Analysis of high temperature effects on piezoelectric based ultrasonic transducers

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Analysis of high temperature effects on piezoelectric based ultrasonic transducers
by
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A dissertation submitted to the graduate faculty
in partial fulfillment of the requirements for the degree of
DOCTOR OF PHILOSOPHY

Major: Engineering Mechanics

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The student author, whose presentation of the scholarship herein was approved by the program of study committee, is solely responsible for the content of this dissertation. The Graduate College will ensure this dissertation is globally accessible and will not permit alterations after a degree is conferred.

Iowa State University

Ames, Iowa

2018

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DEDICATION

To my grandparents and parents.
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<td>BHABHA ATOMIC RESEARCH CENTER, INDIA</td>
</tr>
<tr>
<td>FEA</td>
<td>FINITE ELEMENT ANALYSIS</td>
</tr>
<tr>
<td>HEDL</td>
<td>HANFORD ENGINEERING DEVELOPMENT LABORATORY</td>
</tr>
<tr>
<td>HT</td>
<td>HIGH TEMPERATURE</td>
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<td>IGCAR</td>
<td>INDIRA GANDHI CENTER FOR ATOMIC RESEARCH, INDIA</td>
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<td>IN-SITU INSPECTION AND REPAIR</td>
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ACKNOWLEDGMENTS

I would like to thank my committee chair, Prof. Leonard J. Bond, and my committee members, Prof. Wei Hong, Prof. Ronald Roberts, Prof. Timothy Bigelow, and Dr. Paul Schafbuch, for their guidance and support throughout the course of this research. This work was supported by U.S. Department of Energy’s office of Nuclear Energy under Nuclear Energy University Program (NEUP).

I am grateful to Mr. Daniel J. Barnard (Associate Scientist, CNDE) for helping with the development of the transducer and for the insightful discussions after the measurements. I would also like to take this opportunity to thank Dr. Sunil Chakrapani (Michigan State University) and Dr. Vamshi Krishna Chillara (Las Alamos National Laboratory) for their mentorship during some of the challenging periods in my doctoral research.

Lastly, I would like to thank my friends specifically Tejaswi, and Ketaki, my colleagues, the department faculty and staff for making my time at Iowa State University a wonderful experience.
ABSTRACT

Generation IV fast nuclear reactor designs are being evaluated to support sustainable development, economic competitiveness, and improved safety of the reactors. Under sodium viewing (USV) using pulse-echo techniques offers the potential for the inspection of liquid sodium-cooled fast reactors (SFRs) at the hot stand-by temperature (260°C). However, the harsh environment effects on the transducer reduce the ultrasonic signal strength which also limits the probability of detection (POD) of the defects.

Current work presents a unified modeling and measurement-based methodology to analyze the high temperature effects on the transducer components. Resonance analysis-based characterization has shown that the conventional $d_{33}$ piezoelectric parameter is not a sufficient condition to estimate resonance characteristics of high temperature ultrasonic transducers. A full piezoelectric material matrix needs to be utilized in order to estimate high temperature performance. In seeking materials, BiScO$_3$-PbTiO$_3$ (Bismuth Scandium oxide-lead titanate) is demonstrated to be the piezoelectric material that could potentially be used for hot stand-by mode (260°C) inspection of SFRs.

However, a high temperature ultrasonic transducer is also a multi-layer system where the interaction of different acoustic layers is equally important as the temperature dependence of a single piezoelectric layer. A 3-layer problem was studied which demonstrated the thermal cycling effect on the interfaces, evident from the echo amplitudes and bandwidth of the frequency response. A unique bimodal resonance phenomenon was also found in the transducer due to interaction of the multiple acoustic layers.

Utilizing these insights, design, development and high temperature evaluation of several prototypes was performed in surrogate fluids. This resulted in an air-backed
transducer using BS-PT piezoelectric material, nickel faceplate, and liquid acoustic coupling with silicone oil which demonstrated the ability to image regions of different thickness within the specimen, critical for USV capabilities in SFRs.

At high temperatures, the prototype transducers were seen to have a reduction in signal strength which can lead to a reduction in POD. It was recognized that in assessing performance of an NDT inspection, these POD experiments are, in general, time consuming and expensive, due to the cost of fabricating appropriate sample sets which include representative populations of defects. A more cost-effective approach, which can also be used to supplement a more limited experimental program, and to increase confidence is using physics-based modeling to predict POD.

Current work developed a novel temperature compensated transfer function approach to predict POD at high temperature using room temperature experimental data. Such a model-assisted POD approach for the temperature dependence of PZT-5A material, was demonstrated near 200°C. For a known temperature dependent degradation mechanism, this approach could be extended for other high temperature transducer materials using finite element model and room temperature experimental data to estimate high temperature POD.
CHAPTER 1. INTRODUCTION

Generation IV nuclear power plant (NPP) designs, are being evaluated to potentially support sustainable development, economic competitiveness, and to improve safety for nuclear plants [Locatelli et al. (2014)] as shown in Fig 1.1. Past experience, specifically, with regard to long term maintenance of the French Phenix reactors, has underlined the need to provide effective and reliable inspection of nuclear components [CEA, (2012)]. The use of liquid sodium coolant in NPP enable a low operating pressure and a high-power density although there are major challenges with regards to in-service inspection (ISI) and repair [Wang et al. (2012), Lubeigt et al. (2015)]. The opacity and electrical conductivity of a sodium coolant makes optical and electromagnetic techniques less effective for ISI of NPP components.

Figure 1. 1 Roadmap for Generation IV nuclear reactors

During the operation of a sodium fast reactor as shown in Fig.1.2, various system components can be displaced. For example, the head of a Fuel Sub-Assembly (FSA) as shown in Fig.1.3(a-b), may undergo a lateral shift from its original position due to the fast neutron induced damage to its structural material. Visualization based on ultrasonic imaging has been found feasible to guide robotic arm motion for the inspection of components [Day et al. (1973),
Lions et al. (1974), Taguchi et al. (1980)]. This technique of ultrasound-based scanning is called under sodium viewing in liquid sodium cooled fast reactors (SFRs). The approach was extensively used [Hans et al. (1984), McKnight et al. (1984), Swaminathan et al. (1990)] in a number of countries (especially US, India, Japan, France, Germany and Great Britain) for inspection in fast reactors from 1970 to the 1990s. During the same period the ASME also introduced an inspection code which has an emphasis on reliable methods for inspection of NPP components [ASME BP&V code (1992)]. The importance of non-destructive inspection was further highlighted by the accident at the Monju reactor [Mikami et al. (1996)] which resulted in a sodium leak. Since then, there have been many experimentally based studies reported which seek to establish and evaluate under-sodium viewing capabilities which are reviewed in next chapter.

![Sodium pool type fast reactor](image)

**Figure 1.2** Sodium pool type fast reactor

There is very limited historic data for ultrasound-based fatigue crack detectability in liquid sodium at elevated temperatures, and that which is available was provided by HEDL (Hanford Engineering Development Laboratory) as a result of work carried out in the 1970’s. HEDL reported that for ultrasonic transducers in liquid sodium (~260°C) notches could be found in all cases studied and 50% through wall fatigue cracks could also be detected. However, 25% through wall fatigue cracks in stainless steel samples which could be detected with the
transducer when the sample was in air could not be detected when it was immersed in liquid sodium [Mech et al.(1983)].

The major challenge in proving an effective under sodium viewing capability has been the development of ultrasonic transducers that can operate for an extended period at high temperature (HT), and are not damaged by at least gamma radiation in the reactor environment. The radiation effects on the sensitivity of piezoelectric transducer have been investigated in detail and reported in several studies [Holbert et al. (2005), Augereau et al. (2008), Parks et al. (2011), Rempe et al. (2014), Sinclair et al. (2015)].

The questions that remain unanswered relate to the limited detection capability due to constraints on the transducer performance as shown in Fig.1.4. Current work focuses on the analysis of the temperature effect on the ultrasonic transducer components. In the present work, Chapter 2 reports a technical review of the research and development work performed for under sodium viewing from 1970s. This review has helped to identify key fundamental issue needed to study regarding transducer sensitivity as a function of temperature as shown Fig.1.4. These causal factors limit the signal to noise ratio which has detrimental effect on the probability of detection at high temperatures.
Figure 1.4 Fundamental factors needed to study for development of robust high temperature transducer.

The methodology developed for the high temperature ultrasonic transducer in the current work is shown in Fig.1.5. The challenge of poor signal to noise ratio at high temperature is fundamentally analyzed through a unified modeling and measurements approach. Analysis of the transducer components is leveraged for prototype immersion transducer development and evaluation at high temperatures. Using the evaluation of prototype transducers, a revised design is proposed for the final outcome of the current work.

Figure 1.5 Methodology of the current work

The identification of challenges in Chapter 2 is followed by Chapter 3 which describes the resonance analysis for temperature sensitivity characterization of a piezoelectric disc. This numerical study demonstrates the sensitivity of each piezoelectric material coefficient towards
temperature and resonance frequency. In Chapter 4, temperature effect on the adhesive bond is analyzed with a 3-layer problem consisting of piezoelectric material, adhesive and substrate. The methodology presented in this work could potentially be used to understand long-standing interface issue in high temperature ultrasonic transducers. In Chapter 5, a prototype of immersion high temperature transducer is developed and evaluated in different surrogate fluids. During the benchmarking process of the available transducers, a unique bimodal response was found for which lead to further investigation on effect of adhesive and faceplate thickness on the bimodality. The effect of temperature on the immersed transducer sensitivity was also demonstrated. Utilizing insights from measurements, a revised design is proposed which demonstrated ultrasonic imaging capability required for USV of SFRs. Temperature dependent reduction in the transducer sensitivity limit the probability of detection (POD). Chapter 6 reports a novel temperature compensated transfer function approach to estimate reduction in probability of detection at high temperature using finite element model and room temperature experimental data. Lastly, Chapter 7 and 8 summarizes the conclusions of current work followed by a discussion on the future work.
CHAPTER 2. BACKGROUND

To enable safe operation of advanced reactors, in-situ NDE measurements are becoming important for structural health monitoring (SHM) of the reactors. The various advanced reactor concepts include sodium-cooled fast reactors (SFR), gas-cooled high temperature reactors, and molten salt reactors, and are collectively often referred to as Gen-IV reactors. Ultrasonic technology has the potential to allow inspection with the sensors fully immersed in liquid metal and molten salt coolants. The most basic form of the ultrasound inspection is the pulse-echo A-scan, which can give a wealth of data on defects, changes in density, elastic moduli, and ultrasonic attenuation. Moreover, ultrasonic C-scan is useful in imaging of the reactor components. For this purpose, the transducer should be able to perform at an elevated temperature and produce repeatable measurements. Consequently, it is required to select materials which have an operating temperature greater than the hot stand-by temperature of the SFR (~260°C). It is seen that at high temperature, the sensitivity of a transducer becomes an issue which affects the signal to noise ratio (S:N).

Development of high temperature ultrasonic transducers for under sodium viewing has been performed since the 1970s. Current work presents a technical review of the research and development of ultrasonics transducer for under-sodium viewing. The objective of this chapter is to summarize the efforts in terms of design, characterization for transducer components and signal processing techniques of the low S:N ultrasonic echoes.

Scope of the current literature review:

- Ultrasonic transducer assembly for under-sodium viewing (USV)
- Ultrasonic imaging scanner in liquid sodium
- Beam forming methods and Digital filtering techniques
2.1 Ultrasonic Transducer assembly for USV

Fast reactors (as well as accelerator-driven reactors) use liquid metal as the core coolant and as the heat transfer fluid from the reactor core to the steam generator. While a variety of liquid metal coolants (e.g., mercury, sodium, lead bismuth) have been used worldwide, the current review focuses on the liquid sodium fast reactors. The thermo-physical properties of liquid sodium has effects on ultrasound transduction. Previous studies have shown experimental data for variation in these properties with increasing temperature. A summary of liquid sodium properties relevant to transducer development are reported by Foust (1972) and Leibowitz (1971). Relevant data are given in Table 2.1 as shown below.

Table 2.1 Thermo-physical properties of liquid sodium

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Density (kg/m³)</th>
<th>Dynamic viscosity (Pa·s)*10^{-4}</th>
<th>Speed of sound (m/s) [Foust(1972)]</th>
<th>Thermal conductivity (J/kg·K)</th>
<th>Speed of sound (m/s) [Leibowitz (1971)]</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>9269</td>
<td>6.802</td>
<td>2526</td>
<td>86.9</td>
<td>2524</td>
</tr>
<tr>
<td>150</td>
<td>9153</td>
<td>5.415</td>
<td>2500</td>
<td>84.4</td>
<td>2497</td>
</tr>
<tr>
<td>200</td>
<td>9036</td>
<td>4.519</td>
<td>2474</td>
<td>82</td>
<td>2470</td>
</tr>
<tr>
<td>250</td>
<td>8918</td>
<td>3.900</td>
<td>2448</td>
<td>79.6</td>
<td>2443</td>
</tr>
</tbody>
</table>

Leibowitz (1971) also calculated the speed of sound in liquid sodium up to 1000°C which is given as:

$$c = 2577.6 - 0.536T$$  \hspace{1cm} (2.1)

where $c$ is the velocity of sound in liquid sodium and $T$ is temperature. This equation has been used to give data for modeling of ultrasonic propagation in turbulent liquid sodium with a temperature gradient [Massacret et al. (2014)]. The percentage difference between the speed of sound given by equation (2.1) and as measured by Foust and Liebowitz is less than 1%. Furthermore, when the values of speed of sound in liquid sodium at 250°C from Table 2.1 are
compared with value given by Dierckx, et al. (2014), there is less than 1% of difference. By linear fitting the data with coefficients as determination by equation (2.1), the density, thermal conductivity and dynamic viscosity variation in liquid sodium with temperature is given as

\[ \rho = 951.72 - 0.2396T \]  
\[ J = 91.862 - 0.0496T \]  
\[ \eta = 0.0009 - 3 \times 10^{-6}T - 2 \times 10^{-12}T^3 + 4 \times 10^{-9}T^2 \]

where \( \rho \) is density, \( J \) is thermal conductivity, \( \eta \) is the dynamic viscosity.

In an ultrasonic transducer, electrodes are typically made from gold, silver or platinum are deposited on the piezoelectric disc. The change in the transmission coefficient due to these layers is often not considered due to their small thickness. The thickness of the piezoelectric element is half-wavelength based on the centre frequency of the transducer. In NDT, typically a broad bandwidth pulse is required for which minimizing ring-down of the initial incidence pulse is a critical challenge. The generated wave is reflected back and forth in the transducer layers which decreases the ultrasonic energy transmitted as shown in Fig. 2.1.

![Figure 2.1 Schematic of the ultrasonic transducer for measurements in fluid](image)

To reduce this acoustic noise, a damping material or backing is added behind the piezoelectric element as shown in the Fig. 2.1. Ideally the acoustic impedance of the backing
material should be equal to that for the piezoelectric material. In such a case, the wave travelling into the backing will be attenuated due to the damping coefficient of the backing material which can reduce the resulting ring down. However, adding backing reduces the mechanical quality factor $Q$ and also reduces the energy transmitted into the sample. This limits the input power and hence the signal to noise ratio.

For extended operation of the transducer at high temperatures, two approaches of design have been previously investigated: a) waveguide-based transducers and b) fully immersion transducers as shown in Fig.2.2(a-b) respectively. A waveguide acts as a buffer rod and isolates the transducer piezoelectric element from the high temperature. Recently, a flexible or re-configurable waveguide concept has been developed for temperature sensing applications [Periyannan et al. (2016)]. The challenges with ultrasonic transducers using a waveguide are the limitations in deployment and also the spurious echoes which occur due to the length and shape of the waveguide. These spurious echoes decrease the resolution of the resulting ultrasonic scan and hence, the defect detection ability.

Figure 2.2 a) Waveguide based ultrasound transducer [Daw et al.(2013)] b) Single element immersion transducer [Ensminger and Bond (2011)]
2.1.1 Faceplate

To maximize the energy transmitted into the fluid medium shown in Fig 2.3 [Zhu (2008)], it is necessary to perform acoustic impedance matching which can be obtained using a front surface matching layer which is also called faceplate or front plate of the transducer.

![Figure 2.3 Schematics of ultrasonic transducer in the fluid medium [Zhu (2008)]](image)

Under the plane wave assumption, consider a wave reflected normally from the interface between two media of different acoustic impedances $Z_{01}$ and $Z_{02}$ and the geometry shown in Fig 2.4. [Kino, (1970)].

![Figure 2.4 Reflection and transmission of a plane wave at an interface [Kino, (1970)]](image)

If the thickness of input layer -1 is quarter wavelength, then equivalent input impedance $Z_{in}$ at layer 1 is for complete transmission (transmission coefficient=1) is given by
$Z_{in} = \frac{Z_{01}^2}{Z_{02}} \text{ or } Z_{01} = \sqrt{Z_{in} * Z_{02}}$ (2.5)

However, the faceplate is also the medium which isolates the transducer from the fluid in which the transducer is immersed. From this perspective, wetting of the front surface of the transducer with the fluid is imperative to radiate ultrasound from the transducer into the fluid for performing ultrasonic imaging. This is one of conundrum in the design of ultrasonic transducer for under sodium viewing. In addition to the acoustic impedance matching criteria, the wetting ability of front surface with liquid sodium plays equally important role. Table 2.2 lists faceplate materials used previously for USV transducers.

**Table 2.2** Faceplate materials in the transducer for under sodium viewing (USV)

<table>
<thead>
<tr>
<th>Faceplate material</th>
<th>Observation</th>
<th>Reference literature</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gold coating</td>
<td>Gold sputtering layer sustains up to 1000°C in liquid sodium</td>
<td>[Baba et al. (2016)] [Day and Smith (1973)] [Bond et al. (2012)]</td>
</tr>
<tr>
<td></td>
<td>Wetting time less than 5 seconds at 500°C in liquid sodium</td>
<td></td>
</tr>
<tr>
<td></td>
<td>The gold layer dissolves requiring re-coating for every inspection</td>
<td></td>
</tr>
<tr>
<td>Stainless steel</td>
<td>Slow wetting in liquid sodium</td>
<td>[Tarpara et al. (2016)]</td>
</tr>
<tr>
<td>Polished nickel/nickel</td>
<td>Immediate wetting in liquid sodium at 180 and 260°C Alloy 52 CTE matches</td>
<td>[Swaminathan (1990), Griffin et al. (2011)] [Griffin et al. (2009)]</td>
</tr>
<tr>
<td>alloy</td>
<td>with Bismuth titanate</td>
<td></td>
</tr>
<tr>
<td>Invar</td>
<td>CTE matches with piezoelectric ceramics</td>
<td>[Karasawa et al. (2000)]</td>
</tr>
<tr>
<td>Titanium</td>
<td>CTE matches with Bismuth titanate</td>
<td>[Griffin et al. (2009)]</td>
</tr>
</tbody>
</table>

Figure 2.5(a) shows an experimental set-up used to investigate the reactive wetting of metallic plating materials by liquid sodium at 250°C for the ultrasonic sensor of the under-sodium viewer. Kawaguchi et al. (2014) also simulated the liquid sodium wetting by modeling the reactive and non-reactive wetting of metallic-plated steels as shown in Fig.2.5(b)
2.1.2 Bonding Agent

It has been reported in the literature that 60% of high temperature transducers have an adhesion problem between layers. It is therefore necessary to select adhesives and/or other bonding techniques which can provide structural integrity at high temperatures. When reviewing the literature, there are three different concepts commonly used for acoustic coupling in transducers [Kazys et al. (2008)].

- **Dry coupling:** High quality surface finish is required. Intimate contact with at worst very small air gaps (>0.01 micron) between a piezoelectric element and a front faceplate. Even a small gap can cause a substantial reduction in energy of the transmitted wave. Moreover, a
half-wavelength thick piezoelectric element may not withstand high pressure that may be needed for dry coupling.

- Liquid coupling: Silicone oil has been used successively up to 250°C although it evaporates gradually. During long term operation of an ultrasonic transducer, the liquid may flow out of the interfaces due to vibrations of a piezoelectric element. Couplant E (ultra-therm) has already been used in the temperature range of 260 to 540°C. A major issue with using the specialized high temperature couplants is that they tend to solidify which reduces the energy being transmitted.

- Solid coupling: In this type of coupling several approaches can be used- soldering, diffusion bonding, cementing, and epoxy bonding. The toughness of elastic solder should be able to compensate for differential thermal expansion due to the piezoelectric element, backing element and matching layer. Table 2.3 summarizes the high temperature bonding agent that could be potentially be used for development of USV transducers.

For bond-line monitoring of these bonding agents, electromechanical impedance method is being used for room temperature SHM applications (Dugnani et al, 2016). This method shown in Fig.2.6, utilizes change in the electrical impedance of piezoelectric material for bond-line monitoring demonstrated by a numerical study in Appendix A.

**Figure 2.6** Bond-line monitoring using EMI method [Baptista et al. (2014)].
### Table 2.3 Bonding agents for high temperature application

<table>
<thead>
<tr>
<th>Bonding agent</th>
<th>Operating temp.</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alumina Resbond 989 (cotronics)</td>
<td>1648ºC</td>
<td>High electrical resistance, CTE:8.1*10^-6/ºC</td>
</tr>
<tr>
<td>Duralco 124</td>
<td>343ºC</td>
<td>Silver based epoxy, electrically conductive, cure cycle: 4hrs at 121ºC</td>
</tr>
<tr>
<td>Duralco 4703</td>
<td>343ºC</td>
<td>High electrical resistance, CTE=3.9*10^-5/ºC</td>
</tr>
<tr>
<td>Duralco 952, 954</td>
<td>1093ºC</td>
<td>Aluminum, SS based adhesive, partially offer ductility and impact resistance associated with soldering, welding.</td>
</tr>
<tr>
<td>Epotek 353ND</td>
<td>250ºC</td>
<td>Glass transition temperature&gt;90ºC</td>
</tr>
<tr>
<td>Pyroduct™ 597A</td>
<td>927ºC</td>
<td>Silver based electrically conductive epoxy paste [Giurgiu et al. (2010)]</td>
</tr>
<tr>
<td>Polybenzimidazole (PBI plastic)</td>
<td>343ºC</td>
<td>Celazole®, Thermoplastic, glass transition temperature:399ºC CTE:2.34*10^-5/ºC</td>
</tr>
<tr>
<td>Glass solder</td>
<td>500ºC</td>
<td>At high temp., the solder reacts with transducer components except for gold and platinum [Kazys 2008]</td>
</tr>
<tr>
<td>Ti Braze Al-665 foil</td>
<td>650ºC</td>
<td>Brazed foil successfully used by [Amini et al. (2016)] for continuous inspection</td>
</tr>
<tr>
<td>Silver solder</td>
<td>NA</td>
<td>Used for transducer testing in liquid sodium up to 250 ºC [Griffin et al. (2011)]</td>
</tr>
<tr>
<td>Silicone oil</td>
<td>250ºC</td>
<td>Evaporates gradually [Kazys et al. (2008)].</td>
</tr>
<tr>
<td>88Au-12Ge solder</td>
<td>356 ºC (melting point)</td>
<td>Offers best solder alloy for joining dissimilar metals (Hosking,1999)</td>
</tr>
</tbody>
</table>

### 2.1.3 Piezoelectric Material

In an ultrasonic transducer, a piezoelectric material produces surface charge when a mechanical stress is applied which results in a mechanical wave (direct effect) and conversely produces electrical voltage due to mechanical deformation (inverse effect).

For the transducer applications, the direct piezoelectric effect is given by

\[
d_{ijk} = \left( \frac{\partial D_i}{\partial T_{jk}} \right)_E; \quad g_{ijk} = - \left( \frac{\partial E_i}{\partial T_{jk}} \right)_D
\]  \hspace{1cm} (2.6)

where \(d_{ijk}\) and \(g_{ijk}\) are the piezoelectric charge and voltage coefficient, \(D_i\) is the electric displacement, \(E_i\) is the electric field strength and \(T_{jk}\) is the mechanical stress component.
High temperature piezoelectric material can be broadly classified as ferroelectric material and non-ferroelectric materials. The ferroelectric materials exhibit different material structures namely a) Perovskite b) Tungsten Bronze Structure c) bismuth layer structure, properties of which has been discussed in detail by Zhang et al. (2011). The curie temperature of high temperature piezoelectric materials is listed in the Table 2.4.

Table 2.4 High temperature piezoelectric materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Curie temperature (°C)</th>
<th>Reference literature</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminum nitride</td>
<td>2200 (melting point)</td>
<td>Reinhardt et al. (2017)</td>
</tr>
<tr>
<td>Barium titanate</td>
<td>120</td>
<td>Zhang and Yu (2011)</td>
</tr>
<tr>
<td>Bismuth titanate</td>
<td>600/820</td>
<td>Reinhardt et al. (2017)</td>
</tr>
<tr>
<td>Bismuth scandium oxide- lead titanate (BiScO₃-PbTiO₃)</td>
<td>430</td>
<td>Zhang and Yu (2011)</td>
</tr>
<tr>
<td>Gallium orthophosphate</td>
<td>970 (phase transition)</td>
<td>Damjanovic (1998)</td>
</tr>
<tr>
<td>Lead metaniobate</td>
<td>400-540</td>
<td>Zhang and Yu (2011)</td>
</tr>
<tr>
<td>Lead zirconium titanate</td>
<td>250-360</td>
<td>Zhang and Yu (2011)</td>
</tr>
<tr>
<td>Lithium niobate</td>
<td>1142-1210</td>
<td>Zhang and Yu (2011)</td>
</tr>
<tr>
<td>Langasite</td>
<td>1200-1550 (melting point)</td>
<td>Damjanovic (19908)</td>
</tr>
<tr>
<td>Yttrium calcium oxyborate (YCOB)</td>
<td>1510 (melting point)</td>
<td>Jiang et al. (2013)</td>
</tr>
<tr>
<td>Zinc oxide</td>
<td>1975</td>
<td>Suprock (2016)</td>
</tr>
</tbody>
</table>

Piezoelectric materials typically do not exhibit a center of symmetry. Without the application of external electric field, the center of positive and negative charges may not coincide which is also dependent on the crystal structure. This results in a spontaneous polarization. Ferroelectrics is a class of piezoelectric material which exhibit reorientable spontaneous polarization. At elevated temperatures, the asymmetric structure of these materials converts to a highly symmetric crystal structure which causes loss of polarization. For ferroelectric materials, this polarization is produced by the domain walls of the structure. The stability of these domains is thermally dependent. As the temperature increases, the
orientation of the domains become more random which causes loss in polarization reducing the piezoelectric effect. [Zhang et al. (2011)]

Furthermore, elevated temperatures lead to the phase transition of some of the piezoelectric materials resulting into instability of parameters that include dielectric, piezoelectric and mechanical properties. The stability of these properties up to at least 300°C thus becomes critical to select piezoelectric material for transducers to be used in under-sodium viewing.

Hence, for developing the transducers for under-sodium viewing, it is important to characterize the temperature dependence of piezoelectric material properties. These properties can be measured using resonant ultrasound spectroscopy method (RUS). RUS is fundamentally an inversion problem which uses optimization method to minimize the error between initial guess of a material parameter and experimental value at a particular temperature. Sabat et al. (2007) and Tang et al. (2015) utilized RUS to demonstrate the temperature dependence of the piezoelectric properties.

2.1.4 Backing Material

Backin material attenuates the wave motion caused by transmission and reflection in piezoelectric and multiple transducer materials. The attenuation of ultrasonic wave can be achieved through two mechanisms: a) absorptive damping and b) attenuation due to multiple scattering. The absorptive damping is due to viscoelasticity and it is affected by increase in the temperature. However, multiple scattering due to geometrical shape, inclusions and porosity in the backing material has also demonstrated capability for high temperature applications as shown in Fig.2.7(a-b). Kazys et al. (2005) developed concaved shaped metallic backings for immersion transducers in liquid metal as shown in Fig.2.7(a). Amini et al. (2015) demonstrated application of porous ceramic backing material for transducer used for inspection up to 800°C as shown in Fig.2.7(b).
Figure 2.7 Attenuation of backward travelling at high temperature using (a) concaved metallic (b) porous ceramic backing.

Attenuation due to such backing material can reduce the pulse length and increase axial resolution in the ultrasonic scanning. Moreover, shorter pulse length also indicates broader bandwidth increasing the detectability of defects of size. However, attenuation also causes reduction in the peak amplitude of echoes received by the transducer. Hence, a trade-off is needed between resolution and the peak amplitude criteria. High temperature transducers developed originally by Hanford Engineering development lab (HEDL) used woven wire metallic disc as a backing material whereas PNNL (2011) used steel foam as the backing material as shown in Fig.2.8(a-b) respectively. Lhuillier et al. (2011) applied back plate for attenuation of acoustic noise in the high temperature immersible transducer as shown in Fig.2.9. Characterization of these backing materials at room temperature can be performed using through-transmission method as shown in Fig. 2.10 (a-b) at room temperature to obtain a baseline attenuation coefficient.
2.1.5 Cabling

A two conductor, mineral insulated (MI), co axial cable has been used consistently in under sodium viewing transducers [Taguchi et al. (1980), Griffin et al. (2009)]. Impedance matching
of cable with ultrasonic pulsar-receiver is necessary to reduce the insertion loss. Moreover, these matching circuits have demonstrated low gamma radiation resistance [Sinclair (2015)]. The length of cable used in under sodium viewing has varied from 6 to 15m [Kazys et al. (2006a), Swaminathan et al. (2012)]. Hence, the capacitance of the cable should be low to minimize loading to the pulsar and attenuation of the excitation pulse. In MYRRHA reactor (Belgium), a MI coaxial cable was used up to 900°C [Kazys et al. (2006b)]. The coaxial cable consisted of stainless steel with magnesium oxide insulators. Using these cables, under dose rates in the range of 10-30kGy/h, an underwater gamma irradiation test was carried out with 1mm diameter and 15m long high frequency cable. Glass et al. (2016) has summarized the cable degradation measurement techniques such as tangent loss, indenter modulus measurement and dynamic mechanical analysis as shown in Fig.2.11 (a-c) respectively. These methods help monitor cable degradation which is important to estimate the insertion losses in the transducer due to cabling.

![Image]

**Figure 2.11** Characterization of cabling degradation a) tangent delta instrument b) Indenter modulus measurement c) Dynamic mechanical analysis [Glass et al. (2016)]

### 2.1.6 Acoustic Waveguides

Acoustic waveguide for high temperature measurement act as a buffer rod which isolate the sensing piezoelectric element and other transducer components from high temperature.
These waveguides are basically a mechanical structure which support elastic wave propagation from the transducer to the target specimen. The thermal isolation of the piezoelectric element keeps the material at low temperatures offering better reliability. However, due to the length and shape of waveguides, spurious echoes are generated due to multiple reflection resulting into acoustically noisy signal. There have been multiple geometries of the waveguides used in the transducers for USV which are summarized as below:

- **Smooth rod**: For Liquid metal fast breeder reactor, Anderson (1972) designed steel cylindrical rod waveguide which was attached to the transducer as shown in Fig.2.12. However, sidewall reflection from a smooth rod can cause mode conversion of longitudinal wave to shear wave resulting into transmitted energy loss in the fluid in addition to spurious echoes discussed previously. For this reason, modifications in the geometry was needed to reduce the side wall reflections in the waveguide.

*Figure 2.12 Cylindrical steel waveguide developed for LMFBR inspection [Anderson (1972)]*
- **Threaded rod**: Sidewall reflection in the waveguide tube cause mode-conversion causing energy loss and spurious echoes. The external surface of the waveguide can be threaded to attenuate these side wall reflections in the smooth rod. [Lynnworth (1989)]

- **Focused clad rod**: A clad rod is basically a buffer rod which consists of a double tapered shape with mild steel core and thermal sprayed inner coating at the inner cladding. The outer cladding is made up of bronze. This concept has been successfully used for high temperature ultrasonic imaging in molten melts. [Ihara et al. (2000), Rehman et al. (2001), Ono et al. (2002), Ono et al. (2003,2004, 2006)] as shown in Fig.2.13.

![Focused Clad rod](image)

**Figure 2.13** Focused Clad rod [Ihara et al. (2000)]

- **Bundled waveguide**: This type of waveguide assembly provides dispersion free wave propagation structure as well as acts as a thermal isolation between hot fluid and the transducer. It consists of metal rods with thickness less than the operating wavelength. The rods are closely packed within a sheath and welded closed at each end as shown Fig. 2.14.

![Bundled waveguide](image)

**Figure 2.14** Bundled waveguide [Bohemer et al. (1975)]
Wang et al. (2012) performed under sodium measurements using bundled waveguide which resulted in spurious echoes in which the reflected signal from target is suppressed in electrical noise as shown in Fig. 2.15.

![Bundled waveguide design for under sodium viewing](image)

**Figure 2.15** Bundled waveguide design for under sodium viewing [Wang et al. (2012)]

- **Spiraled sheet waveguide:** A spiraled sheet waveguide is basically a coiled structure with thickness considerable less than the wavelength corresponding to the center frequency of transducer [Boehmer et al. (1976)]. This minimizes wave interference and attenuation due to scattering and dispersion. A similar approach was reported by Periyannan et al. (2016) recently with a re-configurable spring helical waveguide for temperature sensing as shown in Fig. 2.16(a). In these type of waveguides, the transducer is attached at one end whereas the other end is the attached to the surface of specimen being inspected. A-scan data using spiraled sheet rod in liquid sodium is shown Fig.2.16(b).

![Spiraled-sheet Rod](image)

**Figure 2.16** (a) Re-configurable spring like helical waveguide [Periyannan et al. (2016)] (b) A-scan data in liquid sodium using spiraled sheet rod [Wang et al. (2012)]
• **Spiral+bundled geometry:** To overcome the limitation of spiral and bundled waveguides Wang al. (2012) reported hybrid waveguide design for 5MHz transducer. The A-scan of the transducer in liquid sodium using this waveguide is shown in Fig. 2.17.

![A-scan with a Hybrid waveguide transducer immersed in liquid sodium](image)

**Figure 2.17** A-scan with a Hybrid waveguide transducer immersed in liquid sodium [Wang et al. (2012)]

Joo et al. (2013) also developed a beryllium coated high temperature waveguide transducer with an acoustic shielding tube as shown in Fig. 2.18(a-b). The 10-m long waveguide has a lower end section bent at an angle to transmit the beam in the desired direction as shown in Fig. 2.18(b).

![Beryllium coated waveguide transducer](image)

**Figure 2.18** Beryllium coated waveguide transducer (a) Schematic (b) Prototype [Joo et al. (2013)]
2.1.7 Bench-top Experiments for USV

One of the major experimental challenges in the performance evaluation of HT immersion transducers for sodium fast reactors is safely working with liquid sodium for bench-top experiments. This is due to the chemical reactivity of liquid sodium which explodes upon contact with water or burns with air, increasing the risk of accidents [Mikami et al. (1996)]. An experimental set-up developed by Argonne National laboratory is shown in Fig.2.19(a) which includes a dump tank for removal of liquid sodium and prevention of exposed to air.

Figure 2.19 Benchtop under sodium experiments (a) Schematic indicating two separate test tank and dump tank (b) At Argonne National laboratory [Wang et al. (2012)] (c) At Korea Atomic Energy institute [Kim et al. (2014)] (d) At Pacific Northwest National Laboratory (PNNL) [Bond et al. (2012)]
The experimental set-up also shows a separate holder for target and transducer in a jacketed tank as shown in Fig.2.19(b). A similar support guide set-up was also reported by Kim et al. (2014) as shown in Fig.2.19(c). However, the transducer was immersed in an open, metallic vessel containing liquid sodium also shown in Fig. 2.19(d). The measurements were performed at 260°C which is same as the hot stand-by mode temperature of sodium fast reactors. The target specimen used for defect and component identification is shown in Fig.2.20(a-d). As seen from Fig.2.20(a), the surface of the specimen could be machined to introduce thickness variation in the sample to evaluate the ability of under-sodium detection at high temperature. A mock-up of a fuel sub assembly can also be prepared as shown in Fig.2.20(b-c) and arranged in a honeycomb arrangement as shown in Fig.2.20(d).

Figure 2.20  Target specimen for ultrasonic imaging in laboratory experiments (a) for defect identification Wang et al. (2012) (b) component identification Wang et al. (2012) (c) core mock up (d) Fuel sub assembly (FSA) heads [Chaitanya et al. (2017)]
2.2 Ultrasonic Imaging in Liquid Sodium Fast Reactor

For ultrasonic imaging of reactor components, the transducer needs to move a distance of 3 to 8m before reaching the region of the target specimen [Swaminathan et al. (2012)]. For this purpose, long, robotic scanner arms are needed for transducer manipulations. Ideally, the reactor designs should have a provision for sufficient space to be occupied by the scanner assembly. However, conventionally, scanner assemblies are designed to occupy space sufficient to keep at a safe distance from the nuclear components. Since the 1970’s most of the scanner geometries have focused on the sweeper arm scanner (SAS) concept which allows translational and rotational degree of freedom for ultrasonic imaging which will be discussed in this section.

The initial designs focused on sweeper arm design with single element transducer developed by Swaminathan et al. (1990) who deployed the prototype ultrasonic viewer to scan the space below the core cover plate mechanism (CCPM) of the fast breeder test reactor. The viewer was a 10m long and 33mm diameter stainless (Spinner) tube inside a 90 mm diameter guide tube. This design consisted of SAS with transducer is horizontal orientation. Toshiba Corporation, Japan and Power Reactor and Nuclear Fuel Development Corporation (PNC), Japan jointly developed a vertical SAS single transducer which was tested in air at room temperature and in liquid sodium at 250°C [Taguchi et al. (1980)]. Under-sodium image of the test object using the vertical SAS transducer is shown in Fig.2.21(a-b). Hanford Engineering Laboratory (1972) developed SAS assembly with multiple transducers (8) for under sodium viewing as shown in Fig.2.22.
Figure 2.21 (a) Test object (b) Under sodium image at 300 C [Taguchi et al. (1980)]

Figure 2.22 Sweeper Arm scanner with multiple (8) transducers for Fast flux test facility [Ord and Smith (1972)]

Wessels et al. (1981) developed a different multiple probe design for USV in 1980-90’s which consisted of nine transducers. It comprised of a central transmitter probe surrounded by eight receiver probes. All the transducers consisted of focusing lens made up of steel membranes. The axis of the receiver transducer is inclined with respect to the axis of central transducer in such a way that focal point of all the transducers is in the same point of space as shown in Fig. 2.23 (a-b).
Hans et al. (1984) also developed linked sweep arm scanner called snr 300 which consisted of 2.5m long offset arm and multi-head transducer. Servo meters were used for rotation of arm and radial movement of multi-head housing. The housing can contain downward viewing transducers as well as the side viewing transducer. Ford et al. (1996) also reported a linked sweeper arm design as shown in Fig.2.24.
Bhabha Atomic Research Center (BARC), India developed an air-couple ultrasonic system for calibration of the USV system [Patankar et al. (2009)]. The system consists of transmission and receiver pair connected by a tube filled with Argonne gas as shown in Fig. 2.25(a). The transmitted ultrasonic signal travels through the gas and is received by the transducer on the other end confirming proper functioning of both of the transducers. IGCAR and BARC, India developed a conical housing to encapsulate the transducers as shown in Fig. 2.25(b-c).

### 2.3 Beam-forming methods

Beam forming methods use an array of sensors for directional transmission and reception of ultrasonic signals. During transmission, focusing of ultrasound is achieved by introducing a time delay in the emitted signals from each sensor in the array. The delayed echo signals arriving at the different sensing elements are acquired to create an isophase plane. Also, the receiver beamformer creates a pattern of beams pointing in the same direction in a similar way.
These aligned echoes are summed coherently which is also known as delay-sum approach in basic signal processing [Tarpara et al. (2016)].

2.3.1 Phased array imaging

In this technique, transducer elements are used in transmit and receive mode. Focusing and steering of the ultrasonic beam is achieved electronically using delay and apodization law [Tarpara et al. (2016)]. The two-dimensional image of the specimen is constructed in real time by steering the focal point of the beam at an increasing rate. The pitch between phased array elements influences the grating lobes in the generated ultrasound beam. If the pitch distance is less than half the wavelength ($\lambda$), the grating lobes become independent of the steering angle. The under-sodium viewing transducer are generally of frequency 2-5MHz which reduces the required pitch distance causing fabrication of the transducer assembly to be a challenge. Pacific Northwest National laboratory (PNNL) has used two matrix arrays with each of the 30 elements in 10 x 3 configuration for C-scan of target in water at room temperature [Larche et al. (2016)].

2.3.2 Coherent array imaging

Johnson et al.(2005a-b) proposed a coherent array imaging method which reduces the complexity of front end software when compared with that for a full phased array (FPA). This method performs partial transmit and receive beamforming using a subset of adjacent element at each firing step. This method which is also called phased sub array (PSA) imaging approaches the quality of FPA imaging while reducing the front-end hardware.
2.3.3 Sparse array imaging

This technique is based upon the optimal use of a number of elements by placing them in aperiodic sampling pattern. This pattern also reduces the grating lobs in the radiated ultrasound beam. One directional curved array [Kirkebø et al. (2007)], and vernier array [Nikolov et al. (2000)] can be used to reduce the side lobes as well.

2.3.4 Sampling phased array imaging

In this method, a single transducer generated ultrasonic pulse and the reflected echo signals are captured by the all the receiving transducer. This technique provides higher sensitivity for the inspection and increases probability of detection of small discontinuity in highly stressed materials [Verkoijjen (2008)]. Moreover, it can also be used for the real time three-dimensional visualization of the inspection volume [Pudovikov (2010)].

2.3.5 Synthetic aperture imaging

This method is based upon the delay and sum-based signal processing discussed previously. This technique allows building virtual aperture by synthesizing into small real apertures that improves lateral resolution without sacrificing the imaging frame rate. Karasawa et al. (2000) used synthetic aperture focusing technique (SAFT) for 3D under sodium viewing at 200C using the 36x36 matrix arrayed transducer. This work demonstrated a circumferential resolution of <2mm and axial resolution of <0.5mm in liquid sodium environment.

2.3.6 Coded excitation imaging

In ultrasonic imaging there is a tradeoff between penetration depth and spatial resolution due to attenuation associated with the center frequency increase. This limitation is partially overcome by increasing the amplitude of the transmitted energy. In coded excitation imaging, the excitation pulse duration is increased to enhance the total transmitted energy [Chiao et al. (2003)]. Pulse compression is subsequently used to restore axial resolution after the
excitation. The main advantage of coded excitation is the improvement in the signal to noise ratio (S:N). However, it is accompanied by an increase in the side lobes which reduces contrast in the side lobes.

2.4 Digital filtering techniques for low SNR ultrasonic signals

Application of digital filters can provide an economic solution for improving S:N by regularization of the random noise in the signal. Sinding et al. (2016) discussed the ability of total variation (TV) and Tikhonov regularization methods for the ultrasonic echoes. However, Savitzky-golay (SG) smoothing filters have not been explored in the field of high temperature ultrasonic NDE. It should be noted applying digital filters basically refers to data smoothing which assumes the measuring variable is gradually varying and is corrupted by the random noise.

The Savitzky-Golay (1964) filter is based on the least squares polynomial fitting across a moving window within the data in the time domain [Guiñón, et al (2007)]. Typically, a long polynomial, and a moderate order polynomial allows a high level of smoothing without attenuation of signal features of interest. Detail description on SG filer is given by Schafer (2011). The basic idea behind SG filter is least squares local polynomial smoothing. Consider 2M+1 samples of signal x[n] centered at n=0. The coefficients of the polynomial can be given by

\[ p(n) = \sum_{k=0}^{N} a_k n^k \]

Equation (2.8) minimizes the mean squared approximation error of the group of input samples centered at n=0. This error is given by Guiñón et al. (2007) as:
TV denoising is an approach for reduction of noise so that sharp edges in the underlying signal can be preserved. By minimizing a cost function, the output of TV denoising filter can be obtained. This basically represents an optimization problem. TV denoising estimates $x[n]$ by solving an optimization problem [Selesnik et.al (2010,2012)].:

$$\arg \min_{x} \left\{ F(x) = \frac{1}{2} \sum_{n=0}^{N-1} |y(n) - x(n)|^2 + \lambda \sum_{n=1}^{N-1} |x(n) - x(n-1)| \right\}$$

(2.9)

The regularization parameter $\lambda$ is greater than zero and controls the degrees of smoothing. Increasing $\lambda$ gives more weightage to the second term in the equation (2.9). This second term measures the fluctuations in the signal $x[n]$. Such signal processing techniques can facilitate smoothing of the raw signal to minimize the random noise affecting S:N.

2.5 Summary

This review underlined the range of projects which were performed since 1970’s seeking to develop under-sodium viewing transducers. It can be seen that various methods such as phased array which are widely used at room temperature inspections, were also applied at high temperatures. However, it is concluded after the review that the fundamental issue still remains in the acoustical response of degrading transducer components at elevated temperatures. These components are can be mainly classified as: a) piezoelectric material b) bonding agent c) faceplate d) cabling and electrical continuity. The temperature dependent sensitivity of these components is important to understand and when seeking to develop a fundamentally robust transducer for continuous inspection in advanced test reactors. Hence, this review serves to identify design needs for an optimum signal to noise ratio which are summarized as below:
- **Piezoelectric material:** In addition to the high Curie temperature, optimum piezoelectric properties are needed at the hot stand-by temperature (260°C) of the sodium fast reactor. This will potentially give better signal strength at 260°C.

- **Faceplate:** The transducer faceplate material acts as a design constraint and this driven by wetting ability of the front of transducer with the liquid sodium. Currently, nickel appears to be best option to address this constraint. However, nickel introduces an acoustic impedance mismatch between the low impedance fluid medium and high impedance transducer components attached to the faceplate. This will cause multiple reflections in the transducer signal leading to acoustic noise in the ultrasonic C-scan. Hence, an impedance matching scheme is required to minimize the losses in the acoustic noise.

- **Acoustic coupling:** Degradation of acoustic coupling is a major concern for long-term operation of USV transducers. There have been multiple techniques applied for acoustic coupling between transducer layers which might sustain at high temperature but do not maintain transmissivity for wave motion. An innovative approach is needed to minimize the losses due to acoustic coupling at high temperatures. Sol-gel method could potentially help to eliminate the dependence of the bonding agent used in the USV transducers.

- **Electrical continuity:** Poor electrical continuity can introduce random noise into the signal reducing the signal to noise ratio and hence, the detectability of defects. Particularly, an effective electrical grounding is critical for reduction of the electrical noise. Understanding cable degradation and measurement techniques are also critical to relate and understand the insertion losses in the transducer.
Moreover, improvement in reliability and performance of these components can be accompanied by real time digital signal processing to further improve the resolution of under sodium images.

In this way, this literature review underlines the need for fundamental research on temperature effect on transducer components before developing a technology-ready level USV transducers for enhanced safety of sodium fast reactors.
CHAPTER 3. RESONANCE ANALYSIS FOR SENSITIVITY CHARACTERIZATION

Piezoelectric based ultrasonic transducers for high temperature (200°C+) applications are a key enabling technology for energy, chemical and petro-chemical industries. In this chapter, experimental data-based analysis is performed to investigate the fundamental causal factors for the resonance characteristics of a piezoelectric disc at elevated temperatures. The effect of all ten temperature-dependent piezoelectric constants ($\varepsilon_{33}, \varepsilon_{11}, d_{33}, d_{31}, d_{15}, s_{11}, s_{12}, s_{13}, s_{33}, s_{44}$) is studied numerically on both the radial and thickness mode resonances of a piezoelectric disc. It was found that the conventional $d_{33}$ material parameter which is measured to estimate performance of transducer at room temperature is not sufficient to predict resonance characteristics at high temperatures.

3.1 Introduction

Previous experimental studies have shown that design and fabrication of transducers that can simply survive in a reactor or other high temperature environment is challenging and that the signal strength reduces significantly for piezoelectric based ultrasonic transducers from room temperature to operating in liquid sodium at a hot stand-by temperature of ~260°C [Kažys et al. (2008) and Griffin et al. (2009)]. Studies that address aspects of high temperature transducer performance continue to be performed [Baptista et al. (2014), Searfass et al. (2016), Amini et al. (2016), Enciu et al. (2017)]. Recently, Searfass et al. (2016) and Amini et al. (2016) have quantified the ultrasonic signal strength of high temperature piezoelectric transducers for non-destructive evaluation (NDE) up to 800°C. Baptista et al. (2014) utilized the insights from measuring the electrical impedance of the piezoelectric sensor as a method for experimental quantification of the temperature effect. Lately, Enciu et al. (2017) and Haidar et al. (2017) also investigated the temperature
dependence of the piezoelectric material for the structural health monitoring (SHM) applications up to 250°C.

The resonance characteristics which includes signal strength of such ultrasonic transducers, determines the defect detection capability and mainly depends upon the piezoelectric effect. This effect is described by temperature dependent piezoelectric material coefficients [Randall et al. (1998)] which in turn depend upon extrinsic and intrinsic contributions from the ceramic itself [Haun et al. (1987)].

The temperature dependence of these various material coefficients of the different piezoelectric ceramics have been studied experimentally by many authors [Zhang et al. (2005), Sabat et al. (2007), Tang et al. (2015), Qaisar et al. (2017)]. Sabat et al. (2007) quantified the temperature dependence for modified Lead Zirconate Titanate ceramic (PZT-5A) as a function of temperature up to 195°C. Tang et al. (2015) reported the temperature dependence of a set of self-consistent full matrix material constants for PZT ceramics using a single sample for the resonance ultrasound spectroscopy method. This work also emphasized on the need for cost-efficient computer simulations to predict performance of high temperature electromechanical sensors. However, to address this need, it is necessary to describe the cause-effect relationships between the temperature dependent piezoelectric material coefficients and the resonance characteristics of an ultrasonic transducer.

It has been shown in previous experimental data [Sabat et al. (2007)] that each of the ten piezoelectric material parameters vary by different percentages at higher temperatures as compared to their baseline value at room temperature. Roy et al. (2014) modeled seven out of ten-independent piezoelectric material coefficients for temperature up 70°C to simulate the temperature effect on a resulting ultrasonic signal. However, the contribution of each material
coefficient to the ultrasonic signal was not quantified. Recently, Janapati et al. (2016) and Perez et al. (2016) performed numerical sensitivity studies to seek to understand the contribution of each coefficient to performance. This work involved varying selected material parameters by a fixed percentage. The limitation of such a sensitivity analysis approach is that a fixed percentage variation does not adequately capture the physical phenomenon in ceramics which is affecting each of the piezoelectric material parameters at high temperatures.

The effects of this physical phenomenon on the resonance frequencies of a piezoelectric disc need to be fundamentally quantified to enable adequate design and performance prediction for robust high temperature transducers for use in NDE and SHM. The primary objective of this study is thus to quantify the contribution of each piezoelectric material coefficient to the resonance modes of a disc as the temperature is increased. Hence, an experimental data-based methodology is presented by solving an axisymmetric problem in a finite element (FE) model. The changes to the thickness and radial modes in the resonance spectrum due to a temperature dependent material parameter are quantified by a sensitivity index. Lastly, the change in the resonance spectra due to a combined effect of varying all ten material parameters is compared with the experimental observations.

In the present study, section 3.2 describes the theory and relevant assumptions regarding piezoelectricity. Section 3.3 describes the methodology including the governing equations and discretization for the FE based model. Sections 3.4 and 3.5 present the simulation results and the discussion of the contribution for each material coefficient to the resonance modes. Section 3.6 presents conclusions based on the experimental data-based methodology.
3.2 Theory

The assumptions for the current work are described by the theory of piezoelectricity at a corresponding temperature. The fundamental equations can be obtained using Gibbs potentials, which is given by [Baptista et al. (2014)]

\[ S_i = s_{ij}^{E,H,\theta} T_j + d_{mi}^{H,\theta} E_m + d_{mi}^{H,\theta} H_m + \alpha_i^{E,H} d \theta \]  

(3.1)

\[ D_m = d_{mi}^{H,\theta} T_i + \varepsilon_{mk}^{T,H,\theta} E_k + m_{mk}^{T,\theta} H_k + p_m^{T,H} d \theta \]  

(3.2)

where \( S_i \) is the Cauchy’s total mechanical strain tensor, \( D \) is the electric displacement tensor, \( \varepsilon_{mk}^{T,H,\theta} \) is the absolute permittivity \( s_{ij}^{E,H,\theta} \) is the elastic compliance coefficient at at constant mechanical stress \( T \), electric field \( E \), magnetic field \( H \) and a corresponding temperature \( \theta \). The \( d_{mi} \) is the piezoelectric charge coefficient. The thermal expansion coefficient and pyroelectric constant are given by \( \alpha \) and \( p \) respectively. The magneto-dielectric coefficient is given by \( m \).

The phase velocity for elastic waves in a piezoelectric ceramic is significantly less than that for the electromagnetic waves. This implies a time derivative of magnetic field \( H \approx 0 \) indicating absence of magnetization effects and presence of a quasi-static field. Thus, magneto-dielectric coupling from equation (3.1) and (3.2) can be ignored at a corresponding high temperature \( \theta \). In the current work, the temperature difference \( d \theta \) is assumed to be small representing a gradual increase in the temperature of an ultrasonic transducer. Thus, for a quasi-thermal change in the piezoelectric material, the thermal expansion and pyroelectric effect can be ignored. This reduces equation (3.1) and (3.2) to a strain charge form of the piezoelectric effect at a corresponding temperature \( \theta \) which is given by

\[ S_i = s_{ij}^{E,\theta} T_j + d_{mi}^{\theta} E_m \]  

(3.3)

\[ D_m = d_{mi}^{\theta} T_i + \varepsilon_{mk}^{T,\theta} E_k \]  

(3.4)
Ferroelectric ceramics such as soft PZT-5A, which are widely used in the ultrasonic transducers, exhibit both intrinsic and extrinsic contributions to the piezoelectric effect [Haun et al. (1987), Randall et al. (1998)] shown in equation (3.3) and (3.4). This effect is dependent on the elastic, dielectric, and piezoelectric material coefficients [Haun et al. (1987)].

Equations (3.3-3.4) can also be represented in matrix form as

\[
\begin{bmatrix}
S1 \\
S2 \\
S3 \\
S4 \\
S5 \\
S6 \\
D1 \\
D2 \\
D3
\end{bmatrix}
= \begin{bmatrix}
E_1 & E_2 & E_3 \\
T_1 & T_2 & T_3 \\
T_4 & T_5 & T_6 \\
T_7 & T_8 & T_9 \\
T_{10} & T_{11} & T_{12} \\
T_{13} & T_{14} & T_{15} \\
T_{16} & T_{17} & T_{18} \\
T_{19} & T_{20} & T_{21} \\
T_{22} & T_{23} & T_{24}
\end{bmatrix}
\]

Due to the crystal symmetry and polarization of soft PZT-5A, the complete material matrix reduces to the five elastic, three piezoelectric and two dielectric coefficients as shown in equation (3.5). Hence, the piezoelectric effect described by equation (3.5) essentially represents the physical phenomenon that occurs in the PZT-5A as the temperature is increased.

Thus, the temperature dependent data [Sabat et al. (2007)] for these ten mutually independent coefficients from equation (3.5) describe the linear theory for the piezoelectric effect under the assumptions that—\(a)\) the piezoelectric material remains linearly elastic, and \(b)\) the applied electric field and mechanical stress are small, which thus implies that this theory is not applicable for large deformations of the piezoelectric material. This assumption is valid for the low-power piezoelectric based ultrasonic transducers which are used in linear ultrasonic measurements employed in SHM and NDE.
3.3 Method

To develop an understanding of sensitivity for each temperature dependent material coefficient from equation (3.5), a methodology is proposed as shown in Fig.3.1. A material model as described in equation (3.5) is formed using the temperature dependent experimental data for elastic compliance ($s_{ij}$), piezoelectric charge coefficient ($d_{mi}$) and dielectric permittivity ($\varepsilon_{mk}$). The material model forms the input parameters for a frequency domain finite element (FDFE) analysis. The FDFE approach is basically a pseudo-static problem that requires no time stepping [Chillara et al. (2016)]. The current FDFE model utilizes the temperature dependent experimental data for the soft PZT-5A material given by Sabat et al. (2007) in the morphotropic phase boundary (MPB) condition.

![Figure 3.1 Methodology for the sensitivity characterization](image)

The experimental data as a function of temperature for piezoelectric material coefficients is shown in Fig.3.2. Zhang and Yu (2011) reviewed material properties of high temperature piezoelectric materials. The variation in these piezoelectric material properties is dependent on the composition, dopants, grain size, internal defects and the magnitude of temperature increase from the room temperature [Sabat et al. (2007)]. The magnitude of each material coefficient at 195°C is compared with the percentage change from a baseline value at 15°C.
Each material coefficient varies by a different percentage when compared to the baseline value as shown in Fig.3.2.

<table>
<thead>
<tr>
<th>coefficient</th>
<th>(±)% change wrt baseline value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s_{33}^E$</td>
<td>(−)8.5</td>
</tr>
<tr>
<td>$s_{12}^E$</td>
<td>(−)18.5</td>
</tr>
<tr>
<td>$s_{13}^E$</td>
<td>(+)21.7</td>
</tr>
<tr>
<td>$s_{33}^E$</td>
<td>(−)22.9</td>
</tr>
<tr>
<td>$s_{44}^E$</td>
<td>(−)17</td>
</tr>
<tr>
<td>$d_{31}$</td>
<td>(−)14.7</td>
</tr>
<tr>
<td>$d_{33}$</td>
<td>(+)5.6</td>
</tr>
<tr>
<td>$d_{45}$</td>
<td>(−)6</td>
</tr>
<tr>
<td>$K_{11}$</td>
<td>(+)66.4</td>
</tr>
<tr>
<td>$K_{33}$</td>
<td>(+)94.1</td>
</tr>
</tbody>
</table>

Figure 3.2 Temperature dependent piezoelectric material coefficients [Sabat et al. (2007)]

In this work, for the case of soft PZT-5A, the dielectric constant $K_{33}$ in the poling direction demonstrates a maximum (94%) change as compared to the baseline value followed by $K_{11}$ (66%) as shown Fig.3.2. The weakening of the ionic movements in the piezoelectric crystal raises the ionic polarizability [Sabat et al. (2007)] which increase the magnitude of dielectric constants $K_{11}$, and $K_{33}$ with the temperature. The variation in the elastic coefficients of soft Pb [ZrxTi1-x] O3 (PZT-5A) with increasing temperature could be due to the first order phase transition in the crystal structure [Sabat et al. (2007)]. Moreover, the variation in piezoelectric charge coefficients ($d_{15}$, $d_{33}$, $d_{31}$) can be attributed to the increased domain wall motion with the temperature increase, which changes the activation energy required and hence, the extrinsic piezoelectric response [Sabat et al. (2007)].

In the current FDFE approach, electrical admittance [Baptista et al. (2014), Enciu et al. (2017), Haider et al. (2017)] is evaluated to quantify the temperature dependence of intrinsic and extrinsic contributions to the piezoelectric effect. The change in the admittance spectrum
due to a material parameter change is quantified by an index to understand the sensitivity of resonance characteristics to a single temperature dependent material coefficient.

A physics-based model is formulated in COMSOL™ [Comsol Multiphysics, version 5.2. COMSOL AB, Stockholm, Sweden (2016)] using a Lagrangian formulation. An axisymmetric problem with quadratic shape function is solved for the geometric configuration of a piezoelectric disc as shown in Fig.3.3. In the case of an axisymmetric problem, the elastic, piezoelectric and dielectric coefficients in the material matrix shown in equation (3.5) reduce to

\[
S^E = \begin{bmatrix}
 s_1^E & s_2^E & s_3^E & 0 \\
 s_2^E & s_1^E & s_3^E & 0 \\
 s_3^E & s_3^E & s_3^E & 0 \\
 0 & 0 & 0 & s_{44}^E
\end{bmatrix}
\] (3.6)

\[
d = \begin{bmatrix}
 0 & 0 & 0 & d_{15} \\
 d_{31} & d_{31} & d_{33} & 0
\end{bmatrix}, \quad \varepsilon^T = \begin{bmatrix}
 \varepsilon_{11}^T & 0 \\
 0 & \varepsilon_{33}^T
\end{bmatrix}
\] (3.7)

The mechanical strain \( S_i \) is given by

\[
S_1 = s_1^{E1}T_1 + s_2^{E2}T_2 + s_3^{E3}T_3 + d_{31}E_3
\] (3.8)

\[
S_2 = s_1^{E1}T_1 + s_2^{E2}T_2 + s_3^{E3}T_3 + d_{31}E_3
\] (3.9)

\[
S_3 = s_1^{E1}T_1 + s_2^{E2}T_2 + s_3^{E3}T_3 + d_{33}E_3
\] (3.10)

\[
S_4 = s_4^{E4}T_4 + d_{15}E_2
\] (3.11)

The electric displacements \( D_m \) from equation (3.4) and (3.5) reduce to

\[
D_2 = d_{15}T_4 + \varepsilon_{11}^T E_2
\] (3.12)

\[
D_3 = d_{31}T_1 + d_{31}T_2 + d_{33}T_3 + \varepsilon_{33}^T E_3
\] (3.13)
The baseline material parameters (at 15°C) for the current study are given in Table 3.1. The poling direction of the piezoelectric disc is aligned with the \( z \) direction of the model in the cylindrical coordinate system, as shown in Fig. 3.3. The FDFE formulation represents a time harmonic nature for the mechanical displacement \( u = u(r,z)e^{i\omega t} \) and stress \( \sigma = \sigma(r,z)e^{i\omega t} \) in the cylindrical coordinate space \((r,\phi,z)\). The \( e^{i\omega t} \) term describes the harmonic nature of the mechanical displacement \( u \).

**Figure 3.3** Geometric configuration of the problem

The equation for linear momentum balance in the frequency domain is thus given by

\[
\rho \omega^2 u + \nabla \sigma = F_v e^{i\phi}
\]

where \( \rho \) is the assigned material density, \( \omega \) is the angular frequency, and \( F_v \) is the body force. The boundaries \( z=0, z=0.9 \) mm and \( r=9.5 \) mm shown in Fig.3.3 are kept traction free implying \( \sigma_{31}=\sigma_{33}=\sigma_{11}=0 \).

Mechanical damping in the piezoelectric material is modelled using the Rayleigh damping model [Rupitsch et al. (2015)] which is given as

\[
[C_{uu}] = \alpha [M_{uu}] + \beta [K_{uu}]
\]

where \( C_{uu} \) is the damping matrix, \( M_{uu} \) is the mass matrix and \( K_{uu} \) is the stiffness matrix assembled at the global level in the FE model.
Table 3.1 Baseline (15°C) material parameters for PZT-5A

<table>
<thead>
<tr>
<th>Coefficient</th>
<th>Baseline value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s_{11}^E$</td>
<td>16.4e-12 (1/Pa)</td>
</tr>
<tr>
<td>$s_{12}^E$</td>
<td>-5.74e-12 (1/Pa)</td>
</tr>
<tr>
<td>$s_{13}^E$</td>
<td>-7.22e-12 (1/Pa)</td>
</tr>
<tr>
<td>$s_{33}^E$</td>
<td>18.8e-12 (1/Pa)</td>
</tr>
<tr>
<td>$s_{44}^E$</td>
<td>47.5e-12 (1/Pa)</td>
</tr>
<tr>
<td>$d_{31}$</td>
<td>-1.71e-12 (C/N)</td>
</tr>
<tr>
<td>$d_{33}$</td>
<td>3.74e-12 (C/N)</td>
</tr>
<tr>
<td>$d_{15}$</td>
<td>5.85e-12 (C/N)</td>
</tr>
<tr>
<td>$K_{11}$ (relative permittivity)</td>
<td>1730</td>
</tr>
<tr>
<td>$K_{33}$ (relative permittivity)</td>
<td>1700</td>
</tr>
<tr>
<td>Rayleigh damping coeff.($\alpha$)</td>
<td>7.3e4 (1/s)</td>
</tr>
<tr>
<td>Rayleigh damping coeff.($\beta$)</td>
<td>5.48e-9 (s)</td>
</tr>
<tr>
<td>Density ($\rho$)</td>
<td>7750 (kg/m$^3$)</td>
</tr>
</tbody>
</table>

The Rayleigh damping coefficient for mass and stiffness matrix are denoted by $\alpha$ and $\beta$ respectively. For the phenomenological modeling of attenuation due to damping, the Rayleigh damping coefficients are assumed to be constant [Rupitsch et al. (2015)]. Equation (3.14) thus becomes

$$-\rho \omega^2 u + i \omega \alpha_{dm} \rho u = \nabla \cdot (\sigma + i \omega \beta_{dk} c_E \cdot \varepsilon_{ei}) + F_v e^{i\phi}$$ (3.16)

For the piezoelectric media, a quasi-static field is assumed as stated in previous section. Thus, the electric field $E$ is related to the scalar electric potential $V$ as given by

$$E = -\nabla V$$ (3.17)

A unit magnitude voltage is applied to the boundary $z=0.9$ mm by using a terminal boundary condition assigned to PZT-5A as shown in Fig.3.3. The electric current in the piezoelectric domain $\partial^D \Omega_{piezo}$ is given by [Comsol Multiphysics documentation, (2016)]

$$\int_{\partial^D \Omega_{piezo}} D \cdot n = Q_0, \quad \frac{dQ_0}{dt} = I_{cir}$$ (3.18)

Electrical admittance is given by $Y= G+jB$ where $G$ is the real part of admittance known as conductance whereas $B$ is the imaginary part known as susceptance. In the present...
sensitivity study, the susceptance spectrum is plotted to quantify change in the resonance
spectrum due to each material coefficient. The element size for spatial discretization is
determined using the shear wave speed value $c$ (3895 m/s) in the piezoelectric domain at the
frequency $f_0$ (2.25MHz). The number of elements per wavelength $N$ is set equal to 10 by
performing the numerical convergence study. Maximum element size $h_{\text{max}}$ is thus given as
\[
   h_{\text{max}} = \frac{c}{f_0 N}
\]  
(3.19)

For this case, a quadrilateral element of size 0.02mm is used for mapped meshing technique of
the domain $\Omega_{\text{piezo}}$ shown in Fig.3.3. The sensitivity of the temperature dependent material
coefficient in terms of impact on the resonance spectrum is characterized using the metric
indices. One such metric is the root mean square deviation (RMSD) which is given by
\[
   \text{RMSD} = \sqrt{\frac{1}{n_{\text{freq}}} \sum_{n=\omega_I}^{n=\omega_F} \left[ Z_{E,B}(n) - Z_{E,T}(n) \right]^2 / Z_{E,B}^2(n)}
\]  
(3.20)

where $\omega_I$ is the start frequency and $\omega_F$ is the final frequency. $Z_{E,B}$ is the baseline electrical
admittance value at 15C while $Z_{E,T}$ is the admittance signature at the corresponding temperature
$T$ simulated at frequency $n$.

### 3.4 Results

A stationary, Multifrontal Massively Parallel Sparse (MUMPS) direct solver is used for
computation which is the default solver setting for frequency domain studies in COMSOL™.
The model is solved for 193,695 degrees of freedom. A frequency sweep is performed from
10kHz to 4.5 MHz.
3.4.1 Validation of Model with Reference Experimental Results

The baseline FE model is compared with the room temperature experimental data [Ren et al. (2017)] before performing the sensitivity study. Good agreement with the experimental and model data is obtained at room temperature for the resonant and anti-resonant frequencies as shown in Fig. 3.4. The percentage difference for the resonance frequencies $f_1$ and $f_2$ are less than 3% between the model and experimental data. The magnitude of the admittance corresponding to the resonant frequency from model data is also in good agreement with the experimental data shown in Fig. 3.4.

![Comparison of model data (blue line) and experimental data (black dashed line) for the electrical admittance of PZT-5A at room temperature.](image)

**Figure 3.4** Comparison of model data (blue line) and experimental data [Ren et al. (2017)] (black dashed line) for the electrical admittance of PZT-5A at room temperature.

3.4.2 Radial Resonance Modes

The magnitude of $K_{33}$ increases by 94% and $K_{11}$ increases by 66% as the temperature increases from 15 to 195°C [Sabat et al. (2007)]. This increase in $K_{33}$ increases the magnitude of susceptance as shown in Fig. 3.5(b). However, the temperature dependent variation in dielectric constant in the radial direction $K_{11}$ does not appear to affect the radial resonance mode as shown by the data given in Fig. 3.5(a).
Moreover, the temperature dependence of the piezoelectric charge coefficient $d_{33}$ and $d_{15}$ does not significantly affect the radial resonance modes as shown by the data given in Fig. 3.6(a-b). However, with variation in $d_{31}$ the radial resonance mode increases marginally as shown in Fig. 3.6(c) as the temperature is increased from 15 to 195°C.

Interestingly, the radial resonance mode is also unaffected by the temperature dependence of the elastic compliance coefficients $s_{33}$, $s_{13}$ and $s_{44}$ as shown in Fig. 3.7(a-c) which is discussed further in section 3.5. However, the temperature dependence of $s_{12}$ over the temperature range from 15 to 195°C causes a marginal increase in the radial resonance frequency as shown in Fig. 3.7(d). Similarly, a decrease in $s_{11}$ as a function of temperature increases radial resonant frequency from 15 to 195°C as shown in Fig. 3.7(e).
Figure 3.7 Effect on radial resonance mode due to temperature dependent variation in (a) $s_{33}$ (b) $s_{13}$ (c) $s_{44}$ (d) $s_{12}$ (e) $s_{11}$

These temperature dependent changes are further quantified using the RMSD based index as shown in Fig.3.8. It can be seen that the temperature dependence of $K_{33}$ (or $\varepsilon_{33}$) causes maximum sensitivity in terms of changes in the radial resonance mode followed by $s_{11}$, $s_{12}$ and $d_{31}$ at $195^{\circ}$C.

Figure 3.8 Sensitivity index for radial resonance mode due to temperature dependent variation in the ten material coefficients
3.4.3 Thickness Resonance Mode

The thickness resonance mode frequency reduces due to increase in $K_{33}$ as a function of temperature as shown in Fig.3.9(a). In a response which is similar to that for the radial mode frequency, the thickness mode frequency is also unaffected by the 66% variation in temperature dependent $K_{11}$ as shown in Fig.3.9(b).

![Graph showing effect on thickness resonance mode due to temperature dependent variation in $K_{33}$ and $K_{11}$](image)

**Figure 3.9** Effect on thickness resonance mode due to temperature dependent variation in (a) $K_{33}$ and (b) $K_{11}$

However, for a 5.6% increase in $d_{33}$, the magnitude of the susceptance increases with temperature along with a marginal change in the resonance frequency as shown in Fig. 3.10(a). The magnitude of the susceptance decreases for the variation in $d_{31}$ in the temperature range 15 to 195°C as shown in Fig.3.10(b). The thickness resonance mode is completely unaffected by temperature with changes in $d_{15}$ as shown in Fig. 3.10(c).

![Graph showing effect on thickness resonance mode due to temperature dependent variation in $d_{33}$, $d_{31}$, and $d_{15}$](image)

**Figure 3.10** Effect on thickness resonance mode due to temperature dependent variation (a) $d_{33}$ (b) $d_{31}$ and (c) $d_{15}$ coefficient
The reduction in the elastic compliance coefficient $s_{33}$ in the thickness direction increases thickness resonance frequency causing a shift to a higher frequency in the spectrum as shown in Fig. 3.11(a). This also implies that there is an increase in stiffness of the disc due to a reduction of $s_{33}$, as a function of temperature.

**Figure 3.11** Effect on thickness resonance mode due to temperature dependent variation in (a) $s_{33}$ (b) $s_{13}$ (c) $s_{44}$ (d) $s_{12}$ (e) $s_{11}$ coefficient
However, a variation in magnitude of $s_{13}$ from 15 to 105°C causes a reduction in the resonance frequency and an increase in the magnitude of susceptance as shown in Fig.3.11(b). The thickness mode remains unaffected by a 17% reduction in $s_{44}$ as shown in Fig.3.11(c). The temperature dependent variation in $s_{12}$ also increases the resonant frequency as shown in Fig.3.11(d). However, the magnitude of the susceptance decreases with an increase in the temperature. Similar characteristics are observed for the temperature dependence of $s_{11}$ as shown in Fig.3.11(e). The change in the resonance frequency as a function of temperature are further quantified using the RMSD based sensitivity index as shown in Fig.3.12.

![Figure 3.12](image)

**Figure 3.12** Sensitivity index for thickness resonance mode due to temperature dependent variation in the ten temperature dependent material coefficients

It can be seen from Fig.3.12 that $s_{33}$ exhibits the highest sensitivity followed by $K_{33}$, and $s_{11}$ and then followed by the remaining temperature dependent material coefficients.

3.4.4 Combined Effect of Temperature Dependent Material matrix

In previous sub-sections, analysis for the effect of temperature dependent variation in each piezoelectric material coefficient was performed. In physical piezoelectric based transducer, these piezoelectric material parameters vary simultaneously as a function of temperature. In this section, this case is modelled by considering the temperature dependent variation which
occurs in all material parameters simultaneously. The combined effect due to temperature dependent variation of all material coefficients causes an increase in the magnitude of the susceptance as shown in Fig.3.13(a-b). The combined effect due to the temperature dependent variation of all material coefficients causes an increase in the magnitude of the susceptance as shown in Fig.3.13(a-b). The observed shift in the resonance spectrum is due to the temperature effect on the piezoelectric material which is consistent with the previous experimental observations by Baptista et al. (2014) and Enciu et al. (2017). The increase in the magnitude of the susceptance (imaginary part of admittance) at the resonance frequency due to this combined effect is shown in Fig.3.13(b), and this is in agreement with the experimental observations reported by Haider et al. (2017).

**Figure 3.13** Combined temperature effect on (a) Radial resonance mode (b) Thickness resonance mode

This temperature dependent change in magnitude and the resonant frequency is quantified using the RMSD based index as shown in Fig.3.14. The RMSD based index increases from 320 at 105°C to 664 at 150°C and reaches up to 687 at 195°C indicating a non-linear change in the material coefficients of PZT-5A from 105 to 195°C.
Discussion

The sensitivity measured by the metric (RMSD) of each of the ten material coefficients for the temperature dependent variation [Sabat et al. (2007)] as explained in section 3.1 was reported in previous sub-section in Fig. 3.8 and 3.12. Based on the RMSD value, the thickness resonance mode is found to be more sensitive to the temperature change than the radial modes due to the polarization of PZT-5A in the thickness direction.

Here, Table 3.2 gives a summary of the sensitivity value for the percentage change in the material coefficient value at 195°C compared to the baseline value at 15°C.

Table 3.2 Summary of % change and RMSD value at 195°C

<table>
<thead>
<tr>
<th>coefficient</th>
<th>(±)% change as compared to baseline value</th>
<th>RMSD</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s_{11}^E$</td>
<td>(-)8.5</td>
<td>393</td>
</tr>
<tr>
<td>-$s_{12}^E$</td>
<td>(-)18.5</td>
<td>179</td>
</tr>
<tr>
<td>-$s_{13}^E$</td>
<td>(+)21.7</td>
<td>245</td>
</tr>
<tr>
<td>$s_{33}^E$</td>
<td>(-)22.9</td>
<td>874</td>
</tr>
<tr>
<td>$s_{44}^E$</td>
<td>(-)17</td>
<td>4</td>
</tr>
<tr>
<td>-$d_{31}$</td>
<td>(-)14.7</td>
<td>92</td>
</tr>
<tr>
<td>$d_{33}$</td>
<td>(+)5.6</td>
<td>74.1</td>
</tr>
<tr>
<td>$d_{15}$</td>
<td>(-)6</td>
<td>0.18</td>
</tr>
<tr>
<td>$K_{11}$</td>
<td>(+)66.4</td>
<td>0.23</td>
</tr>
<tr>
<td>$K_{33}$</td>
<td>(+)94.1</td>
<td>617</td>
</tr>
</tbody>
</table>

It is evident from Table 3.2 that the percentage change in the baseline value varies for different material coefficients. This indicates that each material coefficient exhibits different absolute
sensitivity to the temperature change. However, a higher percentage change from the baseline value does not ensure a higher influence over the resulting changes in the resonance spectrum as seen from Table 3.2. For example, the relative permittivity $K_{33}$ with a 94% increase, results in a RMSD value of 617, whereas a 66% increase in $K_{11}$ results in RMSD index of only 0.23. This counterintuitive phenomenon can be explained by contributions of a coefficient to the piezoelectric effect. This piezoelectric effect was explained for the axisymmetric problem in terms of mechanical strain and electric displacement given by equation (3.8) through (3.13) previously. The parameter $K_{33}$ contributes to the magnitude of the electric displacement $D_3$ whereas $K_{11}$ contributes to $D_2$. The coefficient $s_{33}^{E}$ contributes to the mechanical strain $S_3$. Similarly, $s_{11}^{E}, s_{12}^{E}$ contribute to $S_1$ and $S_2$ whereas $s_{13}^{E}$ contributes to $S_1, S_2$ and $S_3$ as described in equations (3.8), (3.9) and (3.10). These contributions of $s_{11}^{E}, s_{12}^{E}$ and $s_{13}^{E}$ to the mechanical strain in the principal directions can potentially explain a higher RMSD value for a lower percentage change in their baseline value as quantified in Table 3.2. For the disc shape geometry, $s_{44}^{E}$ does not drive a large change in the resonance spectrum of the PZT-5A. This is evident from a RMSD value of 4 although the baseline value of $s_{44}^{E}$ is reduced by 17% at 195°C. The piezoelectric charge coefficient $d_{15}$ has the lowest RMSD with a value of 0.18 followed by $K_{11}$ with a value of 0.23. Both of these coefficients contribute to electric displacement $D_2$. Mechanical strain $S_4$ and $D_2$ do not affect the magnitude of the susceptance spectrum for a soft PZT-5A disc. The $d_{33}$ coefficient exhibits an RMSD value of 74 for the 5.6% increase in the baseline value. It contributes to $D_3$ and $S_3$ as described in equation (3.10) and (3.13). The parameter $d_{31}$ which contributes to $D_3, S_2$, and $S_1$ results in a RMSD value of 92 for a 14.7% increase in the baseline absolute value.
The changes in the resonance spectrum when all ten material coefficients are varied using the temperature dependent experimental data [Sabat et al. (2007)] is reported in Fig.3.13(a-b) of Section 3.4. This combined temperature effect exhibits a reduction in the thickness resonance frequency as shown in Fig.3.13(b). This effect at 195°C results in an RMSD value of 687 as shown in Fig.3.14 and Table 3.2. The sensitivity metric is 8 times greater than the $d_{33}$ RMSD value of 74. As explained previously, this significant difference is due to the different sensitivity of the material coefficients to the temperature change and their contribution to the mechanical strain and electric displacement.

### 3.6 Conclusions

A 2D axisymmetric FE model has been developed to investigate the effect of temperature dependent material coefficients on the resonance modes of a piezoelectric disc. Initially, the model is validated with the literature experimental data for PZT-5A at room temperature. The FE model is then used to study thickness and radial mode resonance modes for temperature effects on elastic, piezoelectric and dielectric material coefficients using the reference experimental data. The temperature effect for all the material coefficients on the resonance spectrum is quantified using a root mean square deviation (RMSD) based sensitivity index.

It is demonstrated that the sensitivity of resonance modes due to the temperature effects on the material coefficients is dependent on the percentage change in the baseline value as well as the contribution of that coefficient to the mechanical strain and electric displacement. For a soft PZT-5A disc, $s_{33}^E$ demonstrate the highest sensitivity value followed by $K_{33}$, $s_{11}^E$, $s_{13}^E$, $s_{12}^E$, $d_{31}$, and $d_{33}$ in the order of high to low RMSD value. Changes in $d_{15}$, $K_{11}$ and $s_{44}^E$ exhibit minimal effect on the thickness resonance modes at 195°C. For radial resonance modes, the temperature dependence of $e_{33}$ showed highest sensitivity index followed by the $s_{11}$, $s_{12}$ and $d_{31}$ coefficients.
When all the ten material coefficients are varied based on the temperature dependent experimental data, the combined temperature effect results in the reduction of the thickness mode resonance frequency. This causes a reduction in the resonant frequency which is consistent with the observations from experimental work on the temperature effect for piezoelectric sensors. The combined effect results in a RMSD value of 687 which is 8 times the RMSD value for temperature dependent changes in $d_{33}$ at 195°C. This demonstrates that the magnitude of $d_{33}$ is not the sole factor that affects the resonance characteristics of the piezoelectric based ultrasonic transducers at high temperatures. It further appears that a complex interplay between material coefficients results in a reduction of thickness mode resonance frequency as the temperature is increased. This interplay was discussed in this work in terms of the contribution of each of the piezoelectric material coefficients to the mechanical strain and electric displacement. The simulation results also show that the current methodology, based on the temperature dependent experimental data has potential to be a useful tool to estimate the performance of high temperature piezoelectric based ultrasonic transducers. Particularly, for application in nuclear reactor, resonance analysis of radiation tolerant piezoelectric materials could also be performed using the current methodology for the temperature sensitivity characterization.
CHAPTER 4. ANALYSIS OF TEMPERATURE EFFECT ON THE EMBEDDED PIEZOELECTRIC DISC: A 3-LAYER PROBLEM

The previous chapter focused on the temperature dependence of piezoelectric material coefficients and their effect on the resonance frequency. However, piezoelectric material forms only one layer in the multi-layer ultrasonic transducer. Hence, interaction of these multiple layers at high temperature (HT) is important to study for understanding temperature effect on the resonance and sensitivity of transducer. In this chapter, a 3-layer problem is studied for interaction between high temperature piezoelectric material, adhesive and substrate by a numerical and an experimental study.

4.1 Introduction

In an ultrasonic transducer for harsh environment, all components of the transducer, including the piezoelectric material, are directly exposed to the high temperatures. Particularly, the piezoelectric material and adhesive selection is critical [Kazys (2008)] for the development of high temperature transducer.

In meeting the high temperature operation challenge, there has been extensive research which was reviewed in several papers [Patel (1990), Turner (1994), Kazys (2008), Zhang (2011), Lee (2014), Tarpara (2016)]. For temperatures greater than 500°C, aluminum nitride, lithium niobate, gallium orthophosphate and zinc oxide appear to be the validated and verified materials candidates for the piezoelectric element [Giurgiutiu (2010), Baba (2010) ,Cegla (2011), Hou (2013), Searfass (2016), Amini (2016), Dhutti (2016), Hou (2016)]. However, their piezoelectric properties including the electromechanically conversion efficiency, are considerably weaker than the materials used in room temperature ultrasonic transducers, such as those using lead zirconate titanate (PZT) [Bar-Cohen et al. (2017)]. In particular, for measurements at hot stand-by conditions (~250°C) for a SFR, a piezoelectric material with
efficient electro-mechanical conversion properties, and a high Curie temperature, is needed to improve the signal to noise ratio.

Another challenge is the temperature dependence of the bonding agents used in transducer assembly as shown in Fig. 4.1(a). It has been reported that 60% of HT ultrasonic transducers have interface adhesion problems [Kazys et al. (2008)]. For a continuous inspection process, the long-term durability of the transducer is needed for operation under both steady-state conditions at high temperature, and after repeated temperature cycling [Amini et al. (2016)]. Moreover, the ultrasonic transducer for under sodium viewing are estimated to undergo thermal cycling after removal of transducer from the immersed state in the hot liquid sodium.

In the current work, numerical sensitivity analysis is initially performed for temperature effect on adhesive and the reflected ultrasonic signal. In the experimental study, temperature dependence of the piezoelectric material is studied first. Subsequently, the combined temperature effect on the 3-layer system as shown in Fig 4.1(b), is studied up to 260°C which is also the hot-stand by temperature for sodium fast reactors. Lastly, repeated thermal cycling effect on the 3-layers is studied to demonstrate effect of heating and cooling cycle on the transient signal and the resonance spectrum.

![Diagram](image)

**Figure 4.1** Schematic of 3-layer problem to understand interface issue in the HT transducer
4.2 Theory: Transmission Phenomena for Normal Incidence

Under the plane wave assumption, consider a wave reflected normally from the interface between two media of different acoustic impedances $Z_{01}$ and $Z_{02}$ and the geometry shown in Fig. 4.2 [Kino (1970)].

Under the plane wave assumption, consider a wave reflected normally from the interface between two media of different acoustic impedances $Z_{01}$ and $Z_{02}$ and the geometry shown in Fig. 4.2 [Kino (1970)].

At an interface shown in Fig. 4.2, the mechanical stress $T$ and velocity $V$ will be continuous. Thus $T_1(z)$ and $V_1(z)$ on the left side of the interface at $z=0$ can be written as

$$T_1 = T_{F1}e^{-j\beta_1 z} + T_{B1}e^{j\beta_1 z}$$

$$V_1 = V_{F1}e^{-j\beta_1 z} + V_{B1}e^{j\beta_1 z}$$

(4.1)

where $\beta_1(=\omega/c_p)$ is the propagation constant of medium 1, $T_{F1}$, $T_{B1}$ are the amplitudes of forward and backward travelling wave on the left side of $z=0$.

Thus, the reflection ratio at $z=0$ is given by

$$R = \frac{T_{B1}}{T_{F1}}$$

(4.2)

From equation (4.1) and (4.2), it can be written as

$$T_1 = T_{F1}(e^{-j\beta_1 z} + Re^{j\beta_1 z})$$

$$V_1 = -\frac{T_{F1}}{Z_{01}}(e^{-j\beta_1 z} - e^{j\beta_1 z})$$

(4.3)
Similarly, in the region to the right of the interface, there is only a transmitted wave propagating in the forward direction which can be written as

\[ T_1 = T_2 e^{-j\beta_1 z} \]
\[ V_1 = -\frac{T_2}{Z_{02}} e^{-j\beta_1 z} \]  

(4.4)

The boundary condition for equation (4.3) and (4.4) at the plane \( z=0 \) is that the stress \( T \) and velocity \( V \) must be continuous. In this way the reflection ratio \( R \) and transmission ratio \( T_R \) can then be written as

\[ R = \frac{Z_{02} - Z_{01}}{Z_{02} + Z_{01}} \]
\[ T_R = \frac{2Z_{02}}{Z_{02} + Z_{01}} \]  

(4.5)

Thus, the equivalent input impedance of layer \( Z_{01} \) when it is in contact with the \( Z_{02} \), can be given as below:

\[ Z_{in} = Z_{01} \frac{e^{i\beta_1 l} + Re^{-i\beta_1 l}}{e^{i\beta_1 l} - Re^{-i\beta_1 l}} \]  

(4.6)

where \( l \) is the thickness of layer \( Z_{01} \). Equation (4.6) can further be modified as

\[ Z_{in} = Z_{01} \frac{(Z_{02} \cos \beta_1 l + jZ_{01} \sin \beta_1 l)}{(Z_{01} \cos \beta_1 l + jZ_{02} \sin \beta_1 l)} \]  

(4.7)

Equation (4.7) demonstrates that the equivalent impedance of layer 1 when it is in with contact 2 is a function of propagation constant \( \beta \) of each layer, thickness of layer 1 and acoustic impedance \( Z_{01} \) and \( Z_{02} \). Equation (4.7) was derived for transmission-reflection phenomenon for two layers in contact with each other. However, in the multi-layer ultrasonic transducer, the equivalent acoustic impedance of layer would be function of thickness of that layer and function of propagation constants, acoustic impedance of multiple layers as shown Fig.4.1(a). Adhesive layer between piezoelectric and faceplate shown in Fig. 4.1(a) is critical to study as
the generated mechanical wave is transmitted via adhesive to the faceplate and the fluid medium for immersed transducer, specifically for under-sodium viewing. Hence, in this work, we study 3-layers (piezoelectric+ adhesive+ substrate) as shown in Fig. 4.1(b) to understand temperature effect on the critical interfaces in the USV transducers.

### 4.3 Piezoelectric Material Selection

Design and material selection of a high temperature ultrasonic transducer has conventionally only considered $d_{33}$ and the Curie temperature ($T_c$) of piezoelectric material as a maximum design temperature for operation [Kazys et al. (2008)]. However, dielectric characterization of the perovskite structured piezoelectric material indicates that it becomes highly capacitive well before Curie temperature, which is accompanied by a reduction in the ultrasonic signal strength [Bilgunde et al. (2015b)] after a particular transition temperature ($T_p<T_c$) is reached. This implies the need for a transition temperature $T_p$ to be greater than 260°C for optimum performance of the material in the current targeted application of inspection in liquid sodium at hot stand-by temperature (~250°C) for SFRs.

Bismuth Scandium Oxide-Lead titanate, BS-PT [(1-x) BiScO$_3$-xPbTiO$_3$] at its morphotropic phase boundary (MPB) (x=0.64) is a promising candidate for high temperature applications because of its relatively high piezoelectric response as compared to Lithium niobate and aluminum nitride [Eitel et al. (2001,2003), Zhang et al. (2005), Chen et al. (2012,2014), Li et al. (2014)]. Gotmare et al. (2008,2010) performed a systematic study which demonstrated a constant value of relative permittivity indicating minimal thermal degradation of BS-PT at 250°C for 100 days. The dielectric constant characterization of BS-PT (TRS BT200) as a function of temperature is shown in Fig.4.3 demonstrates a transition temperature ($T_p$) higher than 300°C.
In investigating properties of the BS-PT, the high Curie temperature ($T_c > 430^\circ C$) shown in Fig.4.3 could be due to the small perovskite tolerance factor of BiScO$_3$ in the solid-solution system with PbTiO$_3$ [Eitel et al. (2003)].

### 4.4 Adhesive Selection

The adhesive used between piezoelectric element and substrate is required to withstand the temperature encountered at the hot stand-by for continuous inspection. In this study, a two-part epoxy (Epotek 353ND) is used for this purpose which is estimated to have continuous operating temperature of 350$^\circ C$. The physical and electrical properties of the epoxy are listed in Table 4.1 as given below:

#### Table 4.1 Physical and Electrical properties of high temperature epoxy Epotek 353ND

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Continuous operating temperature</td>
<td>-55 to 250$^\circ C$</td>
</tr>
<tr>
<td>Intermittent operating temperature</td>
<td>-55 to 350$^\circ C$</td>
</tr>
<tr>
<td>Storage modulus (at 23°C)</td>
<td>3.56 GPa</td>
</tr>
<tr>
<td>Glass transition temperature ($T_g$)</td>
<td>$&gt;90^\circ C$</td>
</tr>
<tr>
<td>Coefficient of thermal expansion (CTE)</td>
<td>Below $T_g$: 54e-6 in/in $^\circ C$</td>
</tr>
<tr>
<td></td>
<td>Above $T_g$: 206e-6 in/in $^\circ C$</td>
</tr>
<tr>
<td>Dielectric constant (1kHz)</td>
<td>3.17</td>
</tr>
<tr>
<td>Degradation temperature</td>
<td>412$^\circ C$</td>
</tr>
</tbody>
</table>
However, as the temperature of epoxy increases above glass transition temperature \( (T_g) \), the covalent bonds in the epoxy potentially weaken which reduces the storage modulus of the epoxy [Chen et al. (2013)]. This phenomenon is shown schematically in Fig.4.4. Above \( T_g \), the epoxy changes from glassy to a rubbery composition accompanied by a non-linear decrease in the storage modulus [Chen et al. (2013)].

![Graph showing the temperature dependence of storage modulus of polymer-based epoxy](image)

**Figure 4.4** Temperature dependence of storage modulus of polymer-based epoxy

Such a reduction in the storage modulus \( E \) will also reduce the plane wave velocity \( c_p \) in the adhesive, which is given by

\[
c_p = \sqrt{\frac{E(1-\nu)}{\rho(1+\nu)(1-2\nu)}}
\]  
(4.8)

\[
Z_{adh} = \rho c_p = \sqrt{\frac{\rho E(1-\nu)}{(1+\nu)(1-2\nu)}}
\]  
(4.9)

This reduction in the plane wave velocity can also affect the acoustical impedance which is evident from equation (4.9). Using equation (4.9) and (4.7), the equivalent impedance \( Z_{in(adh)} \) of the epoxy based adhesive and the substrate is given by

\[
Z_{in(adh)} = Z_{adh} \frac{(Z_{sub} \cos \beta_{adh}l + jZ_{sub} \sin \beta_{adh}l)}{(Z_{sub} \cos \beta_{adh}l + jZ_{sub} \sin \beta_{adh}l)}
\]  
(4.10)
where \( Z_{sub} \) and \( Z_{adh} \) is the acoustic impedance of substrate and adhesive respectively, \( \beta_{adh} \) is the propagation constant for adhesive, \( l \) is the effective thickness of the adhesive layer. It can be seen that, at elevated temperatures, the equivalent input acoustic impedance \( Z_{in(adh)} \) is dependent on the acoustic impedance of adhesive and substrate at the corresponding temperature, the propagation constants and the effective thickness of the adhesive. Moreover, using equation (4.5) and (4.10), the 3-layer system shown in Fig. 4.1(b) can be reduced to 2-layer system for which transmission and reflection ratio are given by

\[
T = \frac{2 \times Z_{in(adh)}}{Z_{in(adh)} + Z_{pz}}
\]

(4.11)

\[
R = \frac{Z_{in(adh)} - Z_{pz}}{Z_{in(adh)} + Z_{pz}}
\]

(4.12)

where \( T \) is the transmission ratio, \( R \) is the reflection ratio and \( Z_{pz} \) is the acoustic impedance of the piezoelectric material at corresponding temperature.

4.5 Finite Element Model

![Axial symmetry](image)

**Figure 4.5** Geometric configuration of an axisymmetric FE model in Comsol™

To understand the effect of reduction in storage modulus of epoxy on the reflected backwall echo amplitude from the substrate, an axisymmetric coupled problem is solved as shown in Fig. 4.5 which also shows the geometric dimension of the problem which are similar to the
experimental set-up discussed later in the chapter. The material parameters used to study the 3-layer problem numerically are listed in Table 4.2, 4.3 and 4.4.

**Table 4.2 Material parameters for pristine state of epoxy bond**

<table>
<thead>
<tr>
<th>State</th>
<th>$c_p$ (m/s)</th>
<th>$C_s$ (m/s)</th>
<th>$\rho$ (kg/m$^3$)</th>
<th>$Z_{adh}$ (MRayl)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pristine</td>
<td>2600</td>
<td>1300</td>
<td>1200</td>
<td>3.12</td>
</tr>
</tbody>
</table>

**Table 4.3 Material properties for steel domain**

<table>
<thead>
<tr>
<th>Material</th>
<th>$E$ (GPa)</th>
<th>Poisson ratio</th>
<th>$\rho$ (kg/m$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Steel</td>
<td>200</td>
<td>0.29</td>
<td>7870</td>
</tr>
</tbody>
</table>

**Table 4.4 Material parameters for BS-PT**

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Baseline values</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s_{11}^E$</td>
<td>$1.2 \times 10^{-12}$ (1/Pa)</td>
</tr>
<tr>
<td>$s_{12}^E$</td>
<td>$-3.70 \times 10^{-12}$ (1/Pa)</td>
</tr>
<tr>
<td>$s_{13}^E$</td>
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<tr>
<td>$s_{33}^E$</td>
<td>$1.48 \times 10^{-12}$ (1/Pa)</td>
</tr>
<tr>
<td>$s_{44}^E$</td>
<td>$3.2 \times 10^{-12}$ (1/Pa)</td>
</tr>
<tr>
<td>$d_{31}$</td>
<td>$-1.03 \times 10^{-10}$ (C/N)</td>
</tr>
<tr>
<td>$d_{33}$</td>
<td>$2.55 \times 10^{-10}$ (C/N)</td>
</tr>
<tr>
<td>$d_{15}$</td>
<td>$3.40 \times 10^{-10}$ (C/N)</td>
</tr>
<tr>
<td>$K_{11}$ (relative permittivity)</td>
<td>1110</td>
</tr>
<tr>
<td>$K_{33}$</td>
<td>1250</td>
</tr>
<tr>
<td>Rayleigh damping coeff.((\alpha))</td>
<td>$7.3 \times 10^{-4}$ (1/s)</td>
</tr>
<tr>
<td>Rayleigh damping coeff.((\beta))</td>
<td>$5.48 \times 10^{-9}$ (s)</td>
</tr>
<tr>
<td>Density ($\rho$)</td>
<td>7600 (kg/m$^3$)</td>
</tr>
</tbody>
</table>

The equation of linear momentum balance in the time domain for the FE model is given by (Rose, 1994)

$$\rho \frac{\partial^2 u}{\partial t^2} = \nabla s + F_v$$  \hspace{1cm} (4.13)

where $\rho$ is the assigned material density, $u$ is the mechanical displacement, $s$ is second order Piola-Kirchoff stress tensor, and $F_v$ is the body force. A low reflection boundary condition is applied to the sidewall boundary of steel domain which is given by

$$\sigma \cdot n = -pC_p \left( \frac{\partial u}{\partial t} n \right) - pC_s \left( \frac{\partial u}{\partial t} t \right) t$$  \hspace{1cm} (4.14)

Application of such low reflection boundary helps to reduce the computational size of the model.
In the experimental measurements, the piezoelectric transducer is typically connected to the pulsar-receiver circuit using BNC cables and lead wires. This introduces an electrical impedance mismatch between the piezoelectric transducer and the pulse-receiver instrumentation. This is modelled in a finite element model by introducing a resistor with impedance equal to that of pulsar-receiver. In the current study, the impedance value of resistor is set to an ideal 50 Ω. The electric circuit module in Comsol™ is used to evaluates global variables, voltage, and current as a function of time. In the model, a negative spike signal of 160V is applied to the piezoelectric material on the back-electrode boundary. The current at the back-electrode of the piezoelectric element is given by [COMSOL documentation, (2016)] is given by equation (3.18) in the previous chapter. The voltage on the electrode surface is given by

\[ V_{pe}(t) = V_{source}(t) - I_{ei}R \]  \hspace{1cm} (4.15)

Quadrilateral and triangular elements with quadrilateral shape function are used for meshing of the FE model. The maximum element size for a domain is given by equation (3.19) in the previous chapter. Mapped meshing technique is used for the spatial discretization of the piezoelectric and adhesive domains in the model whereas free-triangular mesh was used for steel domain due to relatively larger thickness. The details of the spatial discretization are further given by Table 4.5.

<table>
<thead>
<tr>
<th>Geometric domain</th>
<th>Element size</th>
</tr>
</thead>
<tbody>
<tr>
<td>Piezoelectric material</td>
<td>0.05mm (quadrilateral)</td>
</tr>
<tr>
<td>adhesive</td>
<td>0.01mm (quadrilateral)</td>
</tr>
<tr>
<td>substrate</td>
<td>0.5mm (triangular)</td>
</tr>
</tbody>
</table>
The temporal discretization (time step) $t$ for the model is given by

$$t = \frac{CFL}{f_0 N}$$  \hspace{1cm} (4.16)

where CFL (Courant-Friedrich-Levi) number [Courant et al. (1928)] is set equal to 0.2 from the numerical convergence study [Bilgunde et al. (2015a)]. The transient analysis is run from 0 to 17μs using generalized-alpha solving method which is by-default solver in COMSOL time domain simulations.

Figure 4.6(a) shows effect of reducing the plane wave velocity in the epoxy by 65% when compared to the baseline value listed in Table 4.2. The model results demonstrate a significant 22dB reduction in the backwall echoes from the steel substrate as shown in Fig. 4.6(a). Moreover, Fig.4.6(b) shows Fast Fourier transform of first backwall echo of the model results. A reduction of 0.1MHz is observed in the spectral content in addition to the magnitude reduction due to 65% decrease in the plane wave velocity of epoxy adhesive.

**Figure 4.6** Model results for temperature effect on adhesive (a) Time domain signal (b) Fast Fourier transform of first backwall echo in the model data

Model data demonstrated the effect of reduction in storage modulus of epoxy on the ultrasonic signal and the resonance frequency due to increase in temperature. This further underlines the critical role of adhesive that was also discussed theoretically in the previous section.
4.6 High Temperature Measurements for A 3-Layer Set-Up

The schematic of the experimental set-up for the high temperature pulse-echo solid coupled contact measurements is shown in Fig. 4.7(a-b). The thickness of the BS-PT material is 0.9mm and diameter is 19mm (0.75 inch). The critical challenge in the experimental system is the adhesive bonding between the BS-PT and the low carbon steel specimen. This adhesive bond using Epotek 353ND is cured at 150°C for one hour in the oven.

Figure 4.7 High temperature experimental set-up (a) Schematics (b) Inside the furnace

A 0.2mm (0.008-inch) diameter nickel lead wire is connected to the back electrode of the BS-PT using a silver based conductive two component epoxy paste (Duralco 124). The bond is cured in an oven at 120°C for two hours with post cure heating for an additional two hours at the same temperature, to improve temperature stability of mechanical properties. The front electrode connection is established by using the low-carbon steel substrate itself as shown in
Fig.4.7(a-b). Pyroduct-597A connects the nickel wire to a 12.7 mm (0.5-inch) thick, cold drawn low-carbon steel substrate. The bond is cured at 120°C for two hours. The steel block bonded to BS-PT element is placed in a furnace and a high temperature alumina ceramic tube is used as electrical insulation for the nickel lead wires as shown in Fig.4.7(b). The transmitted and received ultrasonic pulse data acquired from the transducer is dependent upon the energy, damping gain and filter settings of the pulse-receiver (P-R). In the current experimental set-up, a commercial P-R (Panametrics 5052x) is used to excite the transducer with a spike of 100V (energy setting). The damping resistance ($R_d$) is adjusted to 50Ω. Gain is set to be 16dB and the high pass filter is adjusted to the out setting with the repetition rate set at 4. The pulse-echo data from the transducer is acquired using a digital oscilloscope (LeCroy HDO 4034) with a 12-bit vertical resolution. The data is sampled at 450MHz and averaged 512 times to filter random electrical interference noise due to the interaction of heating coil with the exposed surface of nickel wire in the furnace. The high temperature in-situ contact measurements are performed in an electric furnace (Barnstead Thermolyne furnace type 47900). The maximum temperature set point of the furnace was defined as 260°C. The temperature was increased from 20 to 260°C at a 5°C/min rate. The hold (dwell) time at 260°C was set to be 20 minutes in order to achieve a uniform temperature throughout the bonded assembly. It should be noted that the temperature refers to the value indicated by the furnace controller. As the experiment proceeded, the P-R settings were not modified during the contact ultrasonic measurements. The averaged waveform is saved and recorded in the oscilloscope memory in the comma separated values (CSV) file format for every 10°C increase in the furnace temperature and subsequently transferred to a PC for further analysis.
4.6.1 Thermal Cycle-1

The averaged ultrasonic pulse-echo backwall RF signal as a function of furnace temperature from 20°C up to 260°C is shown at a series of temperatures in Fig.4.8(a). The amplitude of the backwall echo reduces linearly up to 150°C and is followed by a non-linear decrease up to 260°C as shown in Fig.4.8(b).

![Figure 4.8](image)

**Figure 4.8** Thermal cycle-1 a) Backwall echoes during heating from 20 to 260°C  b) Peak amplitude reduction as a function of temperature for first backwall echo.

An expanded view of first front wall echo signal as a function of temperature is show in Fig.4.9(a). The increase in time of arrival of the first front wall echo which is due to a reduction of the longitudinal velocity of the wave propagating in the wave path media (steel, epoxy and BSPT) can also be seen in Fig.4.9(a). Spectra obtained by application of an FFT to the first front wall echo are as shown in Fig.4.9(b). The disc used has a nominal thickness mode resonance at 2.4MHz and the spectral data show the fundamental and the third harmonic.

A reduction in the fundamental resonance frequency (2.4 MHz) by 0.1 MHz is seen at 260°C when compared with the resonance frequency of the structure at 20°C and this is shown in the data given in Fig.4.9(c). Similarly, a 0.1 MHz shift to a lower frequency is also observed for the third harmonic resonance frequency of the structure as shown in Fig.4.9(d).
Figure 4.9 (a) Expanded view of first backwall echo (b) Frequency spectrum for first backwall echo as a function of temperature (c) Explored view of fundamental resonance (d) Expanded view of third harmonic indicating resonance frequency shift as a function of temperature

4.6.2 Analysis of Interface Condition Post-Thermal Cycle-1 Measurements

To further understand the interface condition after thermal cycle-1, a high frequency ultrasonic C-scan was obtained using a 12.7mm (0.5-inch) nominal diameter, 10MHz focused transducer with a 50.8mm (2-inch) focal length [GE Panametrics, Type V311] as shown in Fig.4.10(a). The transducer was excited with a square wave pulsar-receiver [Panametrics 5077PR] with 100V pulsar voltage and 18dB receiver gain. The data acquisition gate was set to capture the first backwall signal and to record the peak amplitude for each spatial location. These data are used to define the value for each pixel of the C-scan. The resulting C-scan of
the BS-PT-steel interface is shown in Fig.4.10(b) which shows no obvious de-bonding in the interface after first thermal cycle. The disc is radially constrained by the epoxy extruded during the curing process of BSPT-steel bond. The ultrasonic response of this extruded region can also be seen as the light blue region in the C-scan of interface shown in Fig.4.10(b).

Furthermore, the bonded assembly is again connected to the experimental set up shown in Fig.4.7 for pulse echo measurements post thermal cycle-1. It was found that there was 20dB loss in the backwall echo amplitude compared to the signal before cycle-1 as shown in Fig.4.11. Interestingly, similar loss in dB was observed in the simulation results in section 4.5 which indicates a change in bond properties that might have occurred during thermal cycle-1.

![Diagram](image1)

**Figure 4.10** Anaysis of interface condition post thermal cycle-1 (a) Experimental set-up (b) photograph of item after thermal cycle-1

![Diagram](image2)

**Figure 4.11** Comparison of pulse-echo response after cycle-1 at room temperature with baseline signal
4.6.3 Effect of Repeated Temperature Cycling on the Echo Amplitude

The experimental measurements were repeated to investigate the effect of repeated thermal cycling on the 3 layers set-up. The RF signals for heating cycle-2 as a series of temperatures from 20 to 260°C is shown in Fig.4.12(a). The furnace was set for a dwell time of 20 minutes after the end of heating part of the cycle.

![Figure 4.12](image)

**Figure 4.12** Comparison of time domain signals for (a) heating cycle-1 and 2 at 20°C (b) heating cycle 2 from 20 to 260°C (c) Cooling cycle from 260 to 150°C showing exponential decay (d) Re-appearance of back-wall echoes below 120°C

The experimental arrangement allowed the transducer components to cool at its natural cooling rate in the fume hood and samples returned to room temperature. The ultrasonic signal at 260°C shown in Fig. 4.12(b) was acquired at the end of the dwell time. It can be seen that the RF signal exhibits a characteristic decay. Furthermore, during cooling of the furnace from
260 to 150°C the ultrasonic signal exhibited similar characteristic behavior in the ring-down as shown in Fig.4.12(b). The back-wall echoes begin to re-appear in the RF signal when the furnace temperature dropped below 120°C, as shown in Fig.4.12(c). Furthermore, the ultrasonic waveform at the end of cooling cycle-2 appears to recover the previous form considerably, when compared with the original waveform at the beginning of heating cycle-2, as shown in Fig.4.12(d).

To investigate further for the characteristic ring down, a fast Fourier transform (FFT) of the time domain experimental data was obtained for a windowed signal of same sample length and this gives the spectra as shown in Fig.4.13(a-b). It can be seen that the fundamental and third harmonic frequency response at the beginning of the heating cycle and at the end of cooling cycle are considerably similar. The experimental system cools down to the near-original state to give the ultrasonic signal which was also shown previously in Fig.4.12(d).

![Image](image.png)

**Figure 4.13** Frequency response as a function of temperature in the cooling cycle for (a) Fundamental resonance (b) Third harmonic resonance

During the cooling cycle, the spectral analysis of the experimental data from the thermal cycle revealed that the 6dB bandwidth of fundamental resonance reduced from 0.55 MHz at 20°C to 0.16MHz in the temperature range of 260 to 150°C during cooling of the furnace as shown in Fig. 4.13(a). It should be noted that the bandwidth again increased to 0.5MHz when the furnace
eventually cooled down to 20°C. A similar and consistent series of observations for a 3\textsuperscript{rd} thermal cycle is shown in Fig.4.14(a-b).

![Graphs showing temperature vs. time and frequency response](image)

**Figure 4.14** Element bonded to steel block: (a) RF signal during cooling cycle-3 (b) Bakwall echoes, which re-appear below furnace temperature of 100°C (c) Fundamental resonance (d) Third harmonic resonance

The increase in the bandwidth and magnitude of the frequency response as the furnace temperature reduces from 260 to 20°C can similarly be seen in the fundamental and third harmonic frequency response of the structure as shown in Figs.4.14(c-d). These data are also consistent with the results seen in the previous thermal cycles. For cooling cycles-4 and 5, the RF signal for the temperature dependent phenomenon in the temperature range of 260 to 150°C
is shown in Fig.4.15(a-b). These measurements also exhibit a reduction of the bandwidth and magnitude of the frequency response as demonstrated in previous thermal cycles.

![Figure 4.15 RF signal in the temperature 260 to 140°C during (a) Cooling cycle-4 (b) Cooling cycle-5](image)

The back-wall echoes re-appear in the RF data below a furnace temperature of 100°C as shown in Fig.4.16(a-b) for cooling cycle 4 and 5 respectively.

![Figure 4.16 Backwall echo for element on steel block as the furnace temperature reduces during (a) cooling cycle 4 and (b) cooling cycle 5](image)

A summary of the second back-wall echo amplitude data for five heating and cooling cycles is shown in Figs.4.17(a-b) respectively. The 20dB reduction in the peak amplitude of echo between the first heating and subsequent heating cycles, shown in Fig.4.17(a), indicates
a permanent change in the bond properties after the first heating cycle that was not repeated for subsequent heating cycles.

Figure 4.17 Summary of second back wall echo amplitude (a) during heating cycles and (b) during cooling cycles

Figure 4.17(b) summarizes the persistent observation of the effect on back-wall echo amplitude in the temperature range 260 to 150°C during cooling of the furnace. This response is followed by an increase in the amplitude due to the re-appearance of the back-wall echo signals below 150°C. The spectral analysis of this data also demonstrated that the performance during cooling of the experimental set-up is accompanied by a reduction in the bandwidth in the temperature range of 260 to 150°C. The reduction in the bandwidth can be related to the mechanical quality factor $Q$ given as where $\Delta \omega$ is the -6dB bandwidth and $\omega_r$ is the resonance frequency. In the current experiments, the data seen during cooling in the 260 to 150°C temperature range, indicate there is a reduction in the bandwidth $\Delta \omega$ which corresponds to an increase in the $Q$ factor for a constant resonance frequency $\omega_r$. A higher $Q$ factor qualitatively implies an under-damped oscillatory system from the theory of linear time invariant systems. This indicates further that during cooling cycle, in the temperature range of 260 to 150°C, the ultrasonic system changes to an under-damped system as compared to the system at 20°C.
4.7 Conclusion

Degradation of interface between piezoelectric material and faceplate is a critical issue for high temperature ultrasonic transducer for under sodium viewing. In this work, a 3-layer problem was set-up to analyze temperature effect using numerical and experimental study.

The numerical study demonstrated the effect of reduction in storage modulus of epoxy at temperatures above glass transition temperature. A 22dB reduction in the ultrasonic backwall echoes was observed for 65% reduction in the storage modulus of epoxy.

Subsequently, in-situ measurements were performed up to hot stand-by temperature (260°C) of SFRs which showed significant reduction in the backwall echo amplitude after 200°C accompanied by 0.1MHz reduction in the resonance frequency. A 20dB reduction in the backwall amplitude echo was observed after thermal cycle-1 when compared with original waveform. Interestingly, this 20dB loss in the signal was alignment with the simulation results. Also, the distribution of echo amplitudes in C-scan did not show any visible debonding which further indicated that interface properties might have degraded uniformly.

Moreover, effect of heating and cooling cycle on the ultrasonic signal was demonstrated for multiple thermal cycles up to hot stand-by temperature (260°C) of SFRs. Spectral analysis of the measurements during heating and particularly the cooling part of the cycle demonstrated changes in the bandwidth of frequency response, indicating an increase in the mechanical quality factor of the 3-layer system consisting of BS-PT bonded with the steel substrate. This increase in the mechanical quality further indicates an underdamped system in the temperature resulted in characteristic ring down of the ultrasound signal. In a nutshell, the current work demonstrated methodology based on numerical and experimental methods to analyze the temperature effect characteristics with regards to the long-standing interface degradation issues in the high temperature ultrasonic transducers for under sodium-viewing.
CHAPTER 5. MODELING AND SURROGATE EXPERIMENTS TO EVALUATE IMMERSION TRANSDUCERS FOR UNDER SODIUM VIEWING

The use of liquid sodium as a coolant in a NPP enables a low operating pressure and a high-power density. However, there are major challenges with regards to in-service inspection (ISI) and repair [Wang et al. (2012), Lubeigt et al. (2015)] of reactors. The opacity and electrical conductivity of a sodium coolant makes optical and electromagnetic techniques less effective for ISI of NPP components. During the operation of a sodium fast reactor various system components can be displaced. For example, the head of a Fuel Sub-Assembly (FSA) may undergo a lateral shift from its original position due to the fast neutron induced damage to its structural material. Visualization based on ultrasonic imaging has been found feasible to guide robotic arm motion for the inspection of components which was discussed in Chapter 2.

This technique of ultrasound-based scanning is called under sodium viewing (USV) in liquid sodium cooled fast reactors (SFRs). The major challenge in proving an effective USV capability has been the development of ultrasonic immersion transducers that can operate for an extended period at high temperature (HT), and are not damaged by at least gamma radiation in the reactor environment. The radiation effects on the sensitivity of piezoelectric transducer have been investigated in detail and reported in several studies [Holbert et al. (2005), Augereau et al. (2008), Parks et al. (2011), Rempe et al. (2014), Sinclair et al. (2015)].

For extended operation of the transducer at high temperatures, two approaches of design have been previously investigated: a) waveguide-based transducers and b) fully immersion transducers. A waveguide acts as a buffer rod and isolates the transducer piezoelectric element from the high temperature. Recently, a flexible or re-configurable waveguide concept has been developed for temperature sensing applications [Periyannan et al. (2016)]. The challenges with ultrasonic transducers using a waveguide are the limitations in deployment and also the
spurious echoes which occur due to the length and shape of the waveguide. These spurious echoes decrease the resolution of the resulting ultrasonic scan and hence, the defect detection ability which was discussed in detail in chapter 2.

The previous chapter focused on the analysis of temperature effect seeking to understand interfacial challenges in the high temperature transducer. In an immersion measurement, the thermo-physical properties of the fluid also change as a function of temperature, and such changes are in addition to the temperature dependence of the properties for the transducer element and the target specimen material (steel) studied in the previous chapter. The changes to the thermo-physical properties of a fluid affects the amplitude of the reflected ultrasonic signal which is utilized for high temperature ultrasonic imaging.

One of the major experimental challenges in the performance evaluation of HT immersion transducers for use in sodium fast reactors is safely working with liquid sodium for bench-top experiments. This is due to the chemical reactivity of liquid sodium which explodes upon contact with water or burns with air, increasing the risk of accidents [Mikami et al. (1996)]. For this safety reason, surrogate fluids are utilized in this work for the evaluation of the temperature dependence of the immersion transducer. The sensitivity of the ultrasonic transduction in the transducer is subsequently measured in the fluids with different thermo-physical properties as a function of temperature for each thermal cycle.

5.1 Benchmarking of Ultrasonic Transducers

One of the original PNNL single element ultrasonic transducers [Griffin et al. (2011)] used in HT immersion testing in liquid sodium was provided. Basic pulse-echo measurements are taken at room temperature in water with PNNL high temperature transducer and a commercial (Panametrics V306) transducer as shown in Fig.5.1(a-b) respectively. The purpose was to compare the HT PNNL transducer (nominal dia. 25mm) with the commercial V306 transducer
(nominal dia. 12.5mm) of the same center frequency with the same experiment set-up at the room temperature (20°C). The damping, energy and gain settings of the Panametrics pulsar-receiver 5052 are same for both the experiment using the same BNC cable. This also allowed to eliminate system characteristics (pulsar, cable, and oscilloscope) effect between performance of the two transducers.

Figure 5.1 (a) PNNL research high temperature transducer (b) Panametrics V306 transducer

The reflector used for these measurements is a 25.4mm thick metal disc. The difference in the A-scan signals for these two transducers is evident from Fig.5.2(a-b). The PNNL high temperature transducer exhibits a long (40μs) reverberation in incidence pulse, which is an indicator of “ringing” inside the transducer as shown in Fig.5.2(a).

Figure 5.2 Immersion measurements in water at room temperature using (a) PNNL high temperature transducer (b) Panametrics V306
However, Panametrics transducer in Fig. 5.2(b) shows minimal ringing and electrical noise in the signal. Using windowed signal shown in red in Fig. 5.2(a-b), spectral analysis is performed in Fig. 5.3(a-b). For the Panametrics transducer, the spectral magnitude is maximum around 2.1MHz which appears to predominant mode of resonance. Interestingly, for PNNL transducer two predominant resonance frequencies were observed at 1.26 MHz and 2.98MHz. This is termed as “bimodal resonance” in this work here onwards.

**Figure 5.3** Spectral analysis of the windowed signal for (a) Panametrics transducer (b) PNNL research transducer

Moreover, minor resonance modes were also observed 4MHz and 7.8MHz as shown in Fig. 5.3(b). These room temperature measurements helped to identify a unique bimodal phenomenon which can introduce acoustic noise due to multimodal components in the signal. To understand the factors that might be contributing to it, more theoretical investigation is required which is discussed in the next sub-section.

### 5.1.1 Bi-Modal Resonance in the Ultrasound Transducer

It was hypothesized that the bimodal resonance could be due to multiple reflections within the transducer that could have also caused the reverberations in the incidence pulse as shown in Fig. 5.2(a). Literature review suggested that the PNNL transducer that was tested in water, consisted of silver soldering to attached piezoelectric material to nickel alloy as shown in Fig.
5.4. The theory behind ultrasonic transmission and reflections was described by equation (4.1) through (4.12) previously.

Figure 5.4 Adhesive layer to bond piezoelectric material with the matching layer [Bond, L.J.(2013)]

To investigate this multi-layer phenomenon, a finite element model is developed in COMSOL as shown in Fig. 5.5(a-b). In an ultrasonic transducer, the equivalent acoustic impedance of a layer would be function of thickness of that layer and function of propagation constants, acoustic impedance of multiple layers as shown in Fig.4.1(a). These multiple layers are modeled as shown in Fig.5.5(a). An-axisymmetric coupled problem is solved as shown in Fig.5.5(b), with a governing equation given by (4.13) through (4.16) in the previous chapter.

Figure 5.5 (a) Schematic of multiple layers involved in the transducer (b) Axisymmetric, coupled finite element model to study interaction of multiple layers (c) Boundary conditions of the model
Low reflecting boundary described by equation (4.14) is applied to backing layer to absorb incoming waves for elimination of reflections from the top boundary of backing as shown in Fig.5.5(b). The roller boundary is utilized for radial constraint to model the transducer casing. Pressure wave equation in a lossless fluid is given by [Kinsler et al. (1999)]

\[
\frac{1}{\rho c^2} \frac{\partial^2 p}{\partial t^2} - \nabla^2 p + f = 0
\]  

(5.1)

where \(\rho\) is the density of the fluid, \(c\) is longitudinal wave velocity in the fluid, \(p\) is the total pressure, and \(f\) is the body force. The bi-directional acoustic-structural coupling between solid-fluid domains as shown in Fig.5.5(b) is given by:

\[
-n \cdot \left( -\frac{1}{\rho c} \nabla p_i \right) = n \cdot \frac{\partial^2 u}{\partial t^2}
\]

(5.2)

\[F_A = p_n\]

(5.3)

where \(n\) is normal unit vector \(p_i\) is the total pressure, \(u\) is the structural displacement, \(F_A\) is the boundary load, \(\rho\) is density of the fluid. The sound soft boundary implies acoustic pressure \(p_r=0\) and also indicate a significantly low impedance boundary (Z≈0). For fluid domains, the absorbing boundary can be represented by first order transient radiation boundary condition given by [Bayliss et al. (1982)]

\[
-n \cdot \left( \frac{-1}{p} \nabla p_i \right) + \frac{1}{\rho c} \frac{\partial p}{\partial t} + \kappa(r) p = \frac{1}{\rho c} \frac{1}{\partial t} + \kappa(r) p_i + n \cdot \nabla p_i
\]

(5.4)

where \(p\) is the pressure in the fluid domain, \(p_i\) is the incident pressure field, \(r\) is the shortest distance between \(\kappa(r)\) is function of wave-type which equals 1/2r for cylindrical waves, 1/r for spherical waves and 0 for plane waves. Cylindrical wave radiation and plane wave radiation
also acts as a low reflection boundary for the axisymmetric acoustic problems. Table 5.1 shows material parameters used in the finite element model. Material parameters for PZT-5A are same as listed in Table 3.1. Equation (3.19) and (4.16) are used for spatial and temporal discretization of the model.

### Table 5.1 Material parameters for the epoxy, backing layer, faceplate and fluid medium

<table>
<thead>
<tr>
<th>Medium</th>
<th>$c_p$ (m/s)</th>
<th>$c_s$ (m/s)</th>
<th>$\rho$ (kg/m$^3$)</th>
<th>$Z_{adh}$ (MRayl)</th>
</tr>
</thead>
<tbody>
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<td>epoxy</td>
<td>2660</td>
<td>1330</td>
<td>1200</td>
<td>3.5</td>
</tr>
<tr>
<td>Backing layer</td>
<td>1674</td>
<td>824</td>
<td>5766</td>
<td>9.6</td>
</tr>
<tr>
<td>Faceplate (nickel)</td>
<td>5710</td>
<td>2996</td>
<td>8800</td>
<td>50.2</td>
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<tr>
<td>water</td>
<td>1481</td>
<td>-</td>
<td>1000</td>
<td>1.48</td>
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</table>

Transient simulations from 0 to 5µs are performed to study the effect of multiple layers on the transmitted pulse from the transducer into the water. Fast Fourier transform of the time domain simulation data is performed to understand spectral content of the signal. Bimodality can be defined as the ratio of magnitudes ($b_1/b_2$) of spectral response corresponding to major modes in the resonance spectrum. Figure 5.6 shows the effect of acoustic impedance of a bonding agent/adhesive on the bimodality in the resonance spectrum of the transducer response.

![Figure 5.6](image.png)

**Figure 5.6** Effect of acoustic impedance of adhesive on the bimodality
Furthermore, the effect of acoustic impedance of bonding agent can be quantified as given in Table 5.2. The bimodality increases from 3.12 MRayl to 24.2 MRayl before again reducing to 0.65 for Z=37.8 MRayl.

**Table 5.2** Effect of acoustic impedance on the bimodality for a constant thickness (0.1mm)

<table>
<thead>
<tr>
<th>Material</th>
<th>Z (MRayl)</th>
<th>( b_{1f} ) (MHz)</th>
<th>( b_{2f} ) (MHz)</th>
<th>( b_1 )</th>
<th>( b_2 )</th>
<th>( b_1 / b_2 )</th>
<th>( b_{1f} / b_{2f} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Epoxy</td>
<td>3.12</td>
<td>0.67</td>
<td>2.267</td>
<td>3.12E-04</td>
<td>7.26E-04</td>
<td>0.43</td>
<td>0.3</td>
</tr>
<tr>
<td>Lead</td>
<td>24.6</td>
<td>1.17</td>
<td>2.6</td>
<td>7.11E-04</td>
<td>5.28E-04</td>
<td>1.35</td>
<td>0.45</td>
</tr>
<tr>
<td>Tin</td>
<td>24.2</td>
<td>1.23</td>
<td>2.77</td>
<td>7.44E-04</td>
<td>4.32E-04</td>
<td>1.72</td>
<td>0.44</td>
</tr>
<tr>
<td>Silver</td>
<td>37.8</td>
<td>0.833</td>
<td>2.33</td>
<td>4.45E-04</td>
<td>6.81E-04</td>
<td>0.65</td>
<td>0.36</td>
</tr>
</tbody>
</table>

Moreover, an iterative study was also performed to study effect of bonding layer thickness on bimodality for a fixed acoustic impedance as shown in Fig. 5.7, Table 5.3 and 5.4.

**Figure 5.7** Effect of thickness variation on the bimodality (a) for acoustic impedance of epoxy (3.12 MRayl) (b) lead (24.6 MRayl)

**Table 5.3** Thickness effect for the acoustic impedance of epoxy

<table>
<thead>
<tr>
<th>Thickness (mm)</th>
<th>( b_{1f} ) (MHz)</th>
<th>( b_{2f} ) (MHz)</th>
<th>( b_1 / b_2 )</th>
<th>( b_{1f} / b_{2f} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.15</td>
<td>1.07</td>
<td>2.47</td>
<td>1.04</td>
<td>0.43</td>
</tr>
<tr>
<td>0.1</td>
<td>1.13</td>
<td>2.6</td>
<td>1.36</td>
<td>0.44</td>
</tr>
<tr>
<td>0.07</td>
<td>1.2</td>
<td>2.73</td>
<td>1.65</td>
<td>0.44</td>
</tr>
<tr>
<td>0.05</td>
<td>1.23</td>
<td>2.83</td>
<td>1.91</td>
<td>0.44</td>
</tr>
</tbody>
</table>
Table 5.4 Thickness effect for the acoustic impedance of lead

<table>
<thead>
<tr>
<th>Thickness (mm)</th>
<th>b₁f (MHz)</th>
<th>b₂f (MHz)</th>
<th>b₁/b₂</th>
<th>b₁f/b₂f</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.15</td>
<td>0.7</td>
<td>2.2</td>
<td>0.35</td>
<td>0.32</td>
</tr>
<tr>
<td>0.1</td>
<td>0.83</td>
<td>2.27</td>
<td>0.5</td>
<td>0.37</td>
</tr>
<tr>
<td>0.07</td>
<td>0.97</td>
<td>2.33</td>
<td>0.69</td>
<td>0.41</td>
</tr>
<tr>
<td>0.05</td>
<td>1.1</td>
<td>2.43</td>
<td>0.89</td>
<td>0.45</td>
</tr>
</tbody>
</table>

It was observed that reduction in the thickness of adhesive from 0.15 to 0.05mm increases bimodality of the spectrum in both (Z=3.12, 24.6MRayl) cases as seen from Table 5.3 and 5.4. However, ratio of frequencies (b₁f/b₂f) corresponding to this bimodality, increased in case of lead (Z=24.6MRayl) whereas the ratio was marginally affected in case of epoxy (Z=3.12MRayl). Another factor which can affect the bimodality is the thickness of the faceplate of the transducer. The PNNL transducer consisted of a nickel faceplate with typical acoustical properties listed in Table 5.1. Figure 5.8 (a-c) demonstrate the effect of faceplate thickness on the resonance spectrum.

![Figure 5.8](image)

Figure 5.8 Effect of faceplate thickness variation on bimodality for case of (a) epoxy, 0.1mm thick (b) lead, 0.1 mm thick
At 2.25MHz, the wavelength in nickel corresponds to 2.54mm. The effect of reduction in the thickness of nickel faceplate are further quantified in Table 5.5 and 5.6. In both cases, the thickness of one wavelength as well as half wave length, the simulated resonance spectrum shows multiple modal response instead of a bimodal response. For further reduction in the thickness, the bimodality increases and so as the ratio of frequencies ($b_1/b_2$) corresponding to the bimodal spectrum.

Table 5.5 Effect thickness of nickel faceplate on the bimodality (epoxy,0.1mm thick)

<table>
<thead>
<tr>
<th>Nickel</th>
<th>$b_1$(MHz)</th>
<th>$b_2$(MHz)</th>
<th>$b_1/b_2$</th>
<th>$b_1/b_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\lambda$ (2.54mm)</td>
<td>Multi-modal response (4 modes)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\lambda/2$</td>
<td>Multi-modal response (3 modes)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\lambda/4$</td>
<td>0.67</td>
<td>2.267</td>
<td>0.43</td>
<td>0.3</td>
</tr>
<tr>
<td>$\lambda/8$</td>
<td>0.87</td>
<td>2.267</td>
<td>0.5</td>
<td>0.38</td>
</tr>
<tr>
<td>$\lambda/16$</td>
<td>1.1</td>
<td>2.3</td>
<td>0.61</td>
<td>0.48</td>
</tr>
</tbody>
</table>

Table 5.6 Effect thickness of nickel faceplate on the bimodality (lead,0.1mm thick)

<table>
<thead>
<tr>
<th>Nickel</th>
<th>$b_1$(MHz)</th>
<th>$b_2$(MHz)</th>
<th>$b_1/b_2$</th>
<th>$b_1/b_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\lambda$ (2.54mm)</td>
<td>Multi-modal response (4 modes)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\lambda/2$</td>
<td>Multi-modal response (3 modes)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\lambda/4$</td>
<td>1.133</td>
<td>2.6</td>
<td>1.36</td>
<td>0.44</td>
</tr>
<tr>
<td>$\lambda/8$</td>
<td>1.367</td>
<td>2.9</td>
<td>2.21</td>
<td>0.47</td>
</tr>
<tr>
<td>$\lambda/16$</td>
<td>1.567</td>
<td>3.2</td>
<td>4.27</td>
<td>0.49</td>
</tr>
</tbody>
</table>

Figure 5.9 Literature data on bimodality (a) Kazys, R. et al. (2015)  (b) Dang (2001)
Similar, bimodal spectrum was numerically obtained by Kazys et al. (2015) for different acoustic impedances of adhesive and by Dang (2001) for epoxy adhesive as shown in Fig.5.9(a-b) respectively.

Current modeling approach demonstrated that the acoustic impedance and thickness of transducer components can result in a bimodal response with varying sensitivity. This approach also helped to quantify effect of transducer components properties on the dynamic response of the transducer. As summarized in Chapter 2, nickel with a high acoustical impedance acts as a design constraint due to a better wetting ability with liquid sodium. The benchmarking measurements revealed a unique bimodal phenomenon for which investigation was performed using numerical study. This numerical study further underlined the effects of acoustic impedance, and thickness of bonding, nickel faceplate layer on the bimodal resonance spectrum. Such numerical study will potentially help to develop and analyze new immersion transducer with similar design constraints, which will be discussed in the next section.

5.2 Design and Evaluation of New Immersion Transducer

The major challenge in proving an effective under sodium viewing capability has been the development of ultrasonic immersion transducers that can operate for an extended period at high temperatures. Swaminathan et al. (2012,2013) suggested two different operating frequencies for transducers used for under sodium viewing- a) 5MHz high frequency transducer for imaging b) 1MHz low frequency transducer for ranging purposes. To enable use of the same transducer optimally for ranging and imaging purposes, 2.4 MHz is selected as the center frequency.

Target distances for practical under sodium viewing are generally greater than 200mm which is in the far field of the beam for 10-20 mm diameter and 1-5MHz ultrasonic transducers. Moreover, a focused beam would diverge more at these distances requiring corrective
techniques in imaging [Swaminathan et al. (2013)]. The focusing lens can also introduce gas pockets due to shape of the lens causing attenuation of the ultrasonic signal [Swaminathan et al. (2012)]. Hence, a planer transducer design approach was adopted in the current work for development of the new transducer.

Sensitivity and resolution are two terms that are used in ultrasonic inspection to describe ability of transducer to locate flaws. Sensitivity is the ability to locate small discontinuities. Resolution is the ability of the system to distinguish discontinuities that are close together within the material or located near the part surface. For a constant center frequency, the sensitivity is related to the amplitude of the small discontinuities whereas resolution is related to the pulse length or bandwidth of a transducer.

These criteria are dependent on the acoustic impedance of the backing. There are two schools of thought for the acoustic impedance of backing [State et al. (2010)]. The first thought supports a good match of backing’s acoustical impedance to that of the piezo ceramics that results in a higher transmission coefficient. These materials are called heavy-backings. As a result of heavy backing the generated pulse length is shorter which indicates increase in the bandwidth of the transducer increasing the resolution although it reduces penetration into the material. Moreover, it reduces the amplitude of the pulse causing reduction in the transmitting and receiving sensitivity. The second thought favors the use of low acoustical impedance (low filler ratios). Higher transmit and received sensitivities are achieved in this case because less energy is absorbed into the backing. However, these backings increase the length of the pulse which reduces the axial resolution which is important for under sodium viewing operations.
More importantly, due to differential thermal expansion between backing and piezoelectric material, the bond integrity could be adversely affected. To analyze this particular issue, a transient domain numerical study was performed in COMSOL as shown in Fig. 5.10.

![COMSOL model to study bond integrity between backing layer and piezoelectric material](image)

**Figure 5.10** COMSOL model to study bond integrity between backing layer and piezoelectric material

Thermal expansion simulation is performed using explicit, stationary analysis in ABAQUS. The bonding agent is modelled between the transducer components and assigned with contact stiffness. The current study uses the linear thermal expansion coefficient reported by Piezo-technologies (K350) (Piezotec, LLC) for piezoelectric material and Duralco 4703 (Cotronics Corporation) for matching layer. Table 5.7 lists material properties for the FE model.

<table>
<thead>
<tr>
<th>Material</th>
<th>Density (kg/m³)</th>
<th>Poison Ratio</th>
<th>E(N/m²)</th>
<th>β damping</th>
</tr>
</thead>
<tbody>
<tr>
<td>Araldite</td>
<td>1096</td>
<td>0.34</td>
<td>0.55e10</td>
<td>2.3e-8</td>
</tr>
<tr>
<td>Backing layer</td>
<td>5766</td>
<td>0.34</td>
<td>1.05e10</td>
<td>1.5e-8</td>
</tr>
<tr>
<td>Nylon</td>
<td>1405</td>
<td>0.27</td>
<td>0.74e10</td>
<td>5e-9</td>
</tr>
<tr>
<td>Steel</td>
<td>7890</td>
<td>0.29</td>
<td>26.51e10</td>
<td>10e-8</td>
</tr>
</tbody>
</table>

The differential thermal expansion is dependent on the coefficients of thermal expansion as well as the aspect ratio of the transducer components. The deformed geometry due a temperature gradient of 180°C is shown in Fig.5.11(a). The computed deformation due to this de-bonding is converted into an equivalent acoustic model using an air gap for the
corresponding deformation (20 microns). Hence, the de-bonding process here is basically treated as a special case of a lightly damped ultrasonic transducer in the acoustic model shown in Fig. 5.11(b).

**Figure 5.11** Transformation of thermal expansion issue into equivalent acoustic model (a) Thermal expansion simulation in ABAQUS (b) Acoustic model with air gap to simulate differential thermal expansion effect

PZT-5A and liquid sodium coolant temperature dependent properties used in the finite element model are those reported in previous studies [Sabat et al. (2007), Leibowitz et al. (1971), Dierckx M et al. (2014)]. Fig.5.12 shows results of numerical study regarding the effect reduced bond quality between piezoelectric material and the backing material [Bilgunde and Bond (2015c)]. The de-bonding serves as source of acoustic noise due to increased reverberations in the signal which increases the pulse length as shown in Fig.5.12.

**Figure 5.12** Signal with bonding (blue line) and signal with compromised bond integrity (black)
Hence, in the current approach, an air-backed design concept is adopted as shown in Fig. 5.13 to avoid potential interface issue between backing and piezoelectric material. This concept also facilitates access to the internal components of the transducer which can help in evaluation of transducer during and after high temperature measurements.

Figure 5.13 Concept design of new immersion transducer

5.2.1 Piezoelectric Material Selection

The dielectric constant characterization of BS-PT shown previously in Fig. 4.3 demonstrated transition temperature \( T_p \) to be greater than 350°C and Curie temperature \( T_c \) to be greater than 430°C. These measurements make BS-PT a potential candidate in ultrasonic transducers for under sodium viewing. Moreover, a full material matrix of BS-PT was simulated and compared with the conventional PZT-5A material using experimental database methodology reported in Chapter 3. Computational model discussed in section 5.1 and 5.2 was utilized to simulate temperature dependency of piezoelectric materials and their effect on ultrasonic signal as shown in Fig. 5.14(a-b). The material parameters for PZT-5A as a function of temperature were referred from the literature [Sabat et al. (2007)]. The full material matrix for BS-PT is given by Leist et al. (2012) and Chen et al. (2014) which was utilized in the FE model for corresponding temperatures. It can be seen that PZT-5A based transducer shows 4dB reduction in the transmitted when the temperature is increased from 15 to 195°C (\( \Delta T = 180^\circ C \)). However, simulation results for BS-PT based transducer show only 3dB
reduction when the temperature is increased from 20 to 300°C ($\Delta T=280^\circ C$) as shown in Fig.5.14(b). These simulation results further consolidated the selection of BS-PT piezoelectric material for the development of high temperature immersion transducer.

Figure 5.14 Comparison of temperature effect on piezoelectric material (a) PZT-5A (b) BS-PT

5.2.2 Faceplate Material Selection

One of the major challenges of under sodium viewing is also the wettability for liquid sodium onto the faceplate of a transducer which enables efficient transmission of the acoustic energy from the transducer into the fluid. Wettability of liquid sodium on to the faceplate of the transducer is dependent on the force balance of cohesive and adhesive forces. There have been studies on the effect of a gold [Day and Smith (1973)] and a beryllium [Joo et al. (2013), Kim et al. (2014)] coating for improving wetting of the faceplate with liquid sodium. Other experimental studies [Griffin et al. (2008,2013), Swaminathan et al. (2012,2013)] have shown that for operation of a transducer in liquid sodium, the faceplate consisting of nickel has the required wettability and mechanical properties. Hence, nickel was selected as the faceplate material and a quarter wavelength [Equation (2.5)] thick disc is bonded to the BS-PT piezoelectric material. It should be noted that a high acoustic impedance of nickel introduces a significant impedance mismatch between the low impedance water/oil and epoxy. Hence,
using a nickel faceplate proves to be a design constraint with the disadvantage of the acoustic impedance mismatch between transducer elements and fluid. Consequently, the amplitude of the resulting reflected echo signal that is utilized for ultrasonic imaging could be adversely affected.

5.2.3 Backing Layer and Casing

An air-backed transducer design increases the acoustic energy transmitted into the fluid which is necessary for maximizing capabilities for ultrasonic measurements and imaging. The theory for the design of an air-backed transducer and the resulting wave field is well understood [Kino (1970)]. Such an air-backed piezoelectric transducer element and front face layer does need to be mounted and attached to a suitable case. In the current study, a 24mm diameter and 2mm thick steel casing is used for outer body of transducers. The nickel shim is attached to the casing using high temperature epoxy as shown in Fig.5.15(a-b).

![Figure 5.15](image)

**Figure 5.15** Ultrasonic transducer (a) Schematic and (b) Prototype transducer developed at the in-house facility
5.2.4 Fabrication of Transducer

Table 5.8 lists the components used to develop the new immersion transducer. The transducer is assembled in a specific sequence and tests are performed at each step to ensure efficient generation and transmission of an ultrasonic signal needed for measurements. The piezoelectric element is first selected and electrodes are cleaned to avoid issues with bonding due to contamination, such as that due to oils from handling. Of primary importance is ensuring the electrical connection between the piezoelectric element (BS-PT) and the pulsar receiver. The first step is to join a nickel lead wire to the electrode on the piezoelectric element using the silver based conductive paste (Duralco-124). The bonding compound is cured in an oven at 120°C for two hours with post-cure heating of the bond for an additional two hours at the same temperature. This is followed by the bonding of nickel faceplate to the front electrode of the piezoelectric material (BS-PT) using a thin layer of high temperature epoxy. The bond layer (Epotek 353ND) is cured at 150°C for one hour in an oven. Using the same high temperature epoxy the assembly of nickel with the transducer, BS-PT element and the lead wire is attached to the steel casing at the final stage as shown in Fig.5.15(a-b). For electrical insulation, an alumina ceramic tube is placed around the wire attached to the back electrode.

**Table 5.8 Summary of components in new immersion transducer**

<table>
<thead>
<tr>
<th>Component</th>
<th>Material</th>
</tr>
</thead>
<tbody>
<tr>
<td>Piezoelectric material</td>
<td>BS-PT (BiScO$_3$-PbTiO$_3$)</td>
</tr>
<tr>
<td>Bonding agent</td>
<td>High temperature epoxy (Epotek 353ND)</td>
</tr>
<tr>
<td>Faceplate</td>
<td>Nickel</td>
</tr>
<tr>
<td>Casing</td>
<td>Low carbon steel</td>
</tr>
<tr>
<td>Electrical continuity</td>
<td>Silver based conductive paste (Duralco-124)</td>
</tr>
<tr>
<td>Lead wire</td>
<td>Nickel</td>
</tr>
<tr>
<td>Backing layer</td>
<td>none</td>
</tr>
</tbody>
</table>
5.3 High Temperature Immersion Measurements

A schematic of the experimental set up is shown in Fig.5.16 which is used for the immersion measurements. A borosilicate glass vessel is used to contain the surrogate fluid for the measurements. A K-type thermocouple is immersed in the fluid for the temperature measurement of the liquid in the vessel. The target specimen is a 7mm thick 1018 low carbon steel block. The specimen is kept on a stand to avoid reflection from the bottom of glass vessel. The fluid path is maintained at a distance estimated to be 22mm (±2mm) based on the time of arrival of front-wall echo for each thermal cycle. The fluid path is limited by the experimental limitation of holding the transducer in retort stand which inhibits further travel of transducer in the glass vessel. Hence, the goal of these experiments is to understand temperature effect on pulse-echo measurements for a fixed fluid path during each thermal cycle where temperature is increased from 22 to 142°C.

Figure 5.16 Schematic of the High temperature immersion experimental set-up

The glass vessel is heated using a Thermo-Fischer™ heat plate set in a fume hood as shown in the Fig.5.16. A commercial pulsar receiver (Panametrics 5052) is used with input voltage 100V, damping at 5, gain of 40dB and repetition rate at 4. A high pass filter is set at 0.3MHz to remove low frequency noise. Pulse-echo ultrasonic data is collected at 10°C interval and sampled at 500 MHz using an oscilloscope (HDO 4034) with 12-bit vertical resolution. The
data is averaged 256 times to reduce random electrical noise in the ultrasonic system. For a Newtonian fluid, the decay of plane wave amplitude as a function of distance is given by Stoke’s law of sound attenuation as follows:

\[
\alpha = \frac{\omega^2}{2\rho_0 c^3} \left[ \frac{4}{3} \eta + \eta' + \frac{\kappa(\gamma - 1)}{c_p} \right]
\]  

(5.5)

where \(\alpha\) is the sound absorption coefficient, \(\omega\) is the angular frequency of the plane wave, \(\rho_0\) is the mean density, \(c\) is the speed of sound in fluid, \(\eta\) is the shear viscosity, \(\eta'\) is the volume viscosity, \(\kappa\) is the thermal conductivity, \(\gamma\) is the ratio of specific heats and \(C_p\) is the specific at constant pressure. The attenuation of the plane wave amplitude is thus given by

\[
A(d) = A_0 e^{-\alpha d}
\]

(5.6)

where \(A_0\) is the initial amplitude and \(d\) is the distance travelled by the plane wave in fluid. As the temperature of silicone oil is increased, the sound absorption coefficient will change and cause the attenuation of the plane wave having amplitude \(A_0\) for a certain fixed distance \(d\) in the pulse-echo experiment. Equation (5.5) and (5.6) indicate that the amplitude of reflected signals from immersion measurements would be a function of temperature effect on sound attenuation in the liquid as well as on the transducer components.

### 5.3.1 Surrogate Experiments in Water

Initial series of measurements were performed with the transducer and reflector in water. Examples of A-scan data for the first heating cycle of the transducer in water at 32, 52 and 92°C are shown in Fig.5.17(a). It can be seen that the amplitude for the first, second and third echoes reduces significantly at 92°C. Moreover, an oxidative reaction occurred with the transducer casing after the first heating measurements were completed and the results of this is shown in the photograph given in Fig.5.17(b). The rust was removed using a fine emery
cloth before the next cycle of measurements were performed. To further investigate the significant reduction in the echo amplitude, additional measurements were performed with the same transducer.

![Prototype transducer](image)

Figure 5.17 Prototype transducer, (a) Immersion measurements in water during first heating (b) photograph showing rust which developed due to oxidative reaction between steel casing and water

It was also observed that heating of the water caused dissolved gases to form bubbles as shown in the Fig.5.18(a) and that these adhered to the faceplate of the transducer. The gas bubbles highly attenuated the transmitted energy into the fluid reducing the amplitude of the echo. This is also shown in the A scan data for the third heating cycle as shown in Fig.5.18(b).

![Thermal cycle-3](image)

Figure 5.18 Thermal cycle-3 (a) Gas bubbles adhering to the faceplate of transducer (b) RF signal as a function of temperature

The immersion measurements in water were repeated for three thermal cycles. The amplitude of first front wall echo for each thermal cycle are shown in Fig.5.19(a-c). The
increase in peak amplitude shown in the data in Fig.5.19(a) for cycle-1 at 62°C and in Fig.5.19(c) for cycle 3 at 82°C was due to external cleaning of gas bubbles from the faceplate of the transducer. The variation in peak amplitudes for the first front wall echoes at lower temperatures (<40°C) is potentially influenced by the angular alignment of the transducer with respect to the reflector. Due to the open case prototype, the transducer could not be connected to a gimble which is generally fitted with UHF connector to excite the transducer.

![Figure 5.19](image)

**Figure 5.19** Pulse-echo measurement in water for thermal (a) cycle-1 (b) cycle-2 (c) cycle-3

At temperatures above 90°C the gas bubbles adhere to the majority of surface area of faceplate in each thermal cycle in this temperature range, which has a strong influence on the peak amplitude as shown in Fig. 5.20(a).

![Figure 5.20](image)

**Figure 5.20** Effect of temperature on the mean (a) First front wall echo amplitude (b) Sensitivity of the transducer (dashed line indicates non-linearly fitted data)

In seeking to provide a performance metric, the sensitivity of the transducer is given by the relationship [Kibe et al. (2015)] as follows:
\[ sensitivity = \left(20 \log \frac{V_1}{V_2} - gain\right) \]  

(5.7)

where \(V_2\) is a reference voltage (1 V), \(V_1\) is the peak amplitude of the first front wall echo from the RF signal and gain is given by the settings on the pulsar-receiver. The apparent mean of sensitivity of transducer over three thermal cycles is seen to reduce by about 12 dB as the temperature increased up to 92°C as shown in the data from Fig. 5.20(b).

### 5.3.2 Surrogate Experiments in Silicone oil

Previously, it was seen that water limits the transducer performance evaluation to 92°C. Furthermore, the estimated sensitivity of transducer is significantly affected by the gas bubbles adhering to the nickel faceplate of the transducer. Hence, silicone oil [Rehman (2001), Kibe et al. (2015)] with a smoke point greater than 200°C was selected for the evaluation of the high temperature immersion ultrasonic transducer.

The experimental arrangement and procedure used with oil is the same as shown in Fig. 5.16. The RF data for the time of arrival of the first front wall echo as a function of temperature is shown in Fig. 5.21(a). The data shows that the time of flight change between 22 and 142°C due to temperature dependent changes in the longitudinal velocity for waves propagating in the various media: a) silicone oil b) 1018 low-carbon steel and c) the transducer element. The arrival time for the front wall echo is analyzed to eliminate the effect of change in longitudinal velocity of steel as a function of temperature. The time of arrival of first front wall echo for 4 thermal cycles as a function of temperature is shown in Fig. 5.21(b). It can be seen that the fitted data for time of arrival increases linearly as a function of temperature up to 142°C. The time of arrival was observed to increase by 30% from 22 to 142°C.
The amplitude of first front wall echoes for four thermal cycles in silicone oil are shown in Fig.5.22 (a-b). For temperature greater than 40°C, the echo amplitudes show a characteristic reduction as a function of temperature as seen from Fig.5.22(a-b).

This characteristic reduction is further evident from the mean amplitude and sensitivity data for the transducer shown in Fig.5.23(a-b). The mean sensitivity of the transducer is estimated to reduce by about 8 dB from 22 to 142°C as shown in Fig.5.23(b). After completion of the heating cycle, the oils and transducer were allowed to cool at a natural rate as they returned to room temperature.
The mean amplitudes of front wall echoes in silicone oil as a function of temperature were compared with published data as shown in Fig. 5.24(a-b). Interestingly, the published data also showed that the transducer sensitivities [Kibe et al. (2015)] in Fig. 5.24(a) and voltage amplitude of echoes [Fei et al. (2018)] as shown in Fig. 5.24(b) increase up to 80-90°C in silicone oil. Similar increase in the voltage amplitudes was also observed in current data as shown in Fig. 5.23(a).

Moreover, it is found that by 152°C, the ultrasonic signal is lost completely which requires further investigation. To further verify the observed performance degradation at 152°C in silicone oil and to go to higher temperatures, the fluid used for the measurements was changed.
Immersion measurements are repeated in peanut oil (smoke point 232°C) for two further heating cycles, using the same ultrasonic immersion transducer. The experimental set-up used for these measurements the same as that shown in Fig.5.16. The electrical signal was found to be lost for temperatures above 92°C for measurements in peanut oil during heating cycle-p1 as shown in Fig.5.25(a). Moreover, for heating cycle-p2 in peanut oil, a similar characteristic behavior of the electrical signal was observed for temperature above 92°C as shown in Fig.5.25(b). When the cable from the pulsar-receiver (P-R) shown in Fig.5.16 is connected directly to the surface of back electrode of BS-PT instead of nickel lead wire, the electrical signal showing the front wall-echo reappeared as shown in Fig.5.25(b). Degradation of conductivity of silver-based paste was found to have occurred at elevated temperatures as shown in Fig5.25(c).

Figure 5.25 Immersion measurements in the peanut oil using the same immersion transducer for (a) heating cycle-p1 (b) heating cycle-p2 (c) degraded electrical continuity after 9th thermal cycle
To summarize the current section, the high temperature immersion measurements were performed for 3 heating cycles in water up to 92°C and 4 heating cycles up to 142°C in silicone oil. The sensitivity of the transducer in water was significantly affected by the formation of bubbles on the faceplate of the transducer due to the dissolved gases in water. However, the use of silicone oil for immersion measurements resolved the bubble formation issue and data are compared with results for measurements in water as shown in Fig.5.26. For an increase in the temperature of fluid from 22 to 92°C represented by region-1, the sensitivity of the transducer in water reduced by 12dB whereas it is reduced by only 2.5dB in silicone oil, as shown in Fig.5.26. This further underlines the limitation of water as a surrogate fluid for initial evaluation of HT ultrasonic immersion transducers. When the temperature of silicone oil was increased up to 142°C the sensitivity of the transducer reduced by a further 5.5 dB in region-2 as shown by the data in Fig. 5.26. A total 8dB loss in the sensitivity was observed due to collective changes in sound attenuation from silicone oil and temperature effect on the transducer components.

**Figure 5.26** Thermal cycling effect on mean sensitivity of immersion transducer in water (blue) and silicone oil (black)

Upon further investigation of the prototype transducer, it was also found that the conductive paste showed poor electric continuity as temperature increased above region-1 temperature
(92°C). To further verify this phenomenon, peanut oil (smoke point: 232°C) was used for further temperature dependent immersion measurements as shown in Fig.5.25 (a-b) with the same transducer. These experiments confirmed the conductivity degradation issue of the silver paste which indicates the need to revise electric continuity design in the current prototype transducer.

5.4 Revised Design of the Immersion Transducer

In chapter 2, the 3-layer problem that was studied numerically and experimentally demonstrated the effect of temperature dependence of storage modulus of epoxy on the ultrasonic signal. Furthermore, in the previous section, it was observed that degradation of electrical continuity caused loss of signal after 92°C during high temperature measurements in peanut oil. Hence, an alternative design approach was needed to improve the reliability of the electrical and structural integrity of the transducer. Following modification were performed to the design:

a) Electrical connectivity is achieved by using spring loaded pins instead of conductive paste as shown in Fig.5.27(a). The spring-loaded pins are soldered into a threaded hub as shown in Fig.5.27(b).

Figure 5.27 Electrical continuity in the improved design (a) Spring loaded pins for electrical continuity (b) brass threaded for housing of spring loaded pins
b) Liquid coupling (silicone oil) between nickel faceplate and piezoelectric material was used to eliminate temperature dependent response of epoxy in the solid coupling technique.

c) Mechanical fasteners were utilized for contact of faceplate with the casing which also facilitated the electrical ground as shown in Fig.5.28(a). The quarter-wavelength nickel faceplate is attached to the casing using brass ring and screws located at 120° to each other as shown in Fig.5.28(a). Such design will also help to attach an external focusing lens in the future.

d) A brass knurled cap as shown in Fig.5.28(b) is used as the transducer cover compared to an open case design developed in the previous section. A UHF connector is attached to the brass cap to transfer energy from pulsar-receiver to the spring-loaded pins soldered to the threaded brass hub.

Figure 5.28 Mechanical fasteners in the revised design (a) three screws into brass ring to hold nickel faceplate against the casing (b) Brass, knurled transducer cap with UHF connector

5.5 Ultrasonic Imaging in Water at Room Temperature

The limitation of solid bonding technique and silver conductive paste were understood via high temperature measurements in section 5.2 and 5.3. Hence, in the revised transducer design, emphasis was given on the alternative bonding technique (liquid coupling) and electrical
continuity (spring loaded pins). This was in addition to the use of mechanical fasteners so that transducer assembly can be disassembled easily after the high temperature measurements for inspection of transducer components. To evaluate this design for under-sodium viewing capabilities, a mock-up of ultrasonic imaging operation was set up as shown in Fig. 5.29(a). For evaluating under-sodium viewing (USV) ability, different specimens were reviewed in Chapter 2 which showed that imaging features on the top surface of specimen is of prime importance for USV operations. Hence, a 12.5mm aluminum plate with machined features was selected as shown in Fig. 5.29(b). Moreover, the letters in the specimen have different milled depths. This would be particularly helpful to evaluate ability of transducer to distinguish reduction in thickness of the targets. In reactor environment, this ability could be critical to monitor the corrosion in the reactors components causing reduction in the thickness.

![Experimental set-up for ultrasonic imaging](image)

**Figure 5.29** Experimental set-up for ultrasonic imaging (a) Schematic (b) Aluminum specimen with milled surface of various depths

The transducer is immersed in water and maintained at an estimated fluid path of 60 mm. The revised design which incorporated UHF connector also helped in connecting to a gimble with which precise alignment of the transducer can be performed to avoid errors in echo amplitudes due to misalignment. The repetition rate in pulsar-receiver is kept at 100Hz, energy at 12.5μJ, damping at 100Ω, high pass filter at 100kHz, low pass filter at 20MHz, gain is 0dB
and sampling frequency is set at 250MHz. The signal is averaged only 1 times to avoid smoothing of data and assist in estimate the performance of the transducer. The front wall echo amplitude image is shown in Fig.5.30(a). However, the thickness variation is not evident from the amplitude image, for which time of flight image is obtained as shown in Fig.5.30(b).

![Amplitude Image](image1)

![Time Of Flight Image](image2)

**Figure 5.30** Ultrasonic C-scan (a) Amplitude image(c) time of flight image

The thickness variation in the different milled letters is clearly visible from the color plot image in Fig.5.30(b). The dark blue region represents a non-machined region in the specimen.

To further quantify the thickness variation, A-scan signals on the letter to are referred as shown in Fig.5.31(b-d). These A-scan signals are compared with the baseline signal from the non-machined surface on the aluminum plate as shown in Fig. 5.31(a).
A fast Fourier transform of the A-scan signal is performed for a window starting at 80.8μs to 90.9μs to analyze spectral content in the signal. As seen previously in section 5.2, in this case as well, the bimodal resonance phenomenon persisted. However, the primary resonance mode was observed around 0.88MHz and secondary resonance was observed at 2.5MHz as shown in Fig.5.32. Further investigation and measurements are needed to understand the significant resonance at 0.88MHz.
Figure 5.32 Spectral analysis of the A-scan signals

Subsequently, the machined surface of the specimen was rotated upside down to evaluate the ability to detect back-surface features. Fig.5.33 shows the amplitude C-scan of the backwall surface features in which I, S, U letters are visible with respect to the non-machined surface.

Figure 5.33 Backwall echo amplitude C-scan
5.6 Conclusions

Current work presented modeling and surrogate experiments to evaluate high temperature transducers for potential application in under sodium viewing. The benchmarking procedure of available transducers revealed a unique bimodal resonance phenomenon. The numerical study demonstrated sensitivity of bimodality towards acoustic impedance and thickness change in the multiple layers in the transducer.

Using finite element modeling and experimental characterization, an air backed, BS-PT based single element transducer was developed for evaluation. Surrogate fluids were utilized for evaluation of the temperature dependence of the immersion transducer. Initial immersion measurements in water showed a sensitivity loss of 12dB for the transducer at 92°C and was primarily due to generation of gas bubbles from the dissolved gases. However, silicone oil as a couplant fluid resolved this issue and caused only 2.5dB loss of sensitivity up to 92°C and further 5.5dB loss up to the temperature 142°C. Upon further investigation of the transducer in peanut oil, it was demonstrated that the degradation of electrical continuity of the silver based conductive paste primarily caused the reduction in the sensitivity above 92°C.

In summary, the prototype high temperature immersion transducer developed and evaluated using the current methodology completed a total of nine thermal cycles and exceeded 27 hours of operation while immersed in the surrogate fluid media up to 142°C. Using the analysis of acquired data, the design was modified to develop a final prototype of the transducer. This transducer demonstrated the ability to image regions of different thickness within the specimen, critical for USV capabilities in SFRs. Such a measurement-based design methodology will be extended in the future for the quantitative evaluation of the immersion transducer at the hot stand-by temperature (~260°C) of the sodium fast reactor.
CHAPTER 6. TEMPERATURE COMPENSATED TRANSFER FUNCTION APPROACH FOR PROBABILITY OF DETECTION

Advanced piezoelectric based ultrasonic transducers offer the potential for in-coolant NDT measurements at high temperatures, including during hot stand-by (~260°C) for liquid sodium cooled advanced small modular reactors (SMRs). The reliability of the NDT measurements is typically quantified by the probability of detection (POD) measured at the corresponding temperature. Obtaining such data in liquid sodium is challenging. Using a model assisted probability of detection (MAPOD) approach a transfer function is reported that enables data obtained on low carbon steel specimens at room temperature to give an estimated POD at higher temperature. A primary source of the difference in POD between room and high temperature is due to the transducer material temperature dependent performance. This work demonstrates the transfer function approach using data for the case of modified lead zirconium titanate (PZT-5A).

6.1 Introduction

Generation IV fast nuclear reactor designs are being developed to support sustainable development, economic competitiveness, and improved safety. Past experience, specifically, with regard to long term maintenance experience from the Phoenix reactors (France) has underlined the need to provide effective and reliable inspection of components. The efficacy of the NDT inspection is often quantified by the probability of detection (POD) curve. Previous studies have shown that signal to noise ratio becomes a critical issue for transducers operating in liquid sodium at a hot stand-by temperature of 260°C [Griffin et al. (2009)]. Particularly, the reduction in the signal strength as a function of temperature reduces the ability to identify and distinguish between an ultrasonic response from the defect and electrical noise. This reduction
in signal strength can cause a reduction in POD in a high temperature (HT) environment that needs to be predicted.

From a POD perspective, actual non-destructive inspection is highly variable due to human factors, experimental limitations. Moreover, these experiments are time consuming and expensive due to the cost of the preparation of specimens with defects. A more cost-effective approach to increase confidence is using physics-based modeling to predict POD, and this has been demonstrated for many years [Meeker (2012)]. Sarkar et al. (1998) proposed a modeling methodology to estimate POD as a function of known fixed effect parameters. These models have been an integral part of a unified approach for model assisted probability of detection (MAPOD) [Thompson (2008a-b)]. The basic idea behind MAPOD is to use an understanding of the effects of physical factors on the measurement results [Thompson (2008a)]. Cobb et al. (2009) advocated development of hybrid finite element models to use in the MAPOD approach. The basis of the work states that predictive modeling should be multi-domain consisting of sensor, electronics, and power management. Crack geometry was the primary source of variability in this work. Smith and colleagues developed the Full Model-Assisted (FMAPOD) approach and also presented a transfer function approach [Smith et al. (2005)] that correlated the responses from an artificial defect to that of a real crack geometry. Aldrin et al. (2011) reported simulation-based POD studies for reliability assessment of structural health monitoring (SHM) systems within the framework of MAPOD. Jensen et al. (2010) explored uncertainty propagation through CIVA-multi technique software (CEA-LIST, France) simulation models. Wirdelius et al. (2012) also developed POD based on synthetic data obtained from simSUNDT software. Pavlovi et al. (2012) reported POD data as a function of multiple parameters in contrast to the conventional signal response analysis. Li et. al. (2014)
developed a statistical model for estimating POD based on the physical mechanisms of ultrasonic inspection. This work sufficiently demonstrates the intersection of statistical and physical modeling of ultrasonic wave propagation for the purpose of estimating POD at room temperature.

In a harsh operating environment, such as a nuclear reactor, POD can be expected to reduce over time due to deterioration of sensor performance. Subair et al. (2014) reported finite element modeling for the estimation of POD of nuclear components at room temperature. Roy et al. (2014) developed temperature dependent, physics-based modeling using experimental data up to 80°C. The objective of this work was to propose a temperature compensation strategy for guided waves. Similarly, Wang et al. (2014) reported an adaptive filtering technique for temperature compensation of Lamb waves. Recently, Salmanpour et al. (2017) proposed a new method of temperature correction and used it in conjunction with a delay and sum damage detection algorithm. The proposed method is based on baseline signal stretch with an improved minimum residual allowing correction over a larger temperature range. Gianneo et al. (2016) reported the FMAPOD approach for inspection of a copper canister to plot the POD curve for flat bottom holes (FBH) where data were calculated using CIVA-multi technique software (CEA-LIST, France). In this work, the primary source of variability was structural attenuation. Gianneo et al. (2017) extended this work using finite element models for a multi-parameter POD formulation for a Lamb waves–based SHM for light alloy aeronautical plates. Yusa et al. (2016) evaluated general effects of flaw parameters on the ultrasonic response using the numerical simulations and experimental measurements on 316L steel specimens. Recently, Janapati et al. (2016) discussed the role of POD in NDE and SHM. The focus of this work was quantifying the effect of transducer parameters on damage detection sensitivity.
However, the effect of temperature dependency of piezoelectric material on the POD is not sufficiently quantified. In the current work, temperature dependency of PZT-5A is modelled with regards to its effect on the POD. The objective of this work is to connect a microscopic material phenomenon that occurs within piezoelectric ceramics at a high temperature to an industrial practice of evaluating POD for quantifying performance of NDT inspections. This is achieved by a physics-based model using finite element (FE) software COMSOL™ (Burlington, MA) and temperature dependent material coefficients of PZT-5A [Sabat et al. (2007)]. Using the FE model, temperature correction and transfer factors are proposed to estimate POD value at high temperature using POD of room temperature experimental data.

### 6.2 Method

The problem considered in this work consists of a planar compression wave (P-wave) transducer transmitting at normal incidence through a solid-solid interface as shown in Fig.6.1. Let Ω be the domain of the problem connected by the Lipschitz boundaries ∂Ω. The low carbon steel block of 50mm x 50mm cross section and the piezoelectric transducer are represented by domain Ω_{steel} and Ω_{transducer} respectively. The piezoelectric material PZT-5A domain has a length of 12.7 mm and a thickness of 0.9 mm. Hence, the computational domain is given by Ω = Ω_{steel} ∪ Ω_{transducer} as shown in Fig.6.1.
Figure 6.1 Problem configuration to predict the response from SDHs at normal incidence

The elastic wave is scattered by the side drilled hole (SDH) in the solid. The P-wave response from the SDH is received by the same transducer in a pulse-echo solid-solid interface model. It is assumed that the plane of the transducer is perpendicular to the axis of the SDH.

In the current work, the amplitude of the planar compression wave (P-wave) reflected from SDHs is computed in terms of the vertical component of the displacement field in the finite element model. The assumptions for the physics-based model are summarized as:

(a) plain strain condition (b) the material behaves linearly elastic (c) small deformation of the plate, which thus implies that this theory is not applicable for large deformations of the piezoelectric material. This assumption is valid for the low-power piezoelectric based ultrasonic transducers which are used in linear ultrasonic measurements employed in SHM and NDE. (d) Material is continuous and homogenous which assumes uniform distribution of material properties to solve the problem within a continuum mechanics framework. This prevents the current approach from applying to the materials with significant discontinuity and inhomogeneity. (e) Damping variation due to temperature is assumed negligible since the objective of the current work is to primarily study the effect of temperature dependent piezoelectric material properties as a function of temperature. Damping mainly causes changes in the magnitude and bandwidth of the resonance frequency response. Hence, if the Rayleigh
damping coefficients as a function of temperature are measured, the temperature dependent damping response can be included in the current transient FE model. The pyroelectric and magnetization effect contribute to the mechanical strain and electric displacement of the piezoelectric material as shown in equations (6.1-2). However, the thermoelectric coupling resulting in a pyroelectric effect in the piezoelectric material is not considered and The magnetization effects are assumed negligible, reasons of which are discussed in the next section. The equations for piezoelectricity can be obtained using the Gibbs thermodynamic potential which is given as [Baptista et al. (2014)]

\[ S_i = s_{ij}^{E,H,\theta} T_j + d_{mi}^{H,\theta} E_m + d_{mi}^{H,\theta} H_m + \alpha_i^{E,H} d \theta \]  

(6.1)

\[ D_m = d_{mi}^{H,\theta} T_i + \epsilon_{mk}^{T,H,\theta} E_k + m_{mk}^{T,\theta} H_k + \rho_m^{T,H} d \theta \]  

(6.2)

where \( S_i \) is the Cauchy’s total mechanical strain tensor, \( D \) is the electric displacement tensor, \( \epsilon_{mk}^{T} \) is the absolute permittivity at constant mechanical stress \( T_i \), \( s_{ij}^{E,H,\theta} \) is the elastic compliance coefficient at a constant electric field \( E \), constant magnetic field \( H \) and constant temperature \( \theta \) and \( d_{mi} \) is the piezoelectric charge coefficient. The thermal expansion coefficient and pyroelectric constant are given by \( \alpha \) and \( p \) respectively. The magneto-dielectric coefficient is given by \( m \). Phase velocity of elastic waves for piezoelectric ceramic is significantly less than the electromagnetic waves. This implies time derivative of magnetic field \( H \approx 0 \) indicating absence of magnetization effect and presence of a quasi-static field. Thus magneto-dielectric coupling from equations (6.1-2) can be ignored. In the current work, the temperature difference \( d \theta \) is assumed to be small representing a gradual increase in the temperature of a piezoelectric based sensor. Thus, for a quasi-thermal change in the piezoelectric material, the thermal expansion and pyroelectric effect can be ignored. This reduces equations (6.1-2) to a strain charge form of linear theory of the piezoelectric effect can be given by
The piezoelectric material modeled in this study, PZT 5A, exhibits a crystal structure with a symmetry of a hexagonal 6mm class [Sabat et al. (2007)]. The piezoelectric material is considered transversely isotropic if the poling axis aligns with one of the material symmetry axes [Sabat et al. (2007)] as shown in the expanded view of transducer model in Fig.6.1. Hence, for a 2-D physics-based model, the material matrix reduces to 4 elastic coefficients, 3 piezoelectric coefficients and 2 dielectric coefficients. Hence the piezoelectric stiffness matrix can be given as

\[
\begin{bmatrix}
{s_{11}} & {s_{13}} & 0 & 0 & {d_{31}} \\
{s_{13}} & {s_{33}} & 0 & 0 & {d_{33}} \\
0 & 0 & {s_{44}} & {d_{15}} & 0 \\
0 & 0 & {d_{15}} & {\varepsilon_{11}} & 0 \\
{d_{31}} & {d_{33}} & 0 & 0 & {\varepsilon_{33}}
\end{bmatrix}
\]  

(6.5)

Further information regarding the temperature dependence of a piezoelectric material is given by [Sabat et al. (2007)]. In the current FE model, the in-plane deformation problem is considered in the x-y plane as shown in Fig.6.1. The poling axis of the soft PZT-5A coincides with the y-axis of the model. Assumed electromechanical load for the in-plane problem is given by: a) \(E_1 \neq 0, E_2 = 0\), also implies \(D_1 \neq 0, D_3 \neq 0, D_2 = 0\) and b) mechanical displacement \(u_1 \neq 0, u_3 \neq 0, u_2 = 0\). Thus, the non-zero stress and strain components are given by \(\sigma_{11}, \sigma_{33}, \sigma_{13}\) and \(s_{11}, s_{13}, s_{33}\). The assumption (b) of non-zero mechanical displacement, strain, and stress also applies to linear elastic solids in the physics-based model.

The equation of linear momentum balance in the time domain is given by [Rose, (2004)]

\[
\frac{\partial^2 u}{\partial t^2} = \nabla \sigma + F_v
\]  

(6.6)
where \( \rho \) is the assigned material density, \( u \) is the mechanical displacement, \( \sigma \) is second order Piola-Kirchoff stress tensor, and \( F_v \) is the body force. The top boundary (\( y=L \)) for the \( \Omega_{\text{steel}} \) shown in Fig. 1 is kept traction free (\( \sigma_{31}=\sigma_{33}=\sigma_{11}=0 \)). A low reflection boundary condition is applied to the \( \Omega_{\text{steel}} \) at \( x=0 \) and \( x=L \) boundaries to reduce reflection of the wave from side walls which also reduce the degree of freedom for which the model is solved, is given by [Comsol Documentation (2016)]:

\[
\sigma \cdot n = -pC_p \left( \frac{\partial u}{\partial t} \right) \cdot n - pC_s \left( \frac{\partial u}{\partial t} \cdot t \right) t
\]  
(6.7)

The vertices \( x=0 \) and \( x=L \) on the bottom boundary (\( y=0 \)) of the \( \Omega_{\text{steel}} \) are modelled as fixed constraints (\( u=0 \)) representing the fixed support used to ensure flatness of the structure during ultrasonic contact measurements. The boundaries of SDH are traction free (\( \sigma_{31}=\sigma_{33}=\sigma_{11}=0 \)). As previously stated, for the piezoelectric media, the electric field (piezoelectric media) is assumed to be irrotational. Thus, the electric field \( E \) is related to the scalar electric potential \( V \) by

\[
E = -\nabla V
\]  
(6.8)

\[
n \cdot D = 0
\]  
(6.9)

Terminal boundary is assigned to the top electrode of the piezoelectric material which is coupled to the lumped parametric model discussed in the next section. The bottom boundary of the \( \Omega_{\text{PZT}} \) is grounded (\( V=0 \)). Zero charge constraint is assigned in the domain \( \Omega_{\text{PZT}} \) at boundaries without terminal or ground boundaries given by equation (6.9). Charge density \( \rho_v \) in the domain \( \Omega_{\text{PZT}} \) is given by

\[
\nabla \cdot D = \rho_v
\]  
(6.10)
In the experimental measurements, the piezoelectric transducer is excited with the pulsar-receiver circuit. This introduces electrical impedance mismatch between the transducer and the instrumentation. This can be modelled in a finite element model by introducing a resistor of impedance equal to that of pulsar-receiver. In the current study, the impedance value is set to an ideal 50 $\Omega$. The electric circuit module in COMSOL™ evaluates global variables, voltage, and current as a function of time. The transducer model discussed previously [Bilgunde et al. (2015a)] is excited with a Hamming windowed sinusoid signal with amplitude of 160V. The current at the back electrode of the piezoelectric element $\partial^D \Omega_{PZT}$ is given by

$$\int_{\partial^D \Omega_{PZT}} D\cdot n = Q_0, \quad \frac{dQ_0}{dt} = I_{\text{cir}}$$ \hspace{1cm} (6.11)

Hence the voltage on the electrode surface $\partial^D \Omega_{\text{piezo}}$ is given by:

$$V_{\text{pc}}(t) = V_{\text{source}}(t) - I_{\text{cir}} R$$ \hspace{1cm} (6.12)

A triangular element of maximum size 0.05mm was used for meshing the complete domain $\Omega = \Omega_{SS} \cup \Omega_{\text{Transducer}}$. The total number of elements is 256662 with an average growth rate of 1 and an average element quality equal to 0.99. This simulates the defect as a geometric discontinuity which is kept traction free. The maximum element size is determined by the minimum shear wave speed of the material assigned to the computational domain given by:

$$h_{\text{max}} = \frac{c}{f_0 N}, \quad t = \frac{CFL}{f_0 N}$$ \hspace{1cm} (6.13)

where $h_{\text{max}}$ is the maximum element size, $c$ is the shear wave speed in the material, $f_0$ is the highest frequency required in the spectrum, $N$ is the number of element per wavelength, $t$ is the time step, and CFL (Courant-Freidrich-Levi) number [Courant et al. (1928)] is set equal to 0.2.
6.3 Probability of Detection Formulation

The reflected signal displacement data from the physics-based model corresponds to response \( \hat{a} \). The true size (here, diameter) of the flaw \( a \) is related to response \( \hat{a} \) by [MILHDBK-1823A (Appendix G)]

\[
\hat{a} = \beta_0 + \beta_1 a
\]

which is of the linear equation form given by

\[
\hat{y} = \beta_0 + \beta_1 x
\]

Equation (6.15) is of the form \( Ax = b \). Using the theory of least square solutions from linear algebra, the estimate for \( \beta_1 \) is given by as

\[
\left[ \begin{array}{c} \hat{\beta}_0 \\ \hat{\beta}_1 \end{array} \right] = \left( A^T A \right)^{-1} A^T b
\]

If system noise \( n \) is added to the equation (6.15), the modified \( \hat{y} \) is then given as

\[
\hat{y}_{\text{mod}} = \beta_0 + \beta_1 x + n
\]

where system noise \( n \) has a normal distribution \( N(0, \tau^2) \).

Using Bessel’s correction, \( \tau^2 \) is given by

\[
\tau^2 = \frac{1}{n-1} \sum (\hat{y}^2 - \hat{y}_{\text{mod}}^2)
\]

Using the Wald method, given in the standard handbook, MILHDBK-1823A (Appendix G)

\[
\text{var}(\hat{y}) = \text{var}(\hat{\beta}_0) + x^2 \ast \text{var}(\hat{\beta}_1) + 2x \ast \text{COV}(\hat{\beta}_0, \hat{\beta}_1)
\]

\[
\text{var}(\hat{y}_{\text{mod}}) = \text{var}(\hat{y}) + \tau^2
\]

The decision threshold \( y_{\text{th}} \) for model data is set to exhibit a conventional nature observed in measurements-based POD curves. The detection threshold here is the amplitude of vertical
component of displacement field $u$ from the model data. The upper and lower 95% confidence bounds are plotted using the probit function. The POD curve is computed by the equation given as,

$$POD = \Theta \left( \frac{\hat{\beta}_0 + \hat{\beta}_1 x - y_{th}}{\sqrt{\text{var}(\hat{y}) + \tau^2}} \right)$$

where $\Theta$ is the cumulative distribution function. It should be noted that $\hat{y}$ is the displacement amplitude of the P-wave response from the SDHs computed using the physics-based model described previously.

### 6.4 Room Temperature Benchmark Experiments

For the experimental verification of the model, SDHs are machined into a 1018 low carbon steel test blocks at a depth of 25mm as shown in Fig.6.2. A V306 Panametrics 2.25 MHz planar piezoelectric transducer with 12.7 mm nominal element diameter was used for normal incidence contact measurements. For the near-field length (N) of 15.6 mm, 6dB beam width in the steel at the measured depth of SDHs (1.6N) is 5mm. As the defect size approaches the transducer beam width, the response is not only a function of size of defect, but also a function of reflection ratio and beam width [Sarkar et al. (1998), Li et al. (2014)]. Hence, the largest flaw size is limited to 3.9 mm to reduce the beam width limitation effects. The minimum distance between two adjacent SDHs (22 mm) is also greater than the -6dB beam width at a given depth (1.6N) of the SDHs.
Figure 6.2 Side drilled holes with diameter varying from 0.46mm to 3.9 mm in a 1018 low carbon steel block

The Panametrics 5052 pulsar receiver with energy setting at 2, repetition rate at 4, 40 dB gain and damping set at 5 is used with a 1m coax cable and BNC connectors. The measurements acquired using LeCroy HDO 4034 oscilloscope with sampling frequency of 450MHz are averaged 512 \(2^9\) times to minimize random electrical noise in the A-scan. Contact measurement for each SDH is repeated 6 times for all 16 SDHs to minimize the variability due to contact pressure.

6.5 Results and Interpretation

The mean of the six measurements for each SDH response \(\bar{x}\) is computed to compare with the simulation data. The experimental error for each SDH response is given by \(e = \sigma / \sqrt{N}\) where \(\sigma\) is the standard deviation and \(N\) is the number of measurements for each SDH. The mean value of the experimental error \(\bar{e}\) is assigned to the data as shown in Fig. 6.3(a).
Figure 6.3 P-wave Scattering amplitude for (a) Experimental data (b) Simulation data (c) Literature model data [Lopez et al.(2005)] (d) Comparison of normalized FE simulation and literature model data

FE model-based P-wave scattering amplitude as a function of SDH diameter is shown in Fig.6.3(b). Fig. 6.3(c) shows scattering amplitude model data for SDH reported by Lopez et al.(2005). The increase in the normalized scattering amplitude as a function of flaw size is in good agreement with literature data [Lopez et al. (2005)] as shown in Fig.6.3(d). For Fig.6.3(a) through Fig.6.3(c) scattering amplitude can be fitted logarithmically in the form given by $y=m[ln(x)+c]$. The experimental data shown in Fig.6.3(a) which is acquired in terms of voltage amplitude is dependent upon the excitation pulse using the pulsar-receiver circuit. The scattering amplitude from the FE simulation is dependent on the amplitude of the input...
mechanical displacement to the model. In Table 6.1, this is expressed as the multiplier factor \( m_i \) in the fitted equations.

<table>
<thead>
<tr>
<th>Data</th>
<th>Equation</th>
<th>( m_i )</th>
<th>( R^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Experimental data</td>
<td>( y = 0.0663 \ln(x) + 1.7 )</td>
<td>0.0663</td>
<td>0.95</td>
</tr>
<tr>
<td>FE model data</td>
<td>( y = 2e-8 \ln(x) + 1.5 )</td>
<td>2e-8</td>
<td>0.99</td>
</tr>
<tr>
<td>Literature model data</td>
<td>( y = 0.0395 \ln(x) + 1.89 )</td>
<td>0.0395</td>
<td>0.96</td>
</tr>
</tbody>
</table>

The multiplier factor needs to be compensated to compare model and experimental data. The scattering amplitudes from experiment and models, corrected for the multiplier effect are thus given by

\[
y_{\text{sim}}' = \frac{y_{\text{sim}}}{m_i} = \frac{y_{\text{sim}}}{2e-8} \approx \ln(x) + 1.5 \quad (6.22)
\]

\[
y_{\text{exp}}' = \frac{y_{\text{exp}}}{m_2} = \frac{y_{\text{exp}}}{0.0663} \approx \ln(x) + 1.7 \quad (6.23)
\]

\[
y_{\text{lit}}' = \frac{y_{\text{lit}}}{m_3} = \frac{y_{\text{lit}}}{0.0395} \approx \ln(x) + 1.89 \quad (6.24)
\]

Fig. 6.4 (a-b) shows a good agreement between model and experimental results for \( 0.4 < d/\lambda < 1 \) using the corrected scattering amplitudes of the SDHs from equation (6.22) through (6.24).

**Figure 6.4** Comparison of corrected P-wave scattering amplitudes from simulation data, experimental data and previous published model data [Lopez et al. (2005)] for SDHs
For the \( d/\lambda \) greater than 1, the experimental scattering amplitude shows divergence from the FE model data. The transfer factors \( m_1 \) and \( m_2 \) from equation (6.22) and (6.23) will be used to transform temperature effect from physics-based model to the experimental POD.

### 6.5.1 Scattering Amplitude Variation Due to Temperature Dependency of Piezoelectric Material

In this model case, the material coefficients corresponding to a particular temperature are based upon the temperature dependent material coefficient in the literature [Sabat et al.(2007)]. The summed temperature effect on all the material coefficients from equation (6.5) results in the reduction of the scattering amplitude with increase in temperature from 15 to 195\(^\circ\)C as shown in Fig.6.5.

![Figure 6.5](image)

**Figure 6.5** Displacement versus flaw size (diameter) due to the temperature dependence of material coefficients of PZT-5A

The magnitudes of \( s_{11}, s_{13}, s_{33}, s_{44} \) coefficients reduce whereas \( d_{33}, d_{31}, \varepsilon_{33}, \varepsilon_{11} \) increase as a function of temperature from 15 to 195\(^\circ\)C due to the extrinsic and intrinsic contributions in the piezoelectric ceramic [Sabat et al.(2007)]. The magnitude change in each of the material coefficients as a function of temperature contributes to the change in total mechanical strain \( S_i \) and dielectric displacement \( D_m \) as described by equation (6.3) through (6.5). This influence of
temperature on the piezoelectric effect is related to the P-wave scattering amplitude reduction through equation (6.6-10). The logarithmic fitting of the simulated data in the form of equation (6.14) is shown in Table 6.2 where \( \hat{a} \) is the simulated reflected echo amplitude from SDH and \( a \) is the true size of the SDH.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>Equation</th>
<th>( \beta_1 )</th>
<th>( \beta_0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>15 C</td>
<td>( \hat{a} = 1e^{-8(2*\ln(a)+3)} )</td>
<td>2e-8</td>
<td>3e-8</td>
</tr>
<tr>
<td>105 C</td>
<td>( \hat{a} = 1e^{-8(1*\ln(a)+3)} )</td>
<td>1e-8</td>
<td>3e-8</td>
</tr>
<tr>
<td>150 C</td>
<td>( \hat{a} = 1e^{-8(1*\ln(a)+2)} )</td>
<td>1e-8</td>
<td>2e-8</td>
</tr>
<tr>
<td>195 C</td>
<td>( \hat{a} = 1e^{-8(1*\ln(a)+2)} )</td>
<td>1e-8</td>
<td>2e-8</td>
</tr>
</tbody>
</table>

As the temperature increases from 15 to 105°C, \( \beta_1 \) reduces from 50% and \( \beta_0 \) remains unchanged. Increasing temperature of the piezoelectric material from 105 to 150°C reduces the value of \( \beta_0 \) by 66% while the value of \( \beta_1 \) remains unchanged. It should be noted that from 150 to 195°C there is no change in the equation used for logarithmically fitting the data.

### 6.5.2 Probability of Detection Curve with Temperature Dependency of PZT-5A

Using equations (6.14) through (6.21), POD values are calculated and the resulting data is plotted from the physics-based model data for different temperatures. The detection threshold \( Y_{th} \) is expressed as the amplitude of vertical component of the mechanical displacement field \( u \) from the model data. Fig. 6.6 (a) through Fig.6.7(b) show POD values for 15 to 195°C.

![Figure 6.6 POD at (a) 15°C (b) 105°C for the predefined detection threshold \( Y_{th} \)](image-url)
It should be noted that as the temperature is increased, the detection threshold needed to be lowered due to reduction in the scattering amplitude as explained in the previous section. However, the detection threshold is conventionally a system limitation which should be unmodified irrespective of the temperature. Hence, $Y_{th}$ (3.5e-8 mm) at 15°C is selected as a fixed threshold for temperatures up to 195°C.

Figure 6.7 Probability of detection at: (a) 150°C (b) 195°C for the predefined $Y_{th}$

Figure 6.8 Effect of temperature dependence of PZT-5A on the POD curve for a fixed threshold ($Y_{th}$ at 15°C) (orange dashed line showed flaw size at POD=0.95 from 15 to 195°C) POD values for temperature increasing from 15 to 195°C are shown in Fig.6.8. It can be seen from Fig. 6.8 that, increase in the temperature of the PZT-5A introduces a considerable source of variability in the POD for a given flaw size.
The flaw size corresponding to POD=0.95 increases from 2.1mm (0.81λ) at 15°C to more than 1.5λ at 195°C as shown in Fig.6.8. This shows that detectability of the defects with a probability of 0.95 limits detection to the flaw size of 1.5λ at 195°C as compared 0.81λ at 15°C at 2.25MHz resonance center frequency. Moreover, for SDHs with a diameter between 0.54λ (1.4mm) and 1.17λ (3mm) in the low carbon steel the POD value varies significantly for 2.25MHz transducer as shown in Fig.6.8. For instance, the POD is estimated to reduce by 17% at 195°C as compared to 15°C for a flaw size 3.9mm (1.5λ) as shown in Fig. 6.8. For artificial defects such as SDHs of size 1.1λ, a significant 97% reduction in the POD value between 15 and 195°C can be seen due to the temperature effect on the piezoelectric material. The effect of temperature dependency for PZT-5A piezoelectric material properties at 2.25 MHz on the POD values is listed in Table 6.3.

Table 6.3 Model based data for POD as a function of flaw size for temperature dependence of PZT-5A

<table>
<thead>
<tr>
<th>SDH (d) (mm)</th>
<th>d/λ</th>
<th>POD (Y_{th}=3.5e-8)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>15°C</td>
</tr>
<tr>
<td>0.23</td>
<td>0.09</td>
<td>0.001</td>
</tr>
<tr>
<td>0.46</td>
<td>0.18</td>
<td>0.005</td>
</tr>
<tr>
<td>0.69</td>
<td>0.27</td>
<td>0.024</td>
</tr>
<tr>
<td>0.92</td>
<td>0.36</td>
<td>0.008</td>
</tr>
<tr>
<td>1.15</td>
<td>0.45</td>
<td>0.021</td>
</tr>
<tr>
<td>1.38</td>
<td>0.54</td>
<td>0.041</td>
</tr>
<tr>
<td>1.61</td>
<td>0.63</td>
<td>0.064</td>
</tr>
<tr>
<td>1.84</td>
<td>0.72</td>
<td>0.084</td>
</tr>
<tr>
<td>2.07</td>
<td>0.81</td>
<td>0.094</td>
</tr>
<tr>
<td>2.3</td>
<td>0.9</td>
<td>0.099</td>
</tr>
<tr>
<td>2.53</td>
<td>0.99</td>
<td>1</td>
</tr>
<tr>
<td>2.76</td>
<td>1.08</td>
<td>1</td>
</tr>
<tr>
<td>2.99</td>
<td>1.17</td>
<td>1</td>
</tr>
<tr>
<td>3.22</td>
<td>1.26</td>
<td>1</td>
</tr>
<tr>
<td>3.45</td>
<td>1.35</td>
<td>1</td>
</tr>
<tr>
<td>3.68</td>
<td>1.44</td>
<td>1</td>
</tr>
<tr>
<td>3.91</td>
<td>1.53</td>
<td>1</td>
</tr>
</tbody>
</table>
6.5.3 Temperature correction factor ($k$)

In sections 6.5.1 and 6.5.2, the material coefficients for PZT-5A are altered based on the experimental data at corresponding temperatures. The effect of the temperature dependent coefficients on the scattering amplitudes and hence the simulation-based POD is quantified in Fig. 6.5 and Fig. 6.8 respectively.

In this section, using the simulated data for temperature dependent scattering amplitudes, a correction factor $K$ for the temperature effect of PZT-5A is proposed which is calculated as the ratio of the P-wave scattering amplitudes at $15^\circ$C to the scattering amplitude at the corresponding temperature. The numerical uncertainty in the correction factor is $K \pm 0.02$ with the maximum deviation at SDH of 0.23mm diameter.

![Figure 6.9](image)

**Figure 6.9** Model based POD a) Without correction factor b) With PZT-5A temperature correction factor $K$. (dashed line (red, blue) indicate 95% confidence bounds for POD curve at $15^\circ$C.

As shown in Fig. 6.9(b), the value of $K$ at corresponding temperatures is essentially the magnitude by which the P-wave scattering amplitude needs to be compensated to match the POD at $15^\circ$C.
6.5.4 Estimation of POD at high temperature using benchmark experiments at room temperature

In order to apply the temperature correction factor $K$ developed from the physics-based model data to the actual experimental data, the detection threshold is set as

$$V_{th} = \frac{Y_{th}}{m_1} \approx 0.12V$$  \hspace{1cm} (6.25)

where $Y_{th} = 3.5e-8$ (mm) is detection threshold for the model data at 15°C and the transfer factors $m_1,m_2$ were obtained from equation (6.22) and (6.23). Now, using this temperature correction factor $K$, reduction in the scattering amplitudes for the room temperature experimental data is calculated for 105, 150, and 195°C. In this way, Fig.6.10 shows the estimation of high temperature (HT) POD using room temperature experimental data, the correction factor $K$ and transfer factors $m_1$ and $m_2$.

![Figure 6. 10 Estimation of High temperature(HT) POD using room temperature experimental POD (dashed blue line) with temperature correction factor for PZT-5A, obtained from the physics-based model](image)

**Figure 6.10** Estimation of High temperature(HT) POD using room temperature experimental POD (dashed blue line) with temperature correction factor for PZT-5A, obtained from the physics-based model.
6.6 Conclusion

Current work demonstrated a finite element pulse-echo model for scattering due to a cylindrical side drilled hole (SDH) in a steel specimen. The simulated P-wave scattering amplitude response from SDHs is in good agreement with the experimental and literature data for $0.4 < d/\lambda < 1$ after correcting for experimental and numerical uncertainties. In the experimental and simulated data, near unity value of coefficient of determination ($R^2$) indicated that the P-wave scattering amplitude due to SDH can be well represented by a logarithmic fit, as a function of a SDH diameter.

The P-wave scattering amplitude reduces as a function of temperature due to variation in the piezoelectric material parameters affecting the mechanical strain and electric displacement of the piezoelectric ceramic. This changed the slope and intercept of the logarithmically fitted data. The detection threshold for the simulation-based POD needed to be lowered as the temperature increased from 15 to 195°C. For a constant threshold value, significant reduction in POD was predicted, particularly in the SDHs with diameter from 1.4 mm ($0.54\lambda$) to 3mm ($1.17\lambda$).

A temperature correction factor is proposed using scattering amplitudes estimated for 15, 105, 150, and 195°C from the physics-based model. This correction factor represented the change in the material coefficients of PZT-5A due to temperature dependent intrinsic and extrinsic contributions within the ceramic at a high temperature. Using the transfer factors obtained from experimental and simulated data, a detection threshold is formulated to calculate POD for experimental data at room temperature.

Finally, using this experimental data, the temperature specific correction factor $K$ and the transfer factors $m_1$, and $m_2$, a model-based approach has been demonstrated to estimate POD
at high temperature using low temperature ultrasonic pulse-echo measurements. The estimated POD data shows that detectability with a probability of 0.95 limits to the flaw size of 1.41 \( \lambda \) at 195°C as compared 0.58\( \lambda \) at 15°C. In other words, the temperature dependence of PZT-5A reduces the ability of 2.25 MHz transducer to detect smaller defects which otherwise would be possible to detect at 15°C with POD=0.95.
CHAPTER 7. DISCUSSION

A high temperature ultrasonic transducer is a multi-layer transmission phenomenon which requires analysis at an individual layer as well interaction of different layers. Current work developed and demonstrated a unified analysis, synthesis-based methodology to develop a high temperature ultrasonic transducer as shown in Fig. 7.1. In this chapter, a summary of discussion of results is presented from the previous individual chapters.

Figure 7.1 Summary of current work consisting of analysis, synthesis to develop transducer

It was demonstrated that the sensitivity of resonance modes due to the temperature effects on the piezoelectric material coefficients is dependent on the percentage change in the baseline value as well as the contribution of that coefficient to the mechanical strain and electric displacement. When all the material coefficients are varied based on the temperature dependent experimental data, the combined temperature effect results in the reduction of the thickness mode resonance frequency. This reduction in the resonant frequency which is consistent with the observations from previous experimental work on the temperature effect for piezoelectric sensors. The combined effect results in a sensitivity value of 687 which is 8 times the value for
temperature dependent changes in $d_{33}$ at 195°C. This demonstrated that the magnitude of $d_{33}$ is not the sole factor that affects the resonance characteristics of the piezoelectric based ultrasonic transducers at high temperatures. It further appears that a complex interplay between material coefficients results in a reduction of thickness mode resonance frequency as the temperature is increased. This interplay was discussed in this work in terms of the contribution of each of the piezoelectric material coefficients to the mechanical strain and electric displacement.

Insights from this numerical work were utilized to select Bismuth Scandium oxide-lead titanate Piezoelectric material (BiScO$_3$-PbTiO$_3$). The dielectric constant characterization for the material showed transition temperature more than 300°C and Curie temperature more than 400°C. The full material matrix simulation demonstrated only 3dB reduction up to 300°C when compared to PZT-5A which showed 4dB reduction at a much lower temperature of 195°C. The experimental and numerical study demonstrated the ability of the BS-PT (BiScO$_3$-PbTiO$_3$) material for potential application in hot stand-by mode in sodium fast nuclear reactors.

Nickel faceplate currently acts as a major design constraint for the immersion transducer aimed at use in sodium fast reactor due to the advantage of rapid wetting. However, the acoustic impedance of nickel introduces a significant impedance mismatch causing acoustic noise in the signal and altering the frequency response. This work found a unique bimodal response in the PNNL research transducer and developed model for sensitivity characterization of the bimodal response. It was demonstrated that the thickness variation in nickel faceplate result in a multimodal response which can introduce additional acoustic noise in the ultrasonic signal reducing the resolution of the ultrasonic imaging in the reactor.

Degradation of interface between piezoelectric material and faceplate is a critical issue for
high temperature ultrasonic transducer for under sodium viewing. In this work, a 3-layer problem was set-up to analyze temperature effect using numerical and experimental study. The numerical study demonstrated the effect of reduction in storage modulus of epoxy at temperature above glass transition temperatures. A 22dB reduction in the ultrasonic backwall echoes was observed for 65% reduction in the storage modulus of epoxy. Current work utilized solid-bonding technique using high temperature epoxy between piezoelectric material and the substrate. Subsequently, in-situ measurements were performed up to hot stand-by temperature (260°C) of SFRs which showed significant reduction in the backwall echo amplitude after 200°C accompanied by 0.1MHz reduction in the resonance frequency. Moreover, the effect coupling conditions was pronounced in heating and cooling cycles which was demonstrated by change in the bandwidth and center frequency of the transducer response. Hence, liquid coupling using silicone oil was used in a revised design of the transducer.

Immersion measurements carried out for prototype transducer showed degradation and ultimately loss of signal after multiple thermal cycles. An open case prototype used in this work helped to identify the failure of silver solder paste which affected the electrical continuity. Hence, spring loaded pins were utilized in the revised design of the transducer to improve reliability of the electric continuity. Moreover, mechanical fasteners were utilized to establish contact of faceplate with the steel casing to for electrical grounding. Ultrasonic imaging was carried out using revised design of transducer at room temperature

At high temperatures, the prototype transducers were seen to have a reduction in signal strength which can cause a reduction in POD and such performance change needs to be assessed. It was evident that, in assessing performance of an NDT inspection, the experiments are, in general, time consuming and expensive, due to the cost of fabricating appropriate
sample sets which include representative populations of defects. A more cost-effective approach, which can also be used to supplement a more limited experimental program, and to increase confidence is using physics-based modeling to predict POD which is the basic idea behind model assisted POD approach (MAPOD).

Current work demonstrated a novel temperature compensated transfer function approach to estimate POD at high temperature using room temperature experimental data. The model-based approach was validated with experimental data. It was seen and validated for a PZT-5A ultrasonic transducer operating at 2.25MHz that the 95% POD at 15°C was 0.58λ and due to variation in temperature dependent properties of PZT-5A, the 95% POD was only achieved for a 1.41 λ SDH diameter at 195°C. In other words, the temperature dependence of PZT-5A reduces the ability of 2.25 MHz transducer to detect smaller defects which otherwise would be possible to detect at 15°C with POD=0.95. Hence, this transfer function approach quantified the effect on POD due to the temperature dependence of transducer material.

For a known temperature dependent behavior of materials, this approach could be extended using finite element model and room temperature experimental data to estimate high temperature POD for various non-destructive methods.
CHAPTER 8. CONCLUSIONS AND FUTURE WORK

The aim of this project was to investigate and seek to address the factors which limit the signal to noise ratio for high temperature transducers such as those for use in under sodium viewing in fast spectrum nuclear power plant.

An experimental and modeling study was used to investigate specific issues and insights were obtained to guide material selections and designs for prototype transducers which were tested at high temperatures.

Moreover, a temperature compensated transfer function approach was reported for estimating POD at high temperature using finite element model and room temperature experimental data. Specific and more detailed conclusions of the current work are:

1. **Material selection based on** $d_{33}$ **parameters is not a sufficient condition to** estimate resonance characteristics of high temperature ultrasonic transducer. A full material matrix needs to be captured in order to estimate high temperature performance.

2. **$\text{BiScO}_3$-$\text{PbTiO}_3$ (Bismuth Scandium oxide-lead titanate)** was demonstrated to be a piezoelectric material that could potentially be used at hot stand-by mode (260°C) inspection of sodium fast reactors.

3. **A high temperature ultrasonic transducer is a multi-layer system** where the interaction of different layers is equally important as the temperature dependence of an individual layer which can lead to bi-modal resonance characteristics that was found in the current work.

4. **Design, development and high temperature evaluation several prototypes resulted in an air-backed transducer** using BS-PT piezoelectric material, nickel faceplate, and liquid acoustic coupling using silicone oil which demonstrated the ability to image
regions of different thickness within the same specimen at room temperature. Future work will involve evaluating this ability at temperatures greater than 200°C.

**Future work**

Current work studied the temperature effect on the adhesive bond developed using a solid bonding technique. It was observed that the bonding agent is not suitable for applications beyond 200°C. To eliminate this interface between piezoelectric material and the faceplate spray-on technique could be used. The powdered form of piezoelectric material is sprayed on the faceplate followed by the required poling. The sprayed powdered form of piezoelectric material will potentially eliminate acoustic coupling issue with faceplate at high temperatures. Moreover, nickel faceplate is currently a major design constraint which introduces a significant impedance mismatch causing acoustic noise in the signal. Hence, further study will be required on the optimal selection of faceplate material to satisfy wetting and acoustic impedance matching criteria. A multiple matching layers scheme can potentially be developed to reduce the transmission loss due to impedance mismatch. A focusing lens can also be attached to the final prototype of the transducer presented in this work. This would help to reduce the spot size of the ultrasound beam increasing the resolution of images.
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Current work presents a numerical investigation to classify the in-situ health of the piezoelectric sensors deployed for structural health monitoring (SHM) of large civil, aircraft and automotive structures. The methodology proposed in this work attempts to model the in-homogeneities in the adhesive with which typically the sensor is bonded to the structure for SHM. It was found that weakening of the bond state causes reduction in the resonance frequency of the structure and eventually approaches the resonance characteristics of a piezoelectric material under traction free boundary conditions. These changes in the resonance spectrum are further quantified using root mean square deviation-based damage index. Results demonstrate that electromechanical impedance method can be used to monitor structural integrity of sensor bonded to the host structure. This cost-effective method can potentially reduce misinterpretation of SHM data for critical infrastructures.

Introduction

Adhesively bonded joints have applications increased use in aerospace structures, including in the Boeing 787, Airbus A380, automotive structures and civil structures. However, in-service thermal degradation, dynamic loads, improper curing, and contamination during manufacturing has the potential to reduce the reliability of the bonded joints. To attain certification of bonded joints, the Federal Aviation Administration and the European Aviation Safety Agency have established the requirement that the load bearing capability of the joint must be determined by a repeatable and reliable non-destructive inspection method. Electromechanical impedance (EMI) based bond-line monitoring is one of the promising techniques which could potentially be used for health monitoring of sensors bonded to the large civil, aircraft structures and other application of composites.
This paper reports a numerical investigation to classify the in-situ health of the piezoelectric sensors deployed for structural health monitoring (SHM) of a thin isotropic metallic plate. The methodology proposed in this work models the in-homogeneities in the adhesive with which typically the sensor is bonded to the host structure for SHM.

**Electromechanical Impedance method (EMI)**

The EMI method is based upon the theory of linear piezoelectricity. The piezoelectric material produces surface charge when a mechanical stress is applied, resulting in a mechanical wave (direct effect) and conversely produces electrical voltage due to mechanical deformation (inverse effect). This change in the surface charge and mechanical deformation causes changes in the impedance/admittance signature of the piezoelectric material. Hence, this class of material has the potential to be used in a sensor which can be bonded to the structures using adhesive, and then used for in-situ structural health monitoring (SHM).

However, an inhomogeneous distribution of mechanical properties is often observed in polymer-based adhesives such as epoxy. In adhesive layers, these inhomogeneities result from the formation of interfaces close to the substrates [Diebels et al. (2012)]. Inhomogeneities could also result from temperature and humidity effects on the adhesives. There have been many studies which have considered analytical and numerical modeling of the interface. In a seminal paper on interface modeling, Baik and Thompson (1984) reported a quasi-static model used to study the linear acoustic reflection and transmission from imperfect interfaces. According to the model, the normal and tangential stress need to be continuous at the interface. The normal stiffness $k$ and mass $m$ are used to model the adhesive with a quasi-static model. The local stiffness and inertial effect are given by $k_n$ and $m_n$ respectively, as shown in Fig.A.1.
An acoustic impedance can represent both the stiffness and density of the material. In this paper a methodology is proposed and presented based on acoustic impedance as it seeks to account for the local effects due to the interface formation.

![Axial symmetry diagram](image)

**Figure A.1** A quasi-static model representing local adhesive stiffness $k_i$ and inertial effects of changes in the local density due to inclusion or pores in the adhesive

The model is developed by considering $n$ layers of inhomogeneity in the adhesive as shown in Fig. A.2. The longitudinal wave velocity $C_{Ln}$ for the $n^{th}$ inhomogeneity is given by

$$c_{Ln} = \sqrt{\frac{E_n(1-\mu_n)}{\rho_n(1+\mu_n)(1-2\mu_n)}}$$

(A.1)

where $E_n$ is the local modulus of elasticity representing material stiffness, $\rho_n$ is the local density indicating mass effect and $\mu_n$ is a local Poisson’s ratio. The characteristic acoustic impedance $Z_n$ for the $n^{th}$ inhomogeneity is then defined as

$$Z_n = \rho_n c_{Ln}$$

(A.2)

$$Z_n = \frac{\rho_n E_n(1-\mu_n)}{(1+\mu_n)(1-2\mu_n)}$$

(A.3)

It can be seen from equation (A.3) that the local acoustic impedance $Z_n$ is a function of the local values for density $\rho_n$ and modulus of elasticity $E_n$. A schematic for the distribution of...
local acoustic impedance is shown in Fig.A.2. Thus, it is assumed that the equivalent acoustic impedance of a summation of local acoustic impedances is given by:

\[ Z_{adh} = Z_1 + Z_2 + Z_3 + \ldots + Z_n = \sum_{i=1}^{n} Z_i \]  

(A.4)

For a parallel spring system, the total stiffness constant is given by summation of individual spring stiffness constants. The basis for the summation of local \( n \) acoustic impedances in this work, thus lies in the formulation of a quasi-static model approach with springs in parallel as shown in Fig.A.1. This also implies that the deformation in each \( n^{th} \) inhomogeneity is assumed to be equal due to the excitation from the piezoelectric sensor.

![Axial symmetry](image)

**Figure A. 2** Equivalent acoustic impedance method (\( Z_{adh}=Z_1+Z_2+Z_3+Z_4+Z_5+\ldots+Z_n \))

The global material properties for the pristine epoxy bond state are given in Table A.1. The material parameters for the steel substrate and the piezoelectric material, PZT-5A, properties are given by Table A.2 and Table 3.2 respectively.

**Table A.1 Material parameter for the pristine bond state**

<table>
<thead>
<tr>
<th>State</th>
<th>( C_p ) (m/s)</th>
<th>( C_s ) (m/s)</th>
<th>( \rho ) (kg/m(^3))</th>
<th>( Z_{adh} ) (MRayl)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pristine</td>
<td>2660</td>
<td>1330</td>
<td>1200</td>
<td>3.54</td>
</tr>
</tbody>
</table>
Table A.2 Material parameters for steel substrate

<table>
<thead>
<tr>
<th></th>
<th>E(GPa)</th>
<th>Poisson ratio</th>
<th>ρ(kg/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Steel</td>
<td>200</td>
<td>0.3</td>
<td>7850</td>
</tr>
</tbody>
</table>

An axisymmetric problem was solved using a frequency domain finite element (FDFE) approach for the geometric configuration shown in Fig.3. The thickness of PZT-5A and steel substrate is 1mm whereas thickness of adhesive is 0.1 mm in the finite element model. Perfectly matching layer (PML) boundary is used to absorb side wall reflections as shown in Fig.A.3.

\[ \rho \omega^2 u + \nabla \cdot \sigma = F_i e^{i\omega t} \]  

Where \( \rho \) is the assigned material density, \( \omega \) is the angular frequency, \( F_i \) is the body force. \( Z = 0 \) and \( Z = 2.1 \) surfaces shown in Fig.A.1 are kept traction free (\( \sigma_{xz} = \sigma_{zz} = \sigma_{xx} = 0 \)). For the piezoelectric media, as stated previously, the electric field is assumed irrotational. Thus, the relationship between the electric field \( E \) and the scalar electric potential \( V \) is given as
\[ E = -\nabla V \]  
\[ n \cdot D = 0 \]  
(A.6)  
(A.7)

Zero charge constraint is assigned in the domain \( \Omega_{pz} \) at boundaries \( x=0 \) and \( x=3 \).

Charge density \( \rho_v \) in the domain \( \Omega_{pz} \) is given by
\[ \nabla \cdot D = \rho_v \]  
(A.8)

In the experimental measurements using the EMI method, the piezoelectric element is connected to an impedance analyzer using lead wires soldered onto the metal electrodes. This introduces a potential electrical impedance mismatch between the piezoelectric material and the impedance analyzer. This is modelled by introducing a resistor of impedance equal to the impedance analyzer. In the current study, the impedance value is set to an ideal 50 \( \Omega \) between node 1 and 2. The electric circuit module in COMSOL evaluates the global quantities voltage and current as a function of frequency sweep. In the current model, an AC voltage of amplitude 1.0V is applied to the piezoelectric material via external current coupling of the terminal boundary condition assigned to the piezoelectric material. The current at the piezoelectric positive electrode surface \( \partial^0 \Omega_{pz} \) is given by
\[ \int_{\partial^0 \Omega_{pz}} D \cdot n = Q_0, \quad \frac{dQ_0}{dt} = I_{ir} \]  
(A.9)

The electrical admittance can be calculated as a function of frequency using the frequency dependent current and voltage. The sensitivity of the temperature dependent material coefficient in terms of its effect on the resonance spectrum is characterized using the metric indices. One such metric is the root mean square deviation (RMSD) which is given by
\[ RMSD = \sum_{n=n_0}^{n_{n_0}} \sqrt{\frac{[Z_{E,B}(p)-Z_{E,T}(p)]^2}{Z_{E,B}^2(p)}} \]  

where \( \omega_I \) is the start frequency and \( \omega_F \) is the final frequency. \( Z_{E,B} \) is the baseline electrical admittance value of the piezoelectric + epoxy+ steel assembly; while \( Z_{E,T} \) is the admittance signature for the corresponding changes in the bond state represented by the equivalent acoustic impedance \( Z_{adh} \) as shown in equation (A.4).

**Numerical results**

As the temperature increases, it has been observed experimentally that the storage modulus of the epoxy reduces [Chen et al. (2013)]. This effect essentially reduces the stiffness of bonded assembly and hence causes it to resonate at lower frequencies. The change in the bond state due to temperature is modelled by reduction in the \( Z_{adh} \) of the pristine bond state explained in the previous section. The effect of reduction in \( Z_{adh} \) on the resonance characteristics of the bonded assembly is demonstrated and now quantified.

The reduction in \( Z_{adh} \) causes a corresponding reduction in the resonance frequency when compared with the baseline spectrum as shown in Fig.A.4. When \( Z_{adh} \) is reduced by 90% compared to the pristine bond state, the resonance characteristics approach the resonance characteristics for a free un-loaded piezoelectric disc. The reduction in the stiffness of epoxy bond also reduces the effect of the presence of the bonded steel substrate on the resonance spectrum. This causes the assembly to resonate at a frequency close to that for an un-loaded piezoelectric (PZT) disc as shown in Fig.A.4.
The change in the resonance spectrum due to change in the bond state is further quantified using an RMSD based damage index as shown with the data given in Fig. A.5. As the percentage reduction in $Z_{adh}$ increases, the damage index of the adhesive also increases. At 90% reduction in $Z_{adh}$ the damage index is 95.7 whereas the index for free resonance is 97.8 indicating that the boundary condition on the bonded piezoelectric sensor are approaching the case of traction free boundary conditions. In other words, this indicates the sensor might pop-up out of the bonded assembly. The determination of such in-situ electromechanical impedance can therefore potentially help to monitor health of the piezoelectric sensors which are used for structural health monitoring of large civil and aircraft structures.
Figure A.5 Damage index corresponding to the reduction in storage modulus of the epoxy.

Orange bar plot in Fig.A.5 shows RMSD index for free resonance of PZT when compared with assembly of bonded PZT+ epoxy+ steel assembly.

Discussion

In the previous section, the classification based on damage index was demonstrated for changes in the bond state using the electromechanical impedance (EMI) method. However, there are uncertainties associated with the EMI response which are given as below:

**Temperature dependence of piezoelectric material:**

An increase in the operating temperature can simultaneously degrade the piezoelectric material and the adhesive. This can result in misinterpretation of EMI data acquired from the piezoelectric material regarding the bond state.

This temperature effect on the resonance characteristics has been studied in detail Chapter 3. It was demonstrated that temperature increase of more than 80°C causes a considerable reduction in the thickness mode resonance frequency of the piezoelectric (PZT-5A) disc. This
observation is consistent with experimental observations by Baptista et al. (2014) and Ensiu et al. (2017) on the temperature effect on the electrical impedance signature of a piezoelectric sensor. In such cases, such a major shift of resonance frequency could be considered to be largely due to material properties of piezoelectric material indicating malfunction of piezoelectric sensor itself.

Conclusion

The current work demonstrated the equivalent acoustic impedance approach as a way to model the in-homogeneity in the adhesive. The reduction in the equivalent acoustic impedance causes reduction in the resonance frequency of the bonded assembly essentially approaching the resonance characteristics of the piezoelectric (PZT-5A) element when under the traction free boundary conditions. This change in the resonance spectrum is simulated using the principles of the electromechanical impedance method. A root-mean square deviation-based index quantified the damage of the adhesive in terms of index value. Such an in-situ electromechanical impedance method with an understanding of temperature effect can potentially be help to monitor health of piezoelectric sensors which are being used extensively for structural health monitoring of the large civil and aircraft structures.