured when the reactor was off, the sensor is considered to have failed at $6.27\text{E}+20 \text{ n/cm}^2$ just before the last power cycle. It is unclear what caused the failure, however, the DC resistance was 120Ω , when it should have been several $\text{M}\Omega$.

After taking the sensor capsule apart it was clear that the active element had delaminated from the waveguide and that it had changed from a milky clear color to an orange color. Unfortunately the sensor was too radioactive to take out of the hot box to measure its material properties.

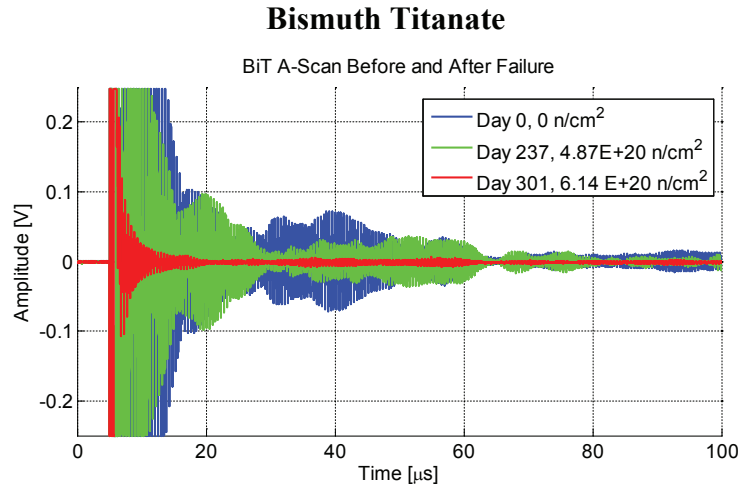


FIGURE 5. Comparison of A-scans from day 1, 237, and 301 from BiT sensor. These represent A-scans from before and after failure of the sensor.

In these experiments, Bismuth Titanate has been demonstrated to operate up to a fast neutron fluence of $5\text{E}+20 \text{ n/cm}^2$, Figure 5. There was an initial decline in the amplitude of the fundamental harmonic starting on day 10 or at $1.28\text{E}+19 \text{ n/cm}^2$, reaching a minimum relative amplitude of 60% at $5.29\text{E}+19 \text{ n/cm}^2$ on day 25. The amplitude gradually recovered to a relative amplitude of 100% at $1.1\text{E}+20 \text{ n/cm}^2$. The sensor failure is noted by a significant decrease in the fundamental amplitude after $5\text{E}+20 \text{ n/cm}^2$. This decline occurred over a period of 17 days, dropping in amplitude by 80%. There was no notable recovery during this time, however, an echo from the back wall of the waveguide was still measureable. At the end of the irradiation the DC impedance was measured to be $11 \text{ k}\Omega$. Further, the d_{33} was $11.5 \pm 1.5 \text{ pC/N}$, measured from the drop in sample after the irradiation. The d_{33} as reported by the material supplier, TRS Technologies, is 20 pC/N .

CONCLUSIONS

A summary of the results are shown in Table 2. Acoustic measurements indicated that all of these sensors were able to transduce during the irradiation. The bismuth titanate sensor began to degrade in performance and failed at approximately $5 \times 10^{20} \text{ n/cm}^2$. This failure was gradual, occurring over a period of several weeks. This failure is suspected to be a result of the irradiation and is consistent with a reduction in remnant polarization. Further, measurements on a bismuth titanate sample, riding along with the experiment, showed a 50% decrease in d_{33} . This supports the conclusion that the remnant polarization had decreased.

The zinc oxide sensor was finicky throughout the entire experiment. The sensor was only capable of transduction during the reactor off cycles making characterization of its pulse-echo amplitude difficult. However, it is clear that the sensor was capable of transduction up to approximately $6.27 \times 10^{20} \text{ n/cm}^2$. At this point, the reactor turned on and

measurements were only obtainable at the end of the experiment. At the end of the irradiation the sensor was pulsed and no acoustic data was measured. It is not known if the failure was due to material property changes in the ZnO or if there was a failure in the construction of the sensor. It should be noted that the ZnO had changed in color.

Lastly, the aluminum nitride sensor, by far, performed the best. The sensor was capable of transduction throughout the entire irradiation. Although the report pulse-echo amplitude is reported to decrease by 57%, this measure is not consistent when measuring each of the echoes. When considering the fourth echo, the drop in pulse-echo amplitude is only 12%. This leaves lots of future work in improving the signal processing algorithms used to analyze the acoustic signals.

Although a huge success, this experiment still requires significant work. Post irradiation examination has only just begun with optical and acoustical inspections being completed. However, numerous impedance and microstructural measurements can be made at facilities such as Idaho National Labs. These measurements help develop constitutive models to describe the evolution of mechanical and electrical properties of the active element while subject to similar neutron fluxes.

TABLE 2. Summary of Irradiation Effects on Ultrasonic Sensors.

Sensor	Fluence (n/cm ²)	Gamma (Gy/cm ²)	%Change PE Amp.	Color
AlN 1	3.88E+18	3.23E+19	-14	Milky Clear/NA
AlN 2	8.68E+20	7.23 E+21	-57	Milky Clear /Black
ZnO	6.43E+20	5.36E+21	-45.7	Milky Clear/ Orange
BiT	4.95E+20	4.12E+21	-93	White/NA

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