Design and Manufacture of an Ultrasonic Transducer for Long-term High Temperature Operation

By

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A thesis submitted in conformity with the requirements for the degree of Doctor of Philosophy
Department of Mechanical and Industrial Engineering University of Toronto

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Doctor of Philosophy Mechanical and Industrial Engineering, University of Toronto

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Abstract

A novel design of ultrasonic transducers suitable for high temperatures is introduced. The transducers are targeted for trouble-free continuous operation at 700-800 °C for at least two years, with sufficiently wide bandwidth to yield good temporal resolution. The design and manufacturing procedures were formatted as a multistep sequence, adaptable to a variety of transducer specifications within a wide range of signal center frequencies (500 kHz to 10 MHz) and a wide range of signal bandwidths.

Two transducer designs were pursued based on two different piezoelectric crystals as the active element: gallium phosphate and lithium niobate. A high Curie temperature of 1200 °C for lithium niobate and a high phase transition temperature of 970 °C for gallium phosphate, make the two piezoelements suitable for our application. A one-dimensional transducer model was used to model the transducer when used as a transmitter-receiver. The optimized acoustic impedance of the transducer backing element was determined to obtain desired signal center frequency of 3 MHz with 3 dB bandwidth of 90-95%. The concept of porous ceramics as a new generation of high
temperature backing element is described. An acoustic model for wave propagation in such a medium was employed to estimate the optimal porosity and pore size for the backing element.

Various high temperature adhesives and brazing alloys were investigated to bond stable bonding at high temperatures. An aluminum-based brazing alloy was found to yield reliable bonding between lithium niobate piezoelement and porous zirconia backing and alumina matching layer. None of the bonding agents resulted in acceptable bonding in the case of gallium phosphate piezoelement.

The performance of the prototyped transducer at high temperature was tested on a steel plate. Despite some fluctuations in back-wall signal amplitude, clear signals were obtained.
Acknowledgments

I would like to express my deepest gratitude to my supervisor Professor Sinclair for his continued guidance during my PhD program. I was privileged to work with him, and benefit from his wisdom. I also would like to thank my co-supervisor Professor Coyle for his generous support and help throughout my research. Without his help, my research would have been an overwhelming pursuit. Also, I express my kind appreciation to my committee members, Professor Cobbold and Professor Ben Mrad. Their thoughtful questions and comments were greatly valued.

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I would like to thank my parents and sisters for all their unconditional support and love during my life. I owe them forever. I especially thank my wife Fatemeh whose love in the past few months has made my life meaningful.
This thesis is dedicated to my parents and my wife.
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Chapter 1

1. Introduction

Ultrasonic technology has become one of the most widely used methods in non-destructive testing (NDT). It has found applications from detecting defects in structures to material characterization [1] and imaging the human body for medical diagnosis [2]. In particular, NDT of engineering components at power, chemical and petroleum plants plays an increasingly important role with respect to safety improvement and extension of plant life.

In many cases, it is highly desirable to perform NDT at elevated temperatures during normal operation of an industrial plant. For instance, ultrasonic NDT of steel components in power plants may be performed at temperatures up to 400 °C [3]. Ultrasonic monitoring has also been used to characterize materials during manufacturing at elevated temperatures, for example, in the high
temperature extrusion and curing processes of graphite/epoxy composites [4]. Also, ultrasound Doppler velocimetry is a promising technique in measuring the flow rate of molten metal [5].

An ultrasonic transducer is a device that converts mechanical displacement to voltage (direct piezoelectric effect) and/or an applied electrical voltage to mechanical displacement (reverse piezoelectric effect). Therefore, it can be used as a transmitter or receiver for generating or detecting ultrasonic waves. In the pulse-echo mode of NDT, the transducer first generates an acoustic pulse; this pulse is then reflected back from defects in an engineering component, and the echoes are captured by the same transducer [6] (Fig. 1.1a). In the transmit-receive mode (sometimes called through-transmission), one transducer emits the ultrasonic pulse and a different transducer receives the signal after it has been transmitted through the engineering component [6] (Fig. 1.1b).

1.1. Ultrasonic transducer components

Conventional ultrasonic transducers are composed of multiple layers of material: An active element (piezoelectric disc), backing element, quarter-wave matching layer, and sometimes an additional protective wear plate, all acoustically coupled together (Fig. 1.2) [7].
The piezoelectric element is the core element of any ultrasonic transducer. NDT often requires short signals containing information over a wide frequency band to obtain good temporal resolution. However, single-crystal piezoelectric elements generally exhibit low internal damping (high value of mechanical quality “Q” factor), such that a relatively long ringdown response is seen to an impulse voltage excitation. To overcome this problem, the back surface of the piezoelement is normally bonded to an attenuative material, called a backing element that makes the output signal shorter in the time domain and wider in the frequency domain, but at the expense of a reduction in signal energy.

One of the main acoustic properties of materials is acoustic impedance. Generally, when a sound wave passes through a medium, acoustic impedance is defined as the ratio of the pressure phasor caused by the wave to the velocity phasor. Energy transmission from the piezoelement to the test piece is poor if there is significant acoustic impedance mismatch between these two materials. To improve the transmission efficiency, an intermediate “matching” layer is attached to the front surface of the piezoelement with an acoustic impedance between those of the
piezoelement and test piece (Fig. 1.2). The matching layer may also serve as a protective wear plate, or that function may be fulfilled by a separate outer layer.

1.2. Ultrasonic NDT challenges in high temperature environments

In a high temperature environment, two types of problems are encountered with ultrasonic transducers. First, individual components of the transducer may lose their functionality due to thermal or chemical instability. The piezoelectric element may lose its piezoelectric properties due to a change in its crystal shape or polarization. The backing element (frequently fabricated from a polymer-based material) may also deteriorate due to chemical decomposition at high temperatures.

Second, the transducer layers must be acoustically linked together to enable ultrasonic energy to move efficiently between them. However, the interfaces among the multiple transducer layers may become stressed and damaged by the differing thermal expansions of the various transducer components if the system is raised to a high temperature. Therefore, the selection of mutually-compatible transducer component materials, and an appropriate bonding technique, are crucial to high temperature ultrasonic NDT. However, commercially available high temperature ultrasonic transducers for continuous operation are very limited in their temperature range (up to 400 °C), relatively expensive (~$5000 per transducer), and subject to early failures.

1.3. Objectives of the project

The purpose of this study is to develop a model-based system for designing ultrasonic transducers, customized for high-temperature industrial applications. The design method will then be applied to a specific NDT challenge in the nuclear power industry. It is noteworthy that failure mechanisms of ultrasonic transducers subjected to very large gamma doses at nuclear plants are often similar in appearance to failure mechanisms at high temperatures [8], e.g., debonding,
material degradation. Therefore, a transducer designed for high temperature operation may also be suitable for use in high radiation fields.

The following specifications have been identified by an industrial partner for the final transducer:

- Signal center frequency of 2.7-3 MHz with minimum 3 dB bandwidth of 2.7 MHz (90%).
- Optimized for contact inspection of low carbon steel material.
- Functional at temperatures up to 700-800 °C.
- Continuous trouble-free operation for at least two years for 90% of transducers.
- Maximum high-volume manufacturing cost of $1,500, compared to currently-available commercial competitor's price of $5,000. (Note that competitor’s transducers are good only to 300° C, with failure rates well above 15% within two years).
- Reliable and stable acoustic coupling of the transducer elements with each other. Transducer capable of accommodating any interfacial stresses originating from differences in coefficients of thermal expansion (CTEs) of transducer components, from room temperature up to the operating temperature range of 700-800 °C.

The given target frequency and bandwidth of the transducer are provided as per the requirements of the industrial partner collaborating on this project. However, the design protocol will be applicable for development of transducers over a wide range of central frequencies and bandwidths.

To achieve the project objectives, suitable piezoelectric materials must first be identified. Candidates must be able to operate at high temperatures over the long term, while maintaining their piezoelectric properties. Complementary designs of backing and matching elements are also
required with the conditions of thermal and chemical stability at high temperatures; they must also have CTEs that are close to that of the piezoelement in order to minimize the interfacial stresses and thereby reduce the chance of interfacial bonding failure.

To this end, various bonding techniques will be investigated to obtain effective, stable bonding at high temperatures up to 800 °C. Electrical connections and assembly must also be designed in a way to avoid failures at high temperatures due to oxidation or thermal stress-induced fractures.

1.4. Thesis outline

In chapter 2, a literature review of various piezoelectric materials suitable for high temperature environments is presented. Several features of these materials are examined, including dielectric properties, mechanical coupling coefficient and thermal stability. Recent developments in high temperature backing elements are described, and advantages and disadvantages of various options are presented. A review of various bonding techniques to link together transducer components is also presented. Based on this background information, final candidates to be use in this project are proposed.

In chapter 3, a simple one-dimensional transducer model is used to estimate the transducer response when it is used as transmitter (one-way) or transmitter-receiver (two-way) as shown in Fig. 1.1. The required inputs to the model are pertinent material properties of each layer, and thicknesses of the piezoelement, backing layer, quarter-wave matching layer, and test specimen. The main output in our use of the model is the required acoustic impedance of the backing element to obtain the desired transducer signal bandwidth. Despite the limitations of this one-dimensional model, it is adequate for this rough calculation.
Having obtained the optimal acoustic properties of the backing element in chapter 3, porous ceramics are introduced in chapter 4 as a new generation of backing elements in ultrasonic transducers. An acoustic model for wave propagation in a porous media with randomly distributed spherical pores is employed to estimate the optimal porosity and pore size for the backing element. Next, suitable ceramics are selected and manufacturing processes are developed to achieve defect-free backing elements with the prescribed porosity and pore size. Manufactured samples are ultrasonically tested to confirm that their acoustic properties are consistent with model predictions and our transducer requirements.

Possible bonding techniques to join together the transducer components are introduced in chapter 5. Various high temperature adhesives and brazing alloys are tested. The bonding quality is assessed by comparing the signal-to-noise ratio (SNR) and bandwidth obtained from prototype transducers using each bonding agent. The best candidate is then identified, and the effects of temperature on signal strength, centre frequency and bandwidth are studied up to 800 °C.

Final conclusions are presented in chapter 6. Proposals for future work to improve the performance of the transducer are described. Also, a suitable design for transducer assembly is proposed. Wire leads and electrical connections are designed to withstand high temperatures. An innovative design is introduced to minimize the negative effects of any differences in the CTE among the components.
Chapter 2

2. Literature review

In this chapter, the principals of piezoelectricity and the constitutive equations for a piezoelectric element are briefly reviewed in Section 2.1. Different types of piezoelectric materials used for high temperature environments, and also recent developments in high temperature piezoelectric materials are examined. Advantages and disadvantages, problems and possible solutions associated with each material are explored, and candidate materials for use in this study are proposed.

The concept of acoustic impedance matching is introduced in Section 2.2. Optimized acoustic and physical properties for the components of an ultrasonic transducer are described to achieve optimal performance.

The backing element has a key role in the performance of the transducer and is described in Section 2.3. Various types of backing materials discussed in the literature are reviewed. A key
constraint to the selection of an optimal material for the backing element is found to be thermal instability. A proposal for a new type of backing material suitable for high temperature environments is introduced.

Various bonding techniques that have been employed for several combinations of piezoelectric element, backing and matching layers at high temperatures are described in Section 2.4. The most promising techniques are identified to be investigated for this development project.

The one-dimensional KLM model for the whole transducer and test piece is introduced in Section 2.5. This model allows a study of the dependence of transducer performance on parameters such as the acoustic properties and thickness of the matching layer.

2.1. Piezoelectric element

2.1.1 Piezoelectric constitutive equations

The piezoelectric effect originates from the polar molecules inside certain materials. As shown in Fig. 2.1a, when voltage is applied to a piezoelectric element with thickness \( l \), positive poles align toward the negative electrode and negative poles toward the positive electrode, consequently the dimensions of the material change (inverse piezoelectric effect). Conversely, the direct piezoelectric effect is observed when an applied mechanical strain produces an electrical field in the material to generate a voltage drop (Fig. 2.1b). The ratio of the mechanical energy stored in the piezoelectric element to the electrical energy supplied (or vice versa) is defined as the piezoelectric electromechanical coupling factor \( k \), which is a measure of the efficiency of the piezoelectric material.
Fig. 2.1. a) Inverse piezoelectric effect, b) Direct piezoelectric effect

For a homogeneous medium, there are six independent stress elements $T_i$. Thus, the stress state can be described by a single column vector.

$$[T] = [T_1 \quad T_2 \quad T_3 \quad T_4 \quad T_5 \quad T_6]^T$$ \hspace{1cm} (2.1)

In the same manner, the 6 components of strain $S_i$ can be written as:

$$[S] = [S_1 \quad S_2 \quad S_3 \quad S_4 \quad S_5 \quad S_6]^T$$ \hspace{1cm} (2.2)

Elements with subscript 1, 2 and 3 represent normal components of stress and strain, while those with subscript 4, 5, and 6 are shear components.

The displacement vector $[D]$ and electric field vector $[E]$ each have three elements, oriented along the 1, 2 and 3 directions.

$$[D] = [D_1 \quad D_2 \quad D_3]^T$$ \hspace{1cm} (2.3)

$$[E] = [E_1 \quad E_2 \quad E_3]^T$$ \hspace{1cm} (2.4)
For a piezoelectric material, the constitutive equations are given as [7]:

\[
[T] = [c^e][S] - [e]^t[D] \tag{2.5}
\]

\[
[D] = [e][S] + [e^s][E] \tag{2.6}
\]

in which \([c^e]\) is the \(6 \times 6\) elastic stiffness matrix measured with the electric field held constant. The \(3 \times 6\) piezoelectric constant matrix \([e]\) is written as:

\[
[e] = \begin{bmatrix}
e_{11} & e_{12} & e_{13} & e_{14} & e_{15} & e_{16} \\
e_{21} & e_{22} & e_{23} & e_{24} & e_{25} & e_{26} \\
e_{31} & e_{32} & e_{33} & e_{34} & e_{35} & e_{36}
\end{bmatrix} \tag{2.7}
\]

The \(3 \times 3\) permittivity matrix \([e^s]\), defined for the condition of constant strain (clamped), is given by:

\[
[e^s] = \begin{bmatrix}
e_{11}^s & e_{12}^s & e_{13}^s \\
e_{21}^s & e_{22}^s & e_{23}^s \\
e_{31}^s & e_{32}^s & e_{33}^s
\end{bmatrix} \tag{2.8}
\]

Equation (5) for the stress \([T]\) can be rearranged as:

\[
[T] = [c^D][S] - [h]^t[D] \tag{2.9}
\]

in which

\[
[c^D] = [c^e] + [e]^t[h] \tag{2.10}
\]

\[
[h] = [e^s]^{-1}[e] \tag{2.11}
\]

\([c^D]\) is the \(6 \times 6\) elastic stiffness matrix measured under constant electrical displacement.
2.1.2. Piezoelectric coupling factor

The general formula to calculate the piezoelectric coupling factor $k$ is given as:

$$
k = \frac{\text{Total work done by the piezoelement}}{\sqrt{\text{Total energy stored in the piezoelement}}} \quad (2.12)$$

The value of the coupling factor can vary with the boundary conditions. For the case of an ultrasonic transducer in which the thickness of the piezoelement is far less than the lateral dimensions, it can be assumed that the crystal is clamped laterally; this simplifies the expression for the thickness expansion mode coupling factor to [7]:

$$
k_t = \frac{e_{33}}{\sqrt{c_{33}^{0.5} e_{33}^{0.5}}} \quad (2.13)$$

2.1.3. Piezoelectric mechanical quality factor

The mechanical quality factor ($Q$) represents the total energy stored in the piezoelement divided by the energy dissipated in every vibration cycle of the material. Therefore, the lower the energy loss in every cycle, the higher the $Q$ value for the piezoelement. Using the result of the electrical admittance (inverse of impedance) measurement, $Q$ can be expressed by:

$$
Q = \frac{f_r}{f_1 - f_2} \quad (2.14)
$$

where $f_r$ is the resonant frequency. $f_1$ and $f_2$ are the frequencies where the admittance is 3 dB below the resonance peak value. Fig. 2.2 shows the admittance of 36° Y-cut LiNbO₃ crystal with
2.87 MHz resonant frequency measured in our laboratory at room temperature using a network analyzer (ADVANTEST R3754A)\textsuperscript{1}.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{Admittance of 36° Y-cut LiNbO$_3$ as a function of frequency measured at room temperature.}
\end{figure}

\textbf{2.1.4. Piezoelectric elements for high temperature applications}

The use of certain types of piezoelectric elements at elevated temperatures is limited by many factors, including potential lowering of the piezoelectric stress constants [$e$] and corresponding lowering of the coupling coefficient $k$, lack of chemical stability, and an increase in internal losses. Also, thermal expansion compatibility of the piezoelectric element with the adjacent layers of the transducer must be considered when selecting the appropriate candidate materials. Finally, the purchase cost or cost of production is always crucial from a commercial point of view.

\textsuperscript{1} Advantest America Inc., USA
Piezoelectric materials fall into two main categories: ferroelectric and non-ferroelectric. Ferroelectricity is a property of the materials that have spontaneous polarization; the polarization can be reversed by applying an external electrical field. Ferroelectric materials with perovskite structure exhibit high sensitivity ($D_{33}$) for ultrasonic wave reception but low temperature stability [9]. A key characteristic in all ferroelectric materials is a parameter called the Curie temperature ($T_c$), above which the piezoelectric material loses its piezoelectric properties. If the temperature is increased towards $T_c$, the crystal asymmetry which is the source of crystal polarity is gradually weakened and the lattice structure becomes more symmetric until the net polarization ultimately disappears. Table 2.1 presents key properties of several piezoelectric materials used for ultrasonic transducers operating at elevated temperatures.

Among ferroelectric piezoelectric materials, lead-zirconate-titanate (PZT) is widely used in transducers and sensors operating at room temperature. This material has relatively high values of piezoelectric constants [$e$] (~25 C/m²) and electromechanical coupling factor $k_t$ (~0.5). The major drawbacks are gradual degradation in piezoelectric properties at high temperatures over time and low $T_c$ of about 160-350 °C, depending on the presence of trace elements [10].

Higher values of $T_c$ can be found in bismuth titanate Bi$_4$Ti$_3$O$_{12}$ (BTO) and lithium niobate (LiNbO$_3$). BTO has a $k_t$ value of approximately 0.2 with low dielectric loss and $T_c$ of 800 °C [11]. LiNbO$_3$ is well-known for its high $T_c$ (1142 – 1210 °C) [11], and has been one of the strongest candidates for high temperature ultrasonic transducers in many studies, e.g., [12]. However, there have been reports that LiNbO$_3$ is limited to operation below 600 °C because of chemical instability and loss of oxygen at higher temperatures [13]. More recent developments have shown the possibility of increasing this limit and extended performance to temperatures higher than 600 °C with no sign of degradation [14].
In contrast to ferroelectric materials, non-ferroelectric piezoelectric materials generally show better thermal stability, but a lower mechanical quality factor $Q$. Unlike ferroelectric materials, non-ferroelectric materials have no Curie temperature $T_c$. For some non-ferroelectric materials, there is a phase transition temperature below the melting point, such that the application of such piezoelectric materials is restricted to below this transition temperature. If there is no phase transition temperature, then the only temperature limitation is the material melting point.

$\alpha$-quartz ($\text{SiO}_2$) is known as the first non-ferroelectric piezoelectric material used in resonators and oscillators. This material has high thermal stability [8], but major drawbacks are the relatively low piezoelectric coupling factor $k_t$ (5%) and the $\alpha$-\(\beta\) phase transition at only 573 °C. From the same group of crystals as $\alpha$-quartz, gallium orthophosphate possesses higher $k_t$ (15%) and sensitivity and has a higher $\alpha$-\(\beta\) phase transition temperature of 970 °C [9]. All these positive features have tempted scholars to investigate the performance and application of this crystal in high temperature environments [15].

Another non-ferroelectric crystal is langasite $\text{La}_3\text{Ga}_5\text{SiO}_{14}$ (LGS) with trigonal crystal structure, which means that material polarization is insensitive to temperature [9]. The main advantage of this crystal over quartz is its higher electromechanical coupling coefficient and melting point of 1470 °C which allows LGS to be used at very high temperatures. However, high temperature promotes oxygen ion diffusion throughout the crystal lattice; this causes an increase in conductivity, and higher mechanical loss [9].

Aluminum nitride (AlN) is another non-ferroelectric piezoelectric material which has attracted significant attention in the manufacture of high frequency sensors and transducers for elevated temperature applications. Ultrasonic transducers in the frequency range of 10–100 MHz have been
manufactured with AlN [16]. Piezoelectric activity of the crystal is reported up to 1100 °C in a hydrogen or carbon dioxide atmosphere. However, for operation in air, surface oxidation limits the temperature of use to below 700 °C [16].

In conclusion, long-term performance in a high-temperature transducer requires a high Curie/phase transition temperature and thermal stability; these requirements are in addition to favorable piezoelectric constants. Although a variety of composite piezoelectric materials have been reported in the literature [17], most of these are not widely available, and their precise material properties have not been published.

Among the aforementioned piezoelectric materials listed in Table 2.1, LiNbO₃ and GaPO₄ were selected as promising, widely-available candidates at the time of initiation of this project. Although a variety of composite piezoelectric materials have been reported in the literature, most of these are not widely available, and their precise material properties have not been published. However, low internal damping (high mechanical quality factor) in both of our selected crystals means that a proper design of backing element is required to obtain broadband signals.

Table 2.1. Properties of piezoelectric materials commonly used in high temperatures ultrasonic transducers (Tᶜ: Curie temp., Tᵖ: phase transition temp., Tᵐ: melting temp.).

<table>
<thead>
<tr>
<th>Piezoelectric Materials</th>
<th>Ferroelectric</th>
<th>Non-ferroelectric</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PZTs</td>
<td>BTO</td>
</tr>
<tr>
<td>limiting temperature (°C)</td>
<td>(Tᶜ)</td>
<td>(Tᶜ)</td>
</tr>
<tr>
<td>kₐ %</td>
<td>50</td>
<td>20</td>
</tr>
<tr>
<td>Q @4 MHz</td>
<td>800-1400</td>
<td>3500</td>
</tr>
</tbody>
</table>
2.2. Acoustic matching layer

To transfer energy efficiently from the piezoelement to the test piece, the thickness of the front matching layer should be one quarter wavelength at the piezoelement’s resonant frequency. Also, the acoustic impedance of the matching layer should be equal to:

\[ Z_m = \sqrt{Z_p \times Z_T} \]  

(2.15)

where \( Z_m \), \( Z_p \) and \( Z_T \) are the acoustic impedances of the matching layer, piezoelement and test piece, respectively. For our purposes, the acoustic impedance of a material is equal to the product of density times the bulk compression wave velocity \( (c_p) \).

For our transducers, the quarter-wave matching layer must have thermal and chemical stability at high temperatures (800 °C). Also, to maintain a stable bond between the piezoelement and the matching layer, their respective CTEs must be closely matched to minimize interfacial stresses at high temperatures.

2.3. Backing element

Two key properties of the backing layer are the acoustic impedance \( Z \) and the attenuation coefficient \( \alpha \). The acoustic impedance mismatch between the backing element material and piezoelement determines the portion of the pulse excitation energy transmitted from the piezoelement into the backing. By absorbing energy, the bandwidth of the transducer signal is increased. However, there is consequently less energy transmitted to the test piece such that the SNR is decreased [18].

It is highly desirable to prevent any of the ultrasound energy that has been transmitted into the backing material from being returned to the piezoelement, where it could contribute to noise or
“ghost” pulses in the received echo signal. This can be accomplished by incorporating highly attenuative materials in the backing layer. Additionally, the boundaries of the backing element can be roughened or angled such that incoming wave energy is not reflected back towards the piezoelement [19].

Designing a backing element with the desired acoustic properties and thermal stability for a wide range of temperatures is another major challenge for high temperature transducers. Backing elements in conventional ultrasonic transducers are usually made from a mixture of epoxy resins loaded with particles of a second material. Such composites have appropriately high attenuation values (1-50 dB/mm), combined with the good bonding strength of the epoxy that maintains long-term integrity of the backing element if used at moderate temperatures [20-22]. Typically, the epoxy matrix is loaded with metallic filler particles that act as acoustic scatterers; popular candidates are tungsten, iron, copper, magnesium or aluminum if high acoustic impedance is required, whereas fillers of wood dust, glass and cork are used for low impedance backings. For example, Wang et al. studied the attenuative properties of two composites based on an epoxy matrix: alumina/EPO-TEK 301² and tungsten/EPO-TEK 301 [23].

It has been observed that that epoxy-based backing materials have good adhesion to piezoelectric materials and favorable acoustic properties. However, they are not suitable for high temperature applications since most of the commonly used epoxies can withstand maximum temperatures of only 100 °C to 300 °C without permanent damage [24-25].

For high temperature use, backing elements must be designed with all of the following characteristics:

² Epoxy Technology Inc. Billerica, MA
(1) High ultrasonic attenuation coefficient.

(2) Acoustic impedance value that allows significant energy ingress from the piezoelement and damping of the pulse (but not excessive energy drain nor excessive distortion of the pulse central frequency).

(3) Thermal stability.

(4) CTE close to that of the piezoelement such that thermal stresses are kept to a minimum.

(5) Amenability to bonding to a ceramic piezoelement.

Ceramics with their characteristic high melting point and low CTE could facilitate long-term durability and stable bonding to the piezoelement. However, most ceramics have low ultrasonic attenuation. In addition, their relatively high acoustic impedance is higher than optimal for good ultrasonic coupling to a piezoelement. A new design and manufacturing technique is required to make ceramics with properties suitable for use as backing elements.

2.4. Coupling Method

Backing and matching layers have to be in intimate contact with the piezoelement over the full range of operating temperatures. There are three general methods [12] currently used to acoustically couple a piezoelectric element to the backing and matching layer: dry coupling, liquid coupling, and solid coupling.

In the dry coupling process, layers are simply pressed together at high pressure; therefore, a high quality surface finish is required to avoid any interfacial gaps. Surface irregularities can lead to total interfacial delamination or failure of the piezoelement. Also, small airgaps between any two transducer elements can substantially impede acoustic energy transmission and thereby distort the ultrasonic pulse. Therefore, pressures up to 300 MPa are required to expel the air even at an
interface between highly polished surfaces [12]. Application of dry coupling is particularly difficult for transducers with very thin piezoelectric elements or matching layer as they cannot withstand the necessary coupling pressure. In addition, for high temperature applications, it is problematic to keep the coupling pressure constant over the entire temperature range of interest due to thermal expansion effects. These challenges were observed when dry coupling was tried to bond AlN to a stainless steel waveguide, where the gaps between piezoelement and waveguide were filled by soft gold foils [26]. Good signals were observed only up to 410 °C at an interfacial pressure (at room temperature) of 60-70 MPa.

To avoid the issues with dry coupling techniques, transducer components can be acoustically bonded together using a liquid couplant. Liquid couplants can be divided into two groups: Those that are liquid at room temperature (e.g. silicone oil), and those that are solid at room temperature and melt only during high-temperature operation (e.g. glass solders). Although they provide good acoustic coupling, the major problems are leakage and chemical instability (corrosivity) of liquid couplants. In the particular case of silicone oil, its chemical stability is lost when an elevated temperature is maintained for a long period. This couplant was successfully used to link two components for ultrasound transmission as the components’ temperature was raised up to 250 °C [27]; however, the couplant then gradually evaporated.

Another liquid couplant candidate is glass solder NaPoLi, which is fluid only near the upper end of our operating temperature range [28]. It was shown that with increase in temperature above 500 °C it becomes chemically active and reacts with most transducer components unless they are coated with platinum or gold. Non-alkaline powdered glass solders LG-K1 and LG-K2 [28] proved to be much less chemically reactive than NaPoLi, but they were thermally stable only up to 600 °C. In addition to thermal stability, one more technical problem yet to be solved is how to
apply the glass solder during ultrasonic probe assembly such that a uniform layer of glass solder is generated; otherwise the piezoelectric membrane might break.

An alternative to liquid and dry bonding is to mechanically bond the layers (solid coupling technique). Solid coupling provides both a good ultrasound transmission link and mechanical bonding of transducer components. It comes in several forms: soldering/brazing, diffusion bonding, adhesive bonding, sol-gel and chemical vapor deposition (CVD) technology. Unlike the case of liquid coupling methods in which the various transducer elements can move relative to each other, close proximity of CTEs of all transducer components is a high priority for solid bonding methods.

Soldering is a widely-used method to bond transducer components together. The solder should be sufficiently compliant to accommodate any differences among CTEs of the matching, backing and piezo layers. It also must not dissolve the thin electrode coatings on the piezoelement. The commercially available 88Au-12Ge solder alloy with melting point of 356 °C has been used to bond stainless steel and copper pieces [29]. The solder alloy was effective in releasing residual stresses generated during the soldering process. In general, however, despite the good adhesion that can be realized with soldering alloys, the low melting point generally limits their application to temperatures below approximately 500 °C [30] – too low for our requirements.

Diffusion bonding is a solid-state welding technique commonly used to join similar and dissimilar metals. There have been several attempts at diffusion bonding of transducer elements, sometimes coated with a thin layer of gold. For ultrasonic inspection up to 400 °C, BTO was bonded to a stainless steel protective layer using gold-to-gold diffusion bonding [31]. A major problem is that at higher temperatures \( T > 0.5 T_c \), cracks in the piezoelectric ceramics developed.
due to localized stress just under the surface. In another project, LiNbO$_3$ was bonded to a metal protective layer by diffusion bonding [32]. Copper filamentary strands bundled together were used as an intermediate layer; this material easily accommodated the different thermal expansions of the assembled components.

Although diffusion bonding has proved to be a promising technique for joining materials, its application is still generally limited to temperatures only up to about 400 °C, even when using gold-to-gold diffusion bonding. In addition, the quality of the bond strongly depends on the surface treatment of the mating parts, pressure and temperature. Trial and error must be used to optimize the process, as theoretical models do not lead to satisfactory results [33].

Ceramic adhesives show exceptional high temperature stability compared to other bonding agents. Baba et al. [34] used a high temperature ceramic adhesive to bond Z-cut LiNbO$_3$ single crystal to austenitic stainless steel in their transducer. Silver conductive paste was used to connect the electrical lead to the top electrode. Multiple echoes from the test piece were observed at temperatures ranging from ambient up to approximately 1000 °C, with no significant loss of signal strength at elevated temperature. However, after the experiment, it was noticed that the surface of the test piece was oxidized and the silver paste appeared to have been heated above its melting temperature. Therefore, for long-term operation in a high temperature and high oxidation-prone environment, an alternative choice of conductive bonding agent was still needed.

A key point in selecting an appropriate adhesive is that any mismatch among the CTEs of the components and the adhesive may lead to the failure in the bonding layer. This prompted the use of a high temperature ceramic adhesive, Contronics 989$^3$, to bond X-cut gallium orthophosphate

---

$^3$ Contronics Corp., Brooklyn, NY
(GaPO₄) crystal to titanium plate [35]. Clear signals were observed in a pitch-catch testing mode at temperatures up 426 °C, but they faded away at 480 °C. The reason was believed to be the failure of the bonding layer between the sensor and substrate. The same adhesive was used to bond AlN single crystal to silicon carbide (SiC) rod [36]. That combination yielded a consistent ultrasonic signal amplitude that was largely independent of temperature, for temperatures well below 1000 °C. However, the response degraded significantly after exposure of the transducer to temperatures of 1000 °C for 8 hours due to oxidation of the AlN crystal.

Despite many attempts, a major challenge with adhesives continues to be their long-term bonding stability at elevated temperatures. This is a strong function of the compatibility of the components with the adhesive in terms of the adhesion mechanism. Trial-and-error continues to be a primary design tool for matching adhesives with transducer components for high temperature operation.

For all of the aforementioned solid coupling methods, individual layers are bonded together after the layers have been manufactured. However, there are bonding techniques in which the component manufacturing stage is integrated with the coupling stage. Two recently-introduced methods of this type for linking together high-temperature transducer components are sol-gel [37-39] and CVD [40] technologies.

Ultrasonic transducers made of LiNbO₃/PZT with an 8 MHz center frequency were fabricated using a sol-gel technique [41]: A piezoelectric composite was deposited on a titanium rod and showed an acceptable SNR at 800 °C. Generally, sol-gel has been shown to be applicable only for thin piezoelement layer deposition (less than 200 μm) and therefore only for relatively high frequencies. Depending on the piezoelement used, the frequency range is 5-30 MHz [37-42].
AlN films with 30 MHz center frequency have been deposited on platinum coated quartz, sapphire, and lithium meta-niobate (LMN) via the CVD process to make ultrasonic transducers [43]. Due to surface oxidation of the AlN layer, ultrasonic measurements were unsuccessful at temperatures above 1150 °C.

Extensive research was also carried out at Dayton University to deposit AlN using CVD [44]. This technique was used to make films up to 0.1 mm thick with deposition times less than 1 hour. Films deposited on tungsten carbide substrates were useful up to temperatures of about 700 °C, where oxidation of the carbide substrates becomes problematic. Titanium was also tried as a substrate for AlN deposition, since its acoustic impedance is closer to that of AlN. However, upon heating in air, the films tended to lose adhesion to the substrate; this was believed to be caused by the mismatch in CTEs of the two materials.

Studies are continuing to find substrates more compatible with AlN deposition and also to examine different materials for coupling the transducer to a test piece at temperatures up to 1000 °C. Despite its favorable acoustic coupling characteristics, CVD is currently limited to thin film deposition of the piezoelectric element to avoid its fracture and/or delamination; this limits application of the technique to relatively high frequency transducers (f>10 MHz).

In conclusion, dry coupling has been shown to be generally unsuitable for transducers with thin layers as they cannot withstand the high pressure required for good acoustic coupling (The method is still used in specialized applications such as coupling a piezoelement to thick buffer rods [45]). As for liquid coupling techniques, the corrosivity and high thermal stresses during heating and cooling cycles limit the application of these materials to temperatures below 600 °C. CVD and
sol-gel techniques have shown promising results for thin films, but can lead to de-bonding and cracking if applied to the manufacture of relatively thick, low-frequency piezoelements [36-45].

Appropriate choices of ceramic-based adhesives and/or high temperature brazing alloys are the options to be pursued in this project for high temperature transducers. They must be chemically stable over the temperature range of 20 °C up to 800 °C. They must also be sufficiently compliant to accommodate any differences in the CTEs of the various transducer layers.

2.5. One-dimensional KLM transducer model

Although wave propagation in ultrasonic transducers is a three dimensional topic, one-dimensional (1D) models provide a useful starting point to predict the behavior of a transducer and optimize the design. In these 1D models, the thickness of each layer is assumed to be far smaller than the two lateral dimensions.

For a piezoelement with cross-sectional area $A$ and thickness $l$ (Fig. 2.3), it is assumed that the polarization is along the axial ($z$) direction and that lateral strains are negligible. $V$ and $I$ are the voltage and current between the two surfaces of the piezoelement. $F_1$ and $F_2$ are the forces on the two surfaces defined to be positive when acting into the surfaces. Similarly, $v_1$ and $v_2$ are the velocities of the two surfaces oriented as shown in Fig. 2.3.
Solving the 1D wave equation along the $z$-direction, it can be shown that [7]:

\[
\begin{bmatrix}
F_1 \\
F_2 \\
V
\end{bmatrix} = -j \begin{bmatrix}
Z_p \cot(\beta l) & Z_p \csc(\beta l) & \frac{h}{\omega} \\
Z_p \csc(\beta l) & Z_p \cot(\beta l) & \frac{h}{\omega} \\
\frac{h}{\omega} & \frac{h}{\omega} & \frac{1}{(\omega C_0)}
\end{bmatrix} \begin{bmatrix}
v_1 \\
v_2 \\
l
\end{bmatrix}
\]

(2.16)

In Eq. 2.16, $Z_p$ and $C_0$ are the piezoelement’s total acoustic impedance and clamped capacitance, respectively; they are defined as:

\[
Z_p = AZ_P = A \sqrt{\rho_p c_D^{33}}
\]

(2.17)

\[
C_0 = A \varepsilon^s / l
\]

(2.18)

$Z_p$ is the piezoelement acoustic impedance in units of Rayls (Pa.s/m). $\beta$ is the wave number defined by:

\[
\beta = \omega / \sqrt{c_D^{33} / \rho_p}
\]

(2.19)

where $\rho_p$ is the piezoelement density.
The KLM model shown in Fig. 2.4 contains two quarter-wave acoustic transmission lines with their common terminals connected to the transformer [7]. The transformer acts as an interface between the electrical and mechanical parts of the circuit. At the electrical port, there are two capacitors in series: the clamped capacitor $C_0$ (Eq. 16) and dynamic capacitor $C'$. $C'$ shows negative capacitance when the frequency is less than resonant frequency ($\omega_0$) and approaches $-\infty$ as the frequency approaches $\omega_0$.

![Fig. 2.4. KLM model for a disc transducer [7].](image)

From a design standpoint, the KLM model [46] provides a means to study the effects of various termination conditions at the two acoustic ports. This provides a useful tool to optimize the design of the transducer. Using the KLM model and transmission matrix method [47], the transmission response (induced pressure over the input voltage) and reception response (received voltage over the input voltage) for the ultrasonic system can be found using the model below:
Fig. 2.5. Transmission matrix method for a) Transmission and b) Reception response of the transducer using KLM model.

In the model shown in Fig. 2.5, a single front matching layer is assumed $[A^M]$. In Fig. 2.5a, the front and back acoustic ports of the transducer are loaded with the acoustic impedances of the test piece ($Z_T$) and backing ($Z_B$) media, respectively. The input voltage ($V_s$) is applied to the piezoelectric element, and leads to a pressure field $F_2$ in the test piece. For the arrangement shown in Fig. 2.5b, the transducer is used as a receiver and the electrical port is connected to an oscilloscope with input impedance of $Z_L$. The reflected pressure $F_{2R}$ from the test piece hits the transducer front surface which generates a voltage $V_R$ between the two piezoelement surfaces. The received voltage $V_R$ is then recorded on the oscilloscope. By combining the two models, the total transmit-receive performance of the transducer can be studied [7].
2.6. Concluding remarks

Two piezoelectric materials are proposed as our candidates for high temperature transducers based on their favorable properties: LiNbO$_3$ and GaPO$_4$. Therefore in this project, two separate, parallel lines of transducer development will be followed, based on LiNbO$_3$ and GaPO$_4$ piezoelectric materials, respectively. This parallel path approach is adopted in case insurmountable obstacles are encountered on one of the two development paths.

To obtain thermally stable backing elements with optimized acoustic properties, the novel idea of porous ceramics for damping of an ultrasonic transducer is introduced in this project. Our motivation is that ceramics with high melting point and CTE close to that of the piezoelement could facilitate long-term durability and stable bonding of the transducer components at high temperature. By tailoring the porosity and pore size of the backing material to appropriate values, the optimal acoustic impedance and attenuation can be obtained. The manufacturing process for the porous ceramics is then tuned to achieve defect free samples.

Commercially available high temperature adhesives and brazing alloy are identified. The performance of each bonding agent is investigated to select those which yield stable bonding and acceptable signal quality. Such bonding techniques are judged to be simpler to implement, more cost-effective, and more stable than other bonding methods such as liquid or dry coupling.
Chapter 3

3. KLM transducer model

As described in the previous chapter, two parallel line of transducer development are to be followed in this project, to improve the chance of a successful outcome. Each of the two transducer designs is based on a piezoelectric element that is commercially available in single crystal form:

(a) 36° Y-cut LiNbO₃ has a high coupling factor $k_t$ of 48%. Its Curie temperature $T_c$ of 1150 °C is a positive indicator for trouble-free long-term application of this crystal at temperatures of the order of 700-800 °C.

(b) X-cut GaPO₄, on the other hand, exhibits no $T_c$, but rather has a phase transition temperature of 970 °C. Its coupling coefficient $k_t$ is only 15%.

Our objective is to design a backing element for a transducer operating at 2.7-3 MHz center frequency, with a 3 dB bandwidth of 90%. The one-dimensional KLM transducer model is
implemented to estimate the acoustic impedance of the backing element ($Z_b$) to obtain this desired transducer bandwidth. In this model, the thickness of the backing layer is assumed to be far greater than that of the piezoelement. Despite the limitations of a one-dimensional model, it is adequate for this rough calculation. The required inputs to the KLM model include material properties and thickness of the piezoelement, backing layer, the quarter-wave matching layer, and the test specimen.

3.1. Matching layer selection

The matching layer material must withstand high temperatures and be highly resistant to oxidation. It also must be made of a material with CTE close to that of the piezoelement to minimize the interfacial shear stress where the two transducer components are joined together. In this study, the target test material is assumed to be low carbon steel with $Z_T \approx 45$ MRayls. The acoustic impedance of the LiNbO₃ crystal⁴ is $Z_p = 31$ MRayls, whereas for the GaPO₄ crystal⁵ it is $Z_p = 15.8$ MRayls, obtained from the manufacturer. Equation (2.15) then yields an optimal value for the matching layer of $Z_m \approx 37$ and 27 MRayls for LiNbO₃ and GaPO₄, respectively.

36° Y-cut LiNbO₃ has lateral CTEs of 15.5 μm/m°C and 10.3 μm/m°C. Black alumina⁶ with acoustic impedance of 36 MRayls (provided by the manufacturer) was found suitable for this application. The material shows high thermal stability, with CTE equal to 8.5 μm/m°C.

X-cut GaPO₄ single crystal has lateral CTE of 12.8 μm/m°C and 3.7 μm/m°C. Looking for a ceramic with CTE close to the average of those of the piezoelement, mullite ceramic with CTE of

---

⁴ Boston Piezo-Optics Inc., MA, USA  
⁵ Piezocryst Advanced Sensorics GmbH, Austria  
⁶ EBL Products Inc., Connecticut, USA
5.7 μm/m°C was found suitable for this application. Mullite has acoustic impedance of 30 MRayls, close enough to the optimal value of 27 MRayls.

Values of impedances, CTEs and thicknesses of the piezoelement and corresponding matching layers are summarized in Table 3.1. Values are calculated based on the material properties of the piezoelements at 800°C. Mechanical and piezoelectric properties at high temperatures were provided by the manufacturer for GaPO₄ piezoelement. For LiNbO₃ they were extracted from study done by Tomeno and Matsumura [48]. The actual effect of the difference in CTEs on the stability of the bonding layer is investigated in chapter 5.

Table 3.1. Acoustic impedance and CTEs of the matching layers associated with each piezoelement.

<table>
<thead>
<tr>
<th>Piezoelement</th>
<th>LiNbO₃ Transducer</th>
<th>GaPO₄ Transducer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Z (MRayls)</td>
<td>CTEs (μm/m°C)</td>
<td>Thickness (mm)</td>
</tr>
<tr>
<td>Piezoelement</td>
<td>31</td>
<td>15.1, 10.3</td>
</tr>
<tr>
<td>Matching layer</td>
<td>36</td>
<td>8.5</td>
</tr>
</tbody>
</table>

3.2. KLM model results

Adding a backing element to the transducer changes the transducer signal frequency and bandwidth. Using the results of the KLM transducer model presented in chapter 2, various acoustic impedances for backing element are tried to obtain a signal with 2.7-3 MHz center frequency and 3dB bandwidth of 80-90%, when transducer is used as a transmitter.

Fig. 3.1 and 3.2 show the output pressure per unit excitation voltage (transducer transfer function) as a function of excitation frequency for various values of the acoustic impedance of the backing element for transducers with piezoelements of LiNbO₃ and GaPO₄, respectively. In Fig. 3.1, as the backing element acoustic impedance Zₜ is increased from 5 to 35 MRayls, the bandwidth
of output signal increases from 86% to 120%. The increase in the bandwidth comes at the cost of losing a part of energy in the backing element. It can be seen that the increase in the backing element acoustic impedance causes a reduction of 44% in the transducer’s maximum output energy. It also shifts the center frequency from 2.85 MHz when the transducer is air-backed down to 2.78 MHz when \( Z_b = 35 \) MRayls. A value of \( Z_b \) in the range of 20-25 MRayls was found to yield a system with center frequency of 2.85 MHz and desired 3 dB bandwidth of 95%-100%.

In Fig. 3.2, with increase in \( Z_b \) from 3 to 15 MRayls, the bandwidth of the output signal increases from 47% to 84%. This is while the maximum of transducer’s signal amplitude decreases by 50% at 3 MHz. A value of \( Z_b \) in the range of 14-15 MRayls was found to yield a system with center frequency of 3 MHz and desired 3 dB bandwidth of 80-85%. A summary of these results is presented in Tables 3.1 and 3.2.

![Fig. 3.1. Transfer function of transducer vs. frequency for various values of backing impedance \( Z_b \) from KLM model, 36° Y-cut LiNbO\(_3\) crystal (\( Z_p = 31 \) MRayls) with alumina quarter-wave matching layer.](image)
Fig. 3.2. Transfer function of transducer vs. frequency for various values of backing impedance $Z_b$ from KLM model, X-cut GaPO$_4$ crystal ($Z_p=15.8$ MRayls) with mullite quarter-wave matching layer.

Table 3.2. Bandwidth and maximum value of transfer function of test system shown in Fig. 3.1 for various values of $Z_b$ (MRayls) from KLM model, using LiNbO$_3$.

<table>
<thead>
<tr>
<th>$Z_b$ (MRayls)</th>
<th>3dB Bandwidth (%)</th>
<th>Max. value of transfer function (pascal/volt)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>86</td>
<td>0.64</td>
</tr>
<tr>
<td>10</td>
<td>87</td>
<td>0.56</td>
</tr>
<tr>
<td>15</td>
<td>90</td>
<td>0.51</td>
</tr>
<tr>
<td>20</td>
<td>94</td>
<td>0.46</td>
</tr>
<tr>
<td>25</td>
<td>100</td>
<td>0.42</td>
</tr>
<tr>
<td>30</td>
<td>107</td>
<td>0.39</td>
</tr>
<tr>
<td>35</td>
<td>120</td>
<td>0.36</td>
</tr>
</tbody>
</table>

Table 3.3. Bandwidth and maximum value of transfer function of test system shown in Fig. 3.2 for various values of $Z_b$ (MRayls) from KLM model, using GaPo$_4$.

<table>
<thead>
<tr>
<th>$Z_b$ (MRayls)</th>
<th>3dB Bandwidth (%)</th>
<th>Max. value of transfer function (pascal/volt)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>72</td>
<td>0.064</td>
</tr>
<tr>
<td>6</td>
<td>73</td>
<td>0.057</td>
</tr>
<tr>
<td>9</td>
<td>74</td>
<td>0.051</td>
</tr>
<tr>
<td>12</td>
<td>80</td>
<td>0.046</td>
</tr>
<tr>
<td>15</td>
<td>85</td>
<td>0.042</td>
</tr>
</tbody>
</table>
Having found the optimal value for the acoustic impedance of the backing elements for each transducers, the major challenge is to design and manufacture a material to yield the desired values while withstand the high temperatures. The backing element is also expected to exhibit CTEs close to those of the piezoelement to reduce the thermal stresses at the interfaces, therefore to ensure stable bonding to the piezoelement over a wide range of temperature (room temperature to 800°C). This subject is addressed in chapter 4.
Chapter 4

4. Modeling and manufacturing of backing elements made of porous ceramics

Solid materials attenuate ultrasonic wave energy in two ways: absorptive damping (often viscoelastic in origin) and scattering from inhomogeneities such as pores and inclusions. Dense ceramics typically have very low values viscoelastic damping coefficients (corresponding to mechanical quality factor $Q>200 \times 10^6$) [49]. By focusing on the scattering component of ultrasonic attenuation, this project introduces a new application of porous ceramics as the backing element in ultrasonic transducers, operating at temperatures up to 700 – 800 °C. In this section, the new concept of porous ceramics as backing element material for high temperature transducers is presented. Modeling and manufacturing of porous ceramics are described.

In every porous medium, acoustic impedance and attenuation are functions of porosity and pore size. Section 4.1 describes the modeling of ultrasonic wave propagation in composite media
containing a random spatial distribution of spherical inclusions or pores. This model yields the optimal porosity (void volume fraction) and pore size for our application. Material selection and the effect of porosity and pore size on acoustic impedance and attenuation of the transducer backing element are presented in Section 4.2. Manufacturing of porous ceramics with specific target values of porosity and pore size is described in Section 4.3. Section 4.4 focuses on the microstructure of a porous ceramic to find the actual 3D pore size distribution after sintering. In Section 4.5, measurements of phase velocity and attenuation of manufactured porous ceramics are reported and compared with those anticipated from the theoretical model.

4.1. Wave propagation model in porous media

Elastic wave propagation in composite materials containing spherical pores or inclusions is dispersive. The relationships linking these microstructural features to wave scattering, attenuation, and phase velocity as functions of frequency have been studied extensively, and are briefly reviewed here. Much of this material is adopted from the Author’s peer-reviewed article [50].

Lord Rayleigh [51] addressed the problem of scattering from a single sphere, neglecting the effect of multiple scattering between inclusions. Yamakawa [52-53] reviewed scattering results for different types of widely-spaced spherical obstacles including cavities, and calculated the attenuation for waves traveling through a medium containing $N$ spherical obstacles per unit volume. Knopoff [54-55] presented compression and shear wave scattering calculations for a single rigid sphere in an elastic medium. A theoretical model for elastic wave propagation in multilayer porous composites was presented by Sev [56], in which the effects of Rayleigh scattering and geometric dispersion due to the voids were included. Bascom and Cobbald [57] developed a theoretical model to study the backscattered Doppler power caused by the red blood cells. They assumed that attenuation and the effect of multi-scattering can be ignored. Also, the
scatterers size and spacing were considered to be small compared to the wavelength. The effect of cell aggregation, which results into scatterers whose size is larger than wavelength, was found crucial on the validity of the model.

With increasing porosity, it becomes necessary to include the effect of multiple scattering interactions on acoustic behavior of the medium. Foldy [58] first presented an approximate closed-form solution for the wavenumber of a coherent longitudinal wave propagating in the presence of randomly distributed spherical point scatterers. Lax [59-60] used the Effective Field Method (EFM) and introduced an approximate solution by employing the Quasi-Crystalline Approximation (QCA: the effective field is assumed to be the same for all inclusions) while considering the statistical distribution of randomly dispersed particles. This approximate method simplifies the problem of diffraction of the incident wave by many inclusions to the problem of diffraction of the effective field by one inclusion (or void) in an infinite matrix. Twersky [61-67] published a series of papers studying various orders of multiple scattering using several forms of pair correlation functions. Varadan et al presented a more complex model for multiple scattering of waves at higher frequencies for which particle size is of the order of one wavelength [68-78].

For this project featuring wave propagation in composite materials with a random set of spherical voids, we will follow the EFM work of Kanaun et al [79-81], who obtained numerical results of dispersion relationships for a wide range of pore radius-to-wavelength ratios. They combined EFM with QCA [80-81] to address the problems of both longitudinal and shear elastic wave propagation in a medium with spherical inclusions (voids). EFM and QCA are used to simplify the effect of interaction between inclusions and reduce the problem of scattering from many inclusions to scattering from one inclusion. The main assumptions in the model are as follows:
1. Every inclusion (or spherical void) acts as an isolated scatterer embedded in the homogeneous medium. The field that acts on this inclusion is the plane wave that propagates in the effective medium.

2. The mean field that propagates in the porous medium is approximated as the field which propagates in a homogeneous with “effective” properties. This is called the self-consistency hypothesis.

3. All of the inclusions are spherical in shape, with a single size.

The resulting dispersion equation is applicable at all pore radius-to-wavelength ratios, and a wide range of volume fraction of inclusions.

The displacement $u$ associated with a wave of initial amplitude $A$ travelling along the x-axis in the positive direction in a porous medium can be written as:

$$ u = A e^{i(k_{e}x-\omega t)} \quad (4.1) $$

where $\omega$ is circular frequency, and $k_{e}$ is the complex wavenumber ($k_{e}=k_{s}+ik_{\omega}$). Subscript “$s$” represents the static values of the parameters while the dynamic components are indicated by subscript “$\omega$”. Also, in the following, subscripts ”$m_{x}$”, “$p$” and “$e$” represent properties related to the matrix, pores and the effective value of the porous medium, respectively. For a small concentration of spherical voids with diameter $d$ much smaller than wavelength $\lambda$ (Rayleigh region), Sev [56] has shown that static and dynamic components of the wavenumber can be approximated as

$$ k_{s} \approx \frac{\omega}{c_{e}} \quad (4.2) $$
in which $c_e$ is the effective phase velocity of the porous medium, equivalent to

$$c_e = c_{mx} \left( \frac{g}{1-p} \right)^{1/2}$$

where $p$ represents porosity, and $c_{mx}$ is the bulk longitudinal wave speed in the matrix. The function $g$ depends on porosity and Poisson’s ratio $\nu_m$ of the matrix as

$$g = \frac{1 - 2\nu_m}{1 - 2\nu_m + \frac{p}{2}(1 - \nu_m)} - \frac{10p(1 - 2\nu_m)}{7 - 5\nu_m + 2p(4 - 5\nu_m)}$$

The scattering parameter $\varepsilon$ is obtained by solving the problem of scattering from a single inclusion. For a dilute mixture of small random voids with diameter $d$, an explicit expression for $\varepsilon$ is given by:

$$\varepsilon = \frac{pg'd^3c_e}{24c_{mx}^4}$$

where the function $g'$ is defined by Ying and Truell [82] as:

$$g' = \frac{4}{3} + \frac{80 + 120\delta^5}{(4 - 9\delta^2)^2} \times \frac{3\delta^2}{2} + \frac{2\delta^3}{3} + \frac{9\delta^4}{16}, \quad \delta^2 = \frac{2(1 - \nu_m)}{1 - 2\nu_m}$$

For high values of porosity however, the interaction among multiple scatterers is not negligible and consequently this model tends to be less accurate. Only approximate solutions such as EFM are available to address this problem. For the Rayleigh scattering region where the wavelength is far greater than the diameter $d$ of the inclusions, Kanaun et al [79] developed the following approximate relationships:
\[ K_e = K_s - i \left( \frac{k_s d}{2} \right)^3 K_\omega, \quad \mu_e = \mu_s - i \left( \frac{k_s d}{2} \right)^3 \mu_\omega, \quad \rho_e = \rho_s + i \left( \frac{k_s d}{2} \right)^3 \rho_\omega \]  

(4.8)

where \( K_e, \mu_e \) and \( \rho_e \) are effective bulk modulus, shear modulus and density, respectively of the porous medium. Kanaun showed that the static (real-part) and dynamic (imaginary-part) parameters in Eq.4.8 could be evaluated by employing the following approximate relations:

\[ K_e = K_{mx} + p(K_p - K_{mx}) \frac{3K_s + 4\mu_s}{3K_p + 4\mu_s}, \]

\[ \mu_s = \mu_{mx} + p(\mu_p - \mu_{mx}) \frac{5\mu_s(3K_s + 4\mu_s)}{\mu_s(9K_s + 8\mu_s) + 6\mu_p(K_s + 2\mu_s)}, \]  

(4.9)

\[ \rho_s = \rho_{mx} + p(\rho_p - \rho_{mx}) \]

\[ K_\omega = p(K_p - K_{mx}) \frac{3K_s + 4\mu_s}{(3K_p - 4\mu_s)^2} \left( K_p - K_s + 3K\frac{\mu_\omega}{\mu_s} \right), \]

\[ \rho_\omega = p(\rho_p - \rho_{mx}) \frac{9\rho_p - \rho_s}{9\rho_s} \left( 1 + \frac{2}{\eta_s^3} \right) \]

\[ \mu_\omega = \frac{10p}{\Delta \omega} \eta_s^2 (\mu_p - \mu_{mx})(\mu_p - \mu_s) \left( 2 + \frac{3}{\eta_s^2} \right), \quad \eta_s^2 = \frac{3\mu_s}{3K_s + 4\mu_s} \]

\[ \Delta \omega = \mu_s \left[ 15 + \frac{2(3 + 2\eta_s^2)}{\mu_s}(\mu_p - \mu_s)^2 - 30p(\mu_p - \mu_{mx}) \frac{\mu}{\mu_s} (3 + 2\eta_s^2) \right] \]

(4.10)

It is noted that Eqs. 4.9 are implicit in terms of \( K_s \) and \( \mu_s \), such they must be solved numerically. \( K_p, \mu_p \) and \( \rho_p \) are bulk modulus, shear modulus and density of the inclusions or pores, while those same parameters with subscript “\( mx \)” refer to the properties of the matrix material. For our case of spherical pores, elastic constants and density of the inclusions are set to zero. The effective wave number \( k_e \) of the porous medium is given by:
\[
k_e = k_s + ik_\omega, \quad k_s = \frac{\omega}{c_{p,e}}, \quad c_e = \sqrt{\frac{3K_s + 4\mu_s}{3\rho_s}} \quad \rightarrow \quad Z_e \approx \rho_s c_e \quad (4.11a)
\]

\[
\alpha_e = \frac{1}{16d^3} p(k_s d)^4 \left( \frac{3K_\omega + 4\mu_\omega}{3K_s + 4\mu_s} \frac{\rho_\omega}{\rho_s} \right) \quad (4.11b)
\]

From Eqs. 4.9 and 4.11a, it can be concluded that in the Rayleigh scattering region, the phase velocity \( c_e \) is frequency-independent if the matrix material has zero attenuation. Also, when \( k_s d^3 \ll 1 \) in Eq.4.8, the dynamic component of density is relatively small compared to the real part [79], such that the effective density \( \rho_e \) of the porous medium approaches its static value \( \rho_s \). Consequently, the effective acoustic impedance \( Z_e \) of the porous medium is independent of frequency within the Rayleigh scattering regime according to the Kanun model. This assumption is later verified by comparing the values of acoustic impedance obtained from experiment and those from the model. The effective acoustic attenuation \( \alpha_e \) (Eq.4.11b) is a fourth-order function of frequency and proportional to the third order of pore diameter \( d \).

The theoretical predictions of this model were compared with experimental results for the case of steel spherical inclusions dispersed in a polymethylmethacrylate (PMMA) matrix [79]. Close agreement between the Kanaun numerical results and the experimental values given in Ref. [83] demonstrated the validity of the Kanaun model.

4.2. Material selection and effect of porosity and pore size

The backing element needs to maintain its mechanical properties at temperatures up to 800 °C, and be resilient against thermal shock. In addition, interfacial stress must be minimized as the transducer temperature is raised to this elevated level. To this end, the CTE of the backing element must be as close as possible to those of the piezoelement.
The lateral CTEs for 36° Y-cut LiNbO₃ are $a_{22}=15.5 \, \mu m/m^\circ C$ and $a_{33}=10.2 \, \mu m/m^\circ C$, as provided by the manufacturer’s data sheets. This leads to our choice of isotropic 3mol% Yttria-stabilized zirconia (3% YSZ) for the ceramic matrix material in the backing layer.

X-cut GaPO₄ single crystal has lateral CTEs of $a_{22}=12.8 \, \mu m/m^\circ C$ and $a_{33}=3.7 \, \mu m/m^\circ C$, per the manufacturer’s datasheet. Mullite ceramic has a CTE of 5.7 $\mu m/m^\circ C$ which is close to the average CTE of GaPO₄, and was therefore selected for the matrix material in the backing layer of this piezoelement.

In every backing element, two ultrasonic propagation parameters are of importance: the acoustic impedance and attenuation. In chapter 3, optimal values of acoustic impedance were found to be 20-25 MRayls and 14-15 MRayls for LiNbO₃ and GaPO₄ piezoelements, respectively. The backing element must have a sufficiently high attenuation coefficient over the operating frequency range of the transducer; this will ensure that any wave launched from the piezoelement into the backing will not reflect back to the piezoelement and interfere with the main signal. In consideration of that objective, the amplitude $A''$ of the wave reflected from the rear wall of the backing layer returning to the backing-piezoelement interface should be at least 40 dB below the amplitude $A'$ of the wave entering the backing layer (Fig. 4.1). For a backing layer of thickness $h_b$ and attenuation coefficient $\alpha$ (dB/mm), this condition can be written as

$$20 \ln \frac{A''}{A'} = -2h_b\alpha < -40 \text{dB}$$  \hspace{1cm} (4.12)

---

7 BOSTON PIEZO-OPTICS INC., MA, USA
8 Tosoh, Japan
9 Piezocryst Advanced Sensorsics GmbH
10 KCM CO., Japan
Geometrical constraints imposed by our industrial partner limit the thickness \( h_b \) of the backing element to a maximum of approximately 20 mm. This leads to a minimum value of \(~1\) dB/mm for ultrasonic attenuation within the useful transducer bandwidth. The values of acoustic impedance corresponding to a series of values of porosity and pore diameter were calculated from Eq. 4.11a. Estimates of the attenuation values can then be determined for any ultrasonic frequency from Eq. 4.11b. These calculations were limited to the center frequency of 3 MHz for the desired ultrasonic transducer, to obtain rough estimates of the optimal porosity and pore size for the backing material to yield an attenuation coefficient of 1 dB/mm; experimental measurements over the entire transducer bandwidth will be performed on manufactured samples to determine if any final adjustments are required (Section 4.4).

Fig. 4.2 shows the effect of fractional porosity \( p \) of the porous zirconia backing on its acoustic impedance \( Z_b \), as given by Eq. 4.11a. The graph indicates that a porosity value of approximately 25\% would yield the target acoustic impedance of \( Z_b = 20-25 \) MRayls.

For the transducers based on a GaPO\(_4\) piezoelement, Fig. 4.3 shows the effect of fractional porosity \( p \) of the porous mullite backing on its acoustic impedance \( Z_b \), as given by Eq. 4.11a. These
results show that a porosity value of slightly less than 30% would yield the target acoustic impedance of $Z_b = 14-15$ MRayls.

Fig. 4.2. Dependence of acoustic impedance $Z$ on porosity $p$ in porous zirconia in the Rayleigh scattering region per Eq. 4.11a.

Fig. 4.3. Dependence of acoustic impedance $Z$ on porosity $p$ in porous mullite in the Rayleigh scattering region per Eq. 4.11a.

The effect of pore size on acoustic attenuation of the porous YSZ backing element, as predicted by Eq. 4.11b, is shown in Fig. 4.4 for a porosity value of 25%. It can be concluded that for the
desired porosity value of $p \approx 25\%$, a pore diameter of about 200 μm provides the required minimum attenuation of 1 dB/mm at the desired central transducer frequency of 3 MHz. In the case of porous mullite with 30% porosity, Fig. 4.5 shows that a pore diameter of slightly under 400μm provides the required minimum attenuation of 1 dB/mm at 3 MHz. Further information on the attenuation over the entire bandwidth (1.5-4.5 MHz) of the transducer is given in the experimental Section 4.4.

Fig. 4.4. Acoustic attenuation ($\alpha_e$) vs. frequency for different values of pore diameter in porous YSZ with 25% porosity per Eq. 4.11b.
Fig. 4.5. Acoustic attenuation ($\alpha$) vs. frequency for different values of pore diameter in porous mullite with 30% porosity per Eq. 4.11b.

Table 4.1 summarizes the required final values of pore diameter, porosity and the resultant acoustic impedance of porous ceramics obtained from the acoustic model.

Table 4.1. Results of acoustic model: Required pore diameter, porosity and acoustic impedance values for the transducer backing materials.

<table>
<thead>
<tr>
<th>Ceramic matrix</th>
<th>Piezoelement</th>
<th>Porosity%</th>
<th>Pore diameter ($\mu$m)</th>
<th>Acoustic impedance (MRayls)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3% YSZ</td>
<td>LinbO$_3$</td>
<td>25</td>
<td>200</td>
<td>24</td>
</tr>
<tr>
<td>Mullite</td>
<td>GaPO$_4$</td>
<td>30</td>
<td>400</td>
<td>15</td>
</tr>
</tbody>
</table>

4.3. Manufacture of porous ceramics

To manufacture the ceramic matrix of the backing element, a conventional die-pressing technique and sintering can be used. To induce pores in the ceramic body, several possible processing routes are identified as: 1) partial sintering, 2) sacrificial fugitives, 3) replica templates, and 4) direct foaming [84]. The sacrificial fugitive technique was recognised as the best suited to induce semi-spherical pores in the ceramic matrix, as it has the advantage of better control over
pore shape and size [85]. In this method, raw ceramic powder is mixed with a second phase material called a sacrificial fugitive that serves as the pore forming agent; a narrow size distribution of pores can be obtained by using a pore forming agent with a single size of sacrificial fugitive particles. Pore forming particles are burned-out during the sintering process, leaving behind closed (i.e. unconnected) pores. The ceramic matrix is then fully sintered, such that the porosity of the final manufactured material is due solely to the spherical voids left by the consumed polymer particles. Therefore, the final porosity value can be fully controlled through adjustment of the amount of polymer agent mixed with the ceramic powder.

4.3.1. Literature review

Various kinds of fugitive agents have been used for manufacturing yttria-stabilized zirconia (YSZ) with spherical pores. For example, porous YSZ with porosity values ranging from 28.9% up to 53% was produced by using starch mixed with ceramic powder [86]. Porosity of the sintered samples was controlled by the volume fraction of added starch, as well as by the sintering temperature. Porous YSZ ceramics were also fabricated using PMMA microspheres as the pore-forming agent [87]; this yielded a very narrow distribution of pore sizes, high compressive strength and excellent thermal insulating properties. Boaro et al [88] used graphite, PMMA and polyethylene (PE) as pore-forming agents in YSZ. They noted that PMMA decomposed to methyl methacrylate at temperatures between 500 K and 700 K, and graphite combusted at temperatures above 900 K; in both cases, gaseous inclusions were left inside the ceramic matrix.

The sintering behavior and mechanical properties of mullite-alumina composites were studied in [89]. Final density varies directly with forming pressure but remains almost constant at pressures over 200 MPa. Density, mechanical hardness and fracture toughness of the mullite increases with increasing $\text{Al}_2\text{O}_3$ concentration of the composite. The effect of calcination temperature and
Al₂O₃/SiO₂ ratio were investigated [90]: samples were sintered in air at 1700 °C for 4 hrs. Homogeneous and dense microstructures were obtained from high Al₂O₃/SiO₂ ratio powders with calcination at temperatures between 600 °C and 1100 °C.

Porous mullite with 69% interconnected porosity was fabricated by freeze-casting and sintering alumina and silica nanoparticles [91]. Flexural strength was improved by increasing the sintering temperature. Fly ash was used as a pore forming agent to fabricate porous mullite [92]. By sintering at 1200 °C, high porosity (48%-72%) samples with low thermal conductivity were obtained.

Sintering a porous ceramic will lead to dimensional and density changes, such that a trial-and-error approach is needed to achieve a specified final porosity and density. In this study, ceramic powder is mixed with PE polymer microspheres as the pore forming agent at various weight ratios and particle sizes, and then each sample is pressed and sintered. Scanning electron microscopy (SEM) can then be used to investigate the sample integrity and final pore size distribution. Measurements of the density of each sample will yield the porosity.

4.3.2. Manufacturing of porous zirconia

A commercial 3mol% YSZ powder\(^\text{11}\) was used for the matrix material. Clear PE spheres\(^\text{12}\) were added to the ceramic powder as the pore forming agent. Each polymer-ceramic mixture was stirred for one hour using a magnetic stirrer to ensure a smooth, uniform mixture of ceramic and polymer. (Later evidence that both the YSZ and pore former powders were well-dispersed came from the uniformly random distribution of the pores in the final ceramic, as revealed in SEM images.)

\(\text{11\ Tosoh Co. Ltd., Japan}\)
\(\text{12\ Cosphoric LLC, Santa Barbara, CA}\)
To obtain various porosity values, several mixtures with different polymer/ceramic weight ratios were prepared (Table 4.2). Nominal density is based on the ratio of the polymer volume to the total volume of samples. However, due to shrinkage in the samples during sintering, the actual final porosities are less than the nominal values.

Samples were first uni-axially die-pressed at 10 MPa to form cylindrical green compacts (Φ≈30mm) and then iso-statically pressed to the final compaction pressure of 70 MPa proposed by the YSZ manufacturer. After compaction, the green samples were heated and then held at a specific temperature (sintering temperature) for a sufficient amount of time to let the matrix material densify completely. For the YSZ used here, the sintering temperature and dwell time were 1500 °C and 2 hours, respectively, as specified by the manufacturer. Careful control of the heating and cooling rates is required to prevent the formation of defects (mainly cracks). In particular, the heating time should be long enough to let the polymer particles to burn completely, and to allow the combustion gases to escape from the ceramic matrix, before the sample is fully sintered. A series of various heating and cooling rates were tested for this purpose. The following values were found suitable: heating at a rate of 2 °C/min, then cooling down at a rate of 4 °C/min to minimize thermal stresses and inhibit formation of cracks.

The weights and dimensions of samples were measured both before and after sintering to determine porosity and linear shrinkage according to Eq. 4.13:

\[
\text{Linear shrinkage} = \left( \frac{l_g - l_{si}}{l_g} \right) \times 100
\]  

(4.13)

where \(l_g\) and \(l_{si}\) are characteristic dimensions of the green and sintered samples, respectively, and can refer to either thicknesses or diameter (Table 4.2). The bulk densities of the sintered pieces
were then calculated from the final dimensions and weights. To find porosity $p$, the 0%-porosity density value of $\rho_0 = 6.05 \text{ g/cm}^3$ given by the ceramic powder manufacturer was taken as a baseline reference. (It is noted that density measurements of our sintered samples manufactured with no polymer added yielded density values of $\rho_0 = 6.07 \text{ g/cm}^3$; this is within experimental measurement uncertainty of the manufacturer’s specified value.) Porosity values of our sintered samples were defined according to:

$$\% \text{ Porosity} = \left(\frac{\rho_0 - \rho_{si}}{\rho_0}\right) \times 100$$

(4.14)

in which $\rho_{si}$ represents the density of the sintered porous sample.

Table 4.2 lists the amount of YSZ ceramic and polymer materials used to make samples with 0% (fully dense), 20% and 25% porosity. Geometrical dimensions such as diameter and thickness of the samples before and after sintering are provided. Using Eqs. 4.13 and 4.14, radial and thickness shrinkages as well as final porosity are calculated and listed in the Table.
Table 4.2. Physical properties of dense and porous YSZ samples before and after sintering.

<table>
<thead>
<tr>
<th>Nominal porosity (%)</th>
<th>0 (Dense)</th>
<th>20</th>
<th>25</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ceramic mass (g)</td>
<td>15±0.01</td>
<td>15±0.01</td>
</tr>
<tr>
<td>Before Sintering</td>
<td>Polymer mass (g)</td>
<td>0</td>
<td>1.36±0.01</td>
</tr>
<tr>
<td></td>
<td>Diameter (mm)</td>
<td>30±0.02</td>
<td>30±0.02</td>
</tr>
<tr>
<td></td>
<td>Thickness(mm)</td>
<td>7.58±0.02</td>
<td>9.6±0.02</td>
</tr>
<tr>
<td>After Sintering</td>
<td>Total mass (g)</td>
<td>14.38±0.01</td>
<td>14.44±0.01</td>
</tr>
<tr>
<td></td>
<td>Diameter (mm)</td>
<td>22.73±0.02</td>
<td>22.59±0.02</td>
</tr>
<tr>
<td></td>
<td>Thickness (mm)</td>
<td>5.83±0.02</td>
<td>7.44±0.02</td>
</tr>
<tr>
<td></td>
<td>Diameter shrinkage%</td>
<td>24</td>
<td>24.7</td>
</tr>
<tr>
<td></td>
<td>Thickness shrinkage%</td>
<td>23.1</td>
<td>22.9</td>
</tr>
<tr>
<td></td>
<td>Density(g/cm³)</td>
<td>6.07</td>
<td>4.83</td>
</tr>
<tr>
<td></td>
<td>Final porosity %</td>
<td>0</td>
<td>20.4</td>
</tr>
</tbody>
</table>

4.3.3. Manufacturing of porous mullite

Unlike YSZ powder which was ready-to-press, mullite powder was received in a pure state, with no binder or dispersant added. The average diameter of the mullite powder particles was 1-2 µm (specification provided by the manufacturer.) The particles were in general not round and had sharp corners. This could cause a large amount of friction between the powder and the die during pressing, which results in a non-uniform compaction along the sample in the die. In addition, the sharp edges do not let the particles roll over each other to fill in gaps during pressing. This would seriously compromise the quality of the final product if no remedial action were taken.

To address this issue, several pre-processes steps were carried out on the raw powders to make it ready-to-press. First, a 40% powder-to-water volume ratio suspension of the mullite was prepared. The suspension was ball-milled for 24 hours. Next, 0.4-0.5 cc of DOLAPIX CE64\textsuperscript{13} dispersant was added.

\textsuperscript{13} Zschimmer & Schwarz Group, Germany
added to the suspension to prevent the particles from settling and forming a sediment. A 2% weight ratio of polyvinyl alcohol (PVA) solution was also prepared and added to the suspension to yield a ratio of 100 g of ceramic powder to 2 g of PVA solution. The PVA acts as a binding agent to keep the particles together during pressing.

The final solution was then ball-milled for 10 hours so that the binder and dispersant would cover all of the ceramic particles. To remove the water, the solution was heated at 90 °C under forced convection conditions to expedite the vaporizing process until all of the moisture was removed. The dried material was then screened through a 200 mesh (74 µm) sieve to obtain agglomerates of particles with average size of 60-80 µm.

Following the same procedure as described in the previous section, the processed mullite powder\textsuperscript{14} was mixed with PE polymer microspheres as the pore forming agent. Three nominal porosities of 0% (fully dense), 25% and 30% were considered. The density of a fully dense sample was taken as $\rho_0 = 3.2$ g/cm$^3$, as specified in data sheets provided by the powder manufacturer. Our experimental measurements on non-porous samples yielded density values of 3.17 g/cm$^3$, 0.9% less than the value specified by the manufacturer. Table 4.3 shows the density and geometrical dimensions of the samples before and after sintering. Linear shrinkage and porosities are calculated using Eqs. 4.13 and 4.14.

\textsuperscript{14} KCM CO., Japan
Mullite-PE mixtures were first uni-axially die-pressed at 10 MPa to form cylindrical green compacts (Φ≈30mm) and then iso-statically pressed at 200 MPa. Sintering was performed at 1700 °C for four hours. The heating rate was 2 °C/min up to 400 °C to let the polymer particles burn completely and the fumes escape from the ceramic matrix, and then increased to 4 °C/min until a final sintering temperature of 1700 °C was reached. After sintering for 4 hours, samples were cooled down at a rate of 4 °C/min to obtain crack-free samples. Analogous to the development of the YSZ sintering routine, the mullite heating and cooling rates were experimentally optimized to yield crack-free samples.
4.4. Microstructure and pore size distribution

4.4.1. Microstructure

Any defect in a transducer’s ceramic backing element could eventually lead to high thermal stresses and possible in-service failure of the part. Therefore, several cross-sections of sintered samples were inspected by surface electron microscopy (SEM) to determine whether any defects could be observed.

Ceramic samples were first cut using a precision diamond saw (Buehler, IsoMet™ 5000 Linear Precision Saw). They were then moved to a fully automated polishing machine (Buehler, EcoMet/AutoMet 250) (Fig.4.7). The machine provides control over the rotational speed of the polishing paper (base) and sample holder (head), direction of rotation, and the polishing pressure. As recommended by the manufacturer\textsuperscript{15}, the following sequence was used to polish the sample cross sections:

1. Polish with 45-µm diamond paper, at a rotational speed of 240-300 RPM and 60 RPM for head and base, respectively. Polishing was continued until a plane surface was obtained. Head and base rotated in the same direction, at an applied load of 36 Newtons.

2. Polish with 15 µm diamond paper, at a rotational speed of 150-200 RPM for 6 minutes. Head and base rotated in opposite directions, at an applied load of 27 Newtons. The rotational speed of the head remained unchanged.

3. Polish with 6 µm diamond paper, at a rotational speed of 120-180 RPM, for 4 minutes. Head and base rotated in the same direction, at an applied load of 27 Newtons.

\textsuperscript{15} Buehler, An ITW Company, IL, USA
4. Polish with 3 µm diamond paper, at a rotational speed of 100-150 RPM for 4 minutes. Head and base rotated in opposite directions, at an applied load of 27 Newtons.

Fig. 4.6. Buehler EcoMet/AutoMet 250 polishing machine.

Fig. 4.6a-d shows cross sectional SEM images of porous YSZ samples with 20% and 25% porosities. One can observe the ceramic inside walls of the empty circular recessions on the surface. As expected, no polymer solid particles remained. No significant defects are visible in the more detailed images (Fig 4.6b and 4.6d) The nominal polymer particle diameters used for samples with 20% and 25% porosity were 106-125 µm and 150-180 µm, respectively, as quoted by the polymer supplier. Therefore, the pores in Fig.4.6d are on average larger than those in Fig. 4.6b.

Similarly, Fig. 4.7 shows cross sectional SEM images of the mullite samples with 25% and 30% nominal porosity. The polymer particle diameters used in the two samples were 212-250 µm, as specified by the supplier. No evidence was found of any residual solid polymer in the sectioned pores. In addition, the SEM images indicate that our final choices of heating rate, cooling rate and sintering temperature do not generate any visible cracks at pore walls nor in the ceramic matrix (Fig. 4.7b and 4.7d).
Fig. 4.7. SEM images of sectioned porous YSZ samples: a) Pore distribution in sample with nominal 20% porosity b) Spherical pore shape observed in sample with nominal 20% porosity c) Pore distribution in sample with nominal 25% porosity d) Spherical pore shape observed in sample with nominal 25% porosity.
Fig. 4.8. SEM images of sectioned porous mullite samples: a) Pore distribution in sample with nominal 25% porosity b) Spherical pore shape observed in sample with nominal 25% porosity c) Pore distribution in sample with nominal 30% porosity d) Spherical pore shape observed in sample with nominal 30% porosity.

4.4.2. Pore size distribution

Due to the specimen shrinkage during the sintering process, the final pore size is less than the size of each corresponding pore-forming polymer particle. To find the actual pore size distribution, multiple SEM images were made of the surfaces of sectioned samples. These images were first
processed to find the apparent two-dimensional size distribution of the exposed circular sections at the cut surface. Employing the statistical approach described in [93], this 2D sectioned size distribution was then mapped onto the actual 3D spherical pore size distribution. The manufacturer’s data indicated that the original polymer particle size distribution had an approximately Gaussian profile; it was therefore assumed as part of the optimization routine that the final 3D pore size distribution in the sintered ceramic would also follow an approximate Gaussian profile.

Fig. 4.8a shows the observed 2D size distribution on the sectioned surface of the porous YSZ sample obtained using particle particles with nominal diameter range of 106-125 μm claimed by the manufacturer. To find the final 3D pore diameter distribution in the ceramic, an iterative technique is used: First, a preliminary guess of the 3D pore Gaussian distribution is fed into the statistical model [93] based on the nominal particle size distribution provided by the manufacturer. Next, the expected distribution of pore diameters visible on the 2D sectioned surface is found based on the postulated 3D size distribution. Then, an optimization algorithm was utilized to update the estimated 3D pore size distribution; the goal is to minimize the difference between the 2D distribution observed on the sectioned surface and the one predicted by the model.

In the optimization algorithm, two gamma distribution functions were fitted onto the experimentally observed 2D size distribution and the 2D distribution predicted by the model. This provides a tool to compare the two distributions and find the best fit. The gamma probability distribution function (PDF) is defined as

$$f(x; \gamma, \beta) = \frac{-1^{\beta-1}}{\Gamma(\beta)\gamma^\beta} x^{\beta-1} e^{-\frac{x}{\gamma}}$$  (4.15)
in which $\gamma$ and $\beta$ are the scale parameter and shape parameter, respectively. Various 3D pore Gaussian distributions were fed into the model so that the difference between parameters $\gamma$ and $\beta$ in the actual and predicted 2D size distributions reached a level of less than 10%.

After the optimization routine has converged, Fig. 4.8b shows the 3D pore size distribution predicted by the model and Fig. 4.8c shows the corresponding theoretical 2D section distribution on a sectioned surface, if pore size distribution were as it is shown in Fig. 4.8b. The mean pore diameter was found to be 96 $\mu$m. Note the good agreement between the observed 2D distribution of pore diameters on the sectioned ceramic surface in fig. 4.8(a), and the distribution predicted by the model in Fig. 4.8 (c). Fig. 4.9 shows the two gamma functions fitted onto the 2D diameter distributions Fig. 4.8a and 4.8c. The final deviations between the distribution parameters $\gamma$ and $\beta$ are 9.4% and 5%, respectively. On that basis, the postulated 3D spherical pore size distribution of Fig. 4.8b represents the actual 3D pore distribution in the porous sample.

Fig. 4.9. Pore diameter distribution generated in porous YSZ by polymer beads with a nominal diameter range of 106-125 $\mu$m: a) Experimental size distribution of pores diameter visible on a 2-dimensional sectioned surface; b) 3D pore diameter (Gaussian) distribution as predicted by numerical model, corresponding to data of Fig. 4.8(a); c) Resultant 2D section distribution of visible pore diameters obtained from the assumed 3D pore distribution.
Fig. 4.10. Gamma distribution functions fitted onto actual (Fig.4.8a) and predicted (Fig.4.8c) 2D size distributions of pore diameters on sectioned surface.

Fig. 4.11. 3D pore diameter distribution in porous YSZ generated by polymer beads with a nominal diameter range of 150-180 μm.

Fig. 4.12. 3D pore diameter distribution in porous YSZ generated by polymer beads with a nominal diameter range of 180-212 μm.
A similar approach was employed to find the 3D pore size distribution in porous YSZ samples made from polymer powder with nominal particle diameters of 150-180 μm and 180-212 μm. In these two cases, the final mean pore diameters were found to be 130 μm (Fig. 4.10) and 160 μm (Fig. 4.11), respectively.

Employing the same statistical approach as used for porous YSZ, the actual 3D pore size distributions of porous mullite samples were extracted from their 2D size distributions. Fig. 4.12 and 4.13 depict the 3D pore distributions obtained from SEM cross-sectional images of porous mullite samples made using a polymer with nominal particle diameter ranges of 212-250 μm and 250-300 μm, respectively. The mean pore diameters were found to be 200 μm and 230 μm, respectively.

![Fig. 4.13. 3D pore diameter (Gaussian) distribution of porous mullite sample as predicted by numerical model generated by polymer beads with a nominal diameter range of 212-250 μm.](image1)

![Fig. 4.14. 3D pore diameter (Gaussian) distribution of porous mullite sample as predicted by numerical model generated by polymer beads with a nominal diameter range of 250-300 μm.](image2)
4.5. Acoustic measurements

Immersion ultrasonic measurements were made to determine the ultrasonic phase velocity and attenuation of each porous sample. A pitch-catch immersion technique was employed (Fig. 4.14). In this method, two signals are captured: one signal with the sample placed in the path of the transmitted beam ($F_s$), and one signal with the sample absent ($F_w$). The phase velocity $c_p(f)$ of the specimen as a function of frequency $f$ can then be obtained from the phase spectra of the two transmitted pulses [94]:

$$\frac{1}{c_p(f)} = \frac{\theta_s(f) - \theta_w(f)}{2\pi f \cdot th} + \frac{1}{c_w}$$  \hspace{1cm} (4.16)

where $c_w$ is the wave speed in water, $\theta_s(f)$ and $\theta_w(f)$ are the unwrapped phase spectra of $F_s(t)$ and $F_w(t)$, respectively, and $th$ is the thickness of the specimen. The result of the measurement can verify whether the phase velocity stays constant in the target frequency range, as anticipated from the model.

Fig. 4.15. Immersion ultrasonic signals captured for two cases: sample present, and sample absent.
The ultrasonic attenuation can theoretically be found from

\[
\alpha(f) = -20 \frac{\log_{10} A_s(f) - \log_{10} A_w(f)}{th}
\]  

(4.17)

where \( \alpha(f) \) is the acoustic attenuation in dB/mm, and \( A_s(f) \) and \( A_w(f) \) are the amplitude spectra of \( F_s(t) \) and \( F_w(t) \), respectively. In practice, because of the finite size of the transducer, diffraction effects cause the beam to spread out and lead to errors in the direct application of Eq. 4.17. To compensate for the effects of diffraction, the technique described by He [94] is utilized here: First, the Fresnel parameter for the water layer between the transmitter and sample is defined as

\[
S_{w1} = \frac{l_{w1} \lambda_w}{a^2}
\]  

(4.18)

In Eq.4.18, \( l_{w1} \) is the distance between the transmitter and sample, \( \lambda_w \) represents the wavelength in water, and \( a \) is the radius of both transmitter and receiver transducers. Similarly, the Fresnel parameters for the sample layer (\( S_s \)) and the water layer between the sample and the receiver (\( S_{w2} \)) can be defined as

\[
S_s = \frac{th \cdot \lambda_s}{a^2}
\]  

(4.19)

\[
S_{w2} = \frac{l_{w2} \lambda_w}{a^2} z
\]  

(4.20)

where \( \lambda_s \) represents the wavelength in the sample and \( l_{w2} \) is the distance between the sample and receiver.

To compensate for the effect of diffraction in the attenuation measurement, the separation distance between transmitter and receiver for the water-only case is adjusted to a value \( l_w^* \) such that \( S_w^* = S_{w1} + S_s + S_{w2} \), where \( S_w^* = l_w^* \lambda_w / a^2 \), yielding

\[
l_w^* = \frac{(l_w - th) \lambda_w + th \cdot \lambda_s}{\lambda_w}
\]  

(4.21)
in which

\[ l_w = l_{w1} + th + l_{w2} \]  \hspace{1cm} (4.22)

Now, Eq.4.17 can be modified as

\[ \alpha(f) = -20 \frac{\log_{10} A_p(f) - \log_{10} A_{w'}(f)}{th} \]  \hspace{1cm} (4.23)

in which \( A_{w'}(f) \) is the amplitude spectrum of the signal taken for the water-only case when transmitter and receiver are separated by a distance \( l_{w} \), as shown in Fig. 4.14.

4.5.1. Acoustic properties of porous YSZ

Immersion measurements of phase velocity and attenuation were carried out on porous YSZ samples with porosities of 20% and 25%, and three different pore size distributions. The acoustic impedance for each sample was then obtained by multiplying the measured phase velocity by the sample density. Given that in our frequency range, phase velocity is approximately frequency-independent, acoustic impedance also shows the same behaviour.

The acoustic model suggests that a porosity of 25% for porous YSZ will yield the target acoustic impedance. However, since the model is only approximate, two porosities were chosen for initial trials to manufacture samples: 20% and 25%.

The first trial YSZ sample was made with 20% nominal porosity and average pore diameter of 96 μm (Fig 4.8b). Measurements yielded an average acoustic impedance of 29.5 MRayls and attenuation of 0.5 dB/mm at 3 MHz. To reduce the acoustic impedance to the optimal value of 20-25 MRayls, further samples were made with nominal 25% porosity. In addition, larger pores with average diameter of 130 μm (Fig 4.10) and 160 μm (Fig 4.11) were induced in these samples to increase acoustic attenuation to the minimum required value of 1 dB/mm.
Measurements on this new set of samples revealed that the acoustic impedance had diminished to 22.76 MRayls and 23.64 MRayls for samples with average pore diameters of approximately 130 μm and 160 μm, respectively. Therefore, it can be concluded that a porosity value of 25% in YSZ yields an acoustic impedance within the desired range (20-25 MRayls). As expected from the theoretical model, experimental measurements showed that pore size has very little effect on acoustic impedance.

Results of attenuation measurements on these samples are shown in Fig.4.15. Two pairs of immersion transducers with 2.25 MHz and 3.25 MHz nominal center frequency were used. The presence of two lines for each pore diameter in Fig.4.15 corresponds to the results obtained from each of the two immersion transducers. As expected, attenuation increases rapidly with pore size in the Rayleigh scattering range. For the sample with nominal 25% porosity and average pore diameter of 160 μm, the minimum required attenuation of 1 dB/mm was obtained over the entire transducer bandwidth of 1.5-4.5 MHz. Even higher levels of attenuation are achievable by introducing larger pores; however, our experience shows that this increases the chance of having cracks in the samples that would imperil specimen integrity at high temperatures.

There is a noticeable difference between values of attenuation anticipated from the acoustic model (Fig.4.4) and those obtained experimentally. It should be noted that the Kanaun model for porous media includes only the attenuation effect of wave scattering from dispersed spherical inclusions of a single diameter, and neglects attenuation due to other factors such as the material’s intrinsic hysteresis. Acoustic measurements on non-porous YSZ samples revealed the significant magnitude of the hysteresis attenuation (Fig.4.16). This accounts for part of the discrepancy between the model and experiment.
SEM images (Fig.4.6d) show that the pores are sufficiently close together that they are not acting as completely independent scatterers – the effects of multiple scattering can therefore be significant. This effect was neglected in the acoustic model which assumes that each pore is an isolated scatterer in an infinite medium. SEM images also revealed that in the manufactured samples, there is a distribution of pore diameters (Fig. 4.8, 4.10 and 4.11), while the acoustic model only accounts for spherical pores with a single diameter. Given that attenuation in the Rayleigh range is roughly proportional to the pore diameter cubed, the pores with larger than average diameter would strongly increase the wave attenuation. This effect was amplified further by some agglomeration of pores as seen in Fig. 4.6d, thereby forming much larger non-spherical pores that have far higher values of attenuation in this Rayleigh scattering regime.

Fig. 4.16. Experimental results for acoustic attenuation of porous YSZ with nominal 25% porosity for three different pore diameters.
4.5.2. Acoustic properties of porous mullite

Mirroring the procedure described in Section 4.5.1 for YSZ, the phase velocity was measured on porous mullite samples with 25% and 30% nominal porosity; negligible dispersion was observed. The acoustic impedances of the two samples were found to be 17.3 MRayls and 14.5 MRayls (Eq.4.16), respectively. It can be concluded that a nominal porosity value of 30% yields an acoustic impedance within the desired range of 14-15 MRayls.

As observed in the manufactured porous YSZ samples, the acoustic model overestimates the minimum pore diameter to obtain the target attenuation. Therefore, samples with smaller pore diameters were manufactured. Results of the attenuation measurements are shown in Fig.4.17, based on Eq.4.23. Two samples were analyzed, both with 30% porosity, but with two different average pore diameters of 200 μm and 230 μm. As expected, the attenuation increases rapidly with pore size in the Rayleigh scattering range. For the sample with 30% porosity and average pore
diameter of 230 μm, the minimum required attenuation of 1 dB/mm was obtained over the entire transducer bandwidth of 1.5-4.5 MHz.

Fig. 4.18. Effect of pore diameter on acoustic attenuation of porous mullite samples with 30% porosity.

4.6. Concluding remarks

In this chapter, a novel backing material for ultrasonic transducers made of a porous ceramic material was designed and manufactured. Porosity in the ceramic was induced by mixing PE microspheres into the ceramic powder, to act as a pore forming agent. The polymer spheres burn during the sintering process, and leave spherical pores inside the ceramic matrix. The main advantage of the new design over current types of backing elements is their high temperature stability up to 700-800 °C, and thermal expansion compatibility with the piezoelement. By controlling the porosity and pore size in the design of the backing element, optimal values of acoustic impedance and attenuation can be obtained.
A 3mol% YSZ ceramic was chosen as the backing element for transducers using LiNbO₃ as the piezoelectric element. Porous zirconia samples with 20% and 25% porosity and three different pore diameters of 96 µm, 130 µm and 160 µm were manufactured. For use with a GaPO₄ piezoelectric crystal, porous mullite was introduced as backing element. Samples with 25% and 30% porosity and pore diameters of 200 µm and 230 µm were manufactured.

SEM images were taken from cut samples to check for micro-cracks, and to measure the pore size distribution of the final sintered samples. From ultrasonic measurements, it was found that the porous YSZ with 25% porosity and average pore diameter of 130-160 µm yielded an acceptable acoustic impedance of 24 MRayls and minimum attenuation of 1 dB/mm over the entire transducer bandwidth of 1.5-4.5 MHz. Similarly, porous mullite with 30% porosity and average pore diameter of 230 µm yields an acceptable acoustic impedance value of 14 MRayls and adequate attenuation.

The design procedure developed in this chapter can be employed in designing the backing element for ultrasonic transducers covering a wide range of frequencies and bandwidths; a simple one-dimensional model of the system was found to be acceptably accurate in selecting the optimal porosity and pore size in the design process. Next steps will be to assemble the transducer using a high-temperature bonding agent, and characterize prototyped transducer properties and endurance over a wide temperature range.
Chapter 5

5. Bonding of Transducer Primary Components

In this chapter, various methods are investigated for bonding together the three primary internal components of the transducer (backing, piezoelement, matching layer). The bonding mechanism is expected to keep intimate contact at the interface between two adherents, withstanding high temperature and thermal cycling, as well as accommodating the shear stresses caused by any difference in CTEs of the adjacent components. The shapes of the internal transducer components as well as the bonding configuration must be designed to provide access to both piezoelement electrodes. The leading wire used to excite the transducer is designed to withstand the high temperature, thermally-induced stresses, and oxidation.

A prototype transducer will be placed in a furnace and its performance studied as the operating temperature is increased from ambient to the target temperature of 800 °C. Using the transducer in pulse-echo mode, the effect of temperature rise on the amplitude, SNR and bandwidth of the received signal can then be investigated.
5.1. Assembly and Bonding System: Description

The backing and matching layers need to be in intimate contact with the piezoelement over the full range of operating temperatures of up to 700-800 °C. Several high temperature commercial adhesives and brazing alloys are investigated for bonding the transducer layers together. Candidates are expected to be thermally and chemically stable, with CTE close to that of adjacent layers to promote stable bonding with minimal thermal stress. In addition, an electrically conductive bonding agent would facilitate the electrical connections to the piezoelement. For brazing candidates in particular, it is important to consider the chemical composition of the alloy as it reacts with the components. An additional consideration is the capability of the alloy to wet the entire ceramic surface. Given that there is no model anticipating the bonding result, it is a matter of trial-and-error to find a suitable bonding agent. Inquiring from specialist in the companies, more promising candidates were identified and investigated.

Two main routes were chosen: adhesives and brazing alloys. Table 5.1 lists the bonding agents evaluated in this study; only electrically conductive agents were explored at this point, as these would facilitate connection of the “hot” wire and ground to the two surfaces of the piezoelement. TiBraze®Al-665 and TiBraze®800 are sold commercially as a foil, while the other candidates are in the form of paste. The maximum operating temperature is not provided by the manufacturer for the brazing alloys. This is primarily because the chemical composition of the bonding layer is changed by the brazing operation; such changes could produce compounds with melting points that are even higher than the given brazing temperature, depending on both the brazing alloy and the elements being brazed together.
Table 5.1. High temperature bonding agents.

<table>
<thead>
<tr>
<th>Bonding Agent</th>
<th>Adhesives</th>
<th>Brazing alloys</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pyro-Duct 597-A paste(^{16})</td>
<td>TiBraze®Al-665 foil(^{18})</td>
</tr>
<tr>
<td></td>
<td>Duralco(^{TM}) 956 paste(^{17})</td>
<td>TiBraze®800 foil(^{3})</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Active-braze(^{TM}) No.10 paste(^{3})</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Base material</th>
<th>Silver</th>
<th>Titanium</th>
<th>Aluminum</th>
<th>Titanium</th>
<th>Silver</th>
</tr>
</thead>
<tbody>
<tr>
<td>Max service temperature (°C)</td>
<td>950</td>
<td>930</td>
<td>Not provided</td>
<td>Not provided</td>
<td>Not provided</td>
</tr>
<tr>
<td>Brazing temperature (°C)</td>
<td>N/A</td>
<td>N/A</td>
<td>650</td>
<td>850</td>
<td>750</td>
</tr>
</tbody>
</table>

For all the agents shown in Table 5.1, the bonding operation was carried out in two steps: First, one face of the piezoelement was bonded to the ceramic backing. Using each of the bonding agents, LiNbO₃ and GaPO₄ crystals were bonded to porous zirconia and mullite backing elements, respectively. When this step yielded a mechanically strong interface, the other face of the piezoelement was then bonded to the quarter-wave matching layer. The curing process of the adhesives, and the brazing procedures were carried out as per the directions provided by the manufacturers of the bonding agent.

The diameter of the ceramic backings is 15 mm, compared to the piezoelement diameter of 10mm (Fig 5.1). For the case of an electrically conductive bonding agent, the relatively large backing diameter provides access to the piezoelectric electrode after bonding to the non-conductive ceramic backing. The edge of the backing elements is rounded as shown in Fig. 5.1,

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\(^{16}\) Aremco Products Inc.
\(^{17}\) Cotronics Corp.
\(^{18}\) Titanium Brazing, Inc.
such that any ultrasonic wave travelling along the bottom surface of the backing is not reflected back towards the piezoelement.

![Diagram of Piezoelement bonded to porous ceramic backing.](image)

**Fig. 5.1.** Piezoelement bonded to porous ceramic backing.

All three of the internal transducer components must be precisely centered to generate symmetrical longitudinal waves. This symmetry also reduces the chance of exciting laterally propagating modes which could lead to extra pulses in the transmitted wave and a lowered SNR in the received signal. To achieve such transverse symmetry, the assembly jig shown in Fig. 5.2 was designed and constructed to aid in the bonding process. The jig is composed of two main parts held together by three screws spaced 120 degrees apart. Two transducer components are placed in the receptacle with the bonding agent between them. In the first bonding stage, the backing is secured in part 1 while the piezoelement is placed in part 2. In the subsequent bonding step, the quarter-wave matching layer (10mm diameter) is placed in the recess in part 1 and bonded to the piezoelement. To achieve precise concentricity, a tight manufacturing tolerance of 0.001 inches (≈25µm) was specified for manufacturing the assembly jig.
As a first step, the bonding surface on the ceramic backing was polished using diamond polishing papers with the following size sequence: 45 µm, 9 µm and 3 µm. The final average surface roughness on the backing surface was measured\textsuperscript{19} and found to be 0.15 µm. Next, the backing element was washed in an ultrasonic bath to remove any debris from the surface pores and then was dried completely. To remove any contamination such as grease from the backing, matching layer and piezoelement, they were cleaned using acetone.

For those cases where the bonding agent was a paste, the bonding surfaces were first wetted with the paste and placed in the receptacle. To obtain the best bonding strength, the bonding layer must be uniform over the entire surface with constant thickness of 50 µm (as specified by the manufacturer). To achieve this condition, a short piece (5-6 mm long) of nickel alloy wire (Inconel\textsuperscript{®} 625) was placed between the bonding components as a spacer. Then, the bonding components were placed and centered in the receptacle, pressed gently together (Fig. 5.2) so that

\textsuperscript{19} Alpha-step D-120 stylus profiler, KLA-Tencor Corporation, CA, USA
the extra bonding agent seeps out from the interface. Finally, the extra paste was cleaned from the interface.

5.2. Assembly and Bonding System: Experimental trials

First, Duralco™ 956 adhesive was used to bond LiNbO₃ to a porous zirconia backing element. Very poor adhesion was obtained between the adhesive and the piezoelement, regardless of the surface finish on the crystal, such that the bond would not hold even in an unloaded condition. However, this particular adhesive showed good wetting and adhesion to the zirconia backing.

In the next trial, the adhesive Pyro-Duct 597-A yielded a mechanically stable bond between the lithium niobate and porous zirconia backing, even when the bonded system temperature was raised up to 800 °C. However, due to the presence of silver flakes in the paste, ultrasound transmission from the piezoelement to the backing was found to be very poor. As a consequence, the backing element was ineffective as a vibration damper, such that the assembled transducer yielded a very narrow-band signal.

For the alternate transducer configuration featuring a GaPO₄ piezoelement and porous mullite backing, neither of the high temperature adhesives resulted in proper adhesion to either the piezoelement or the backing. After the adhesive curing cycle, no adhesive was found on the piezoelement surface and the adhesive layer on the backing element could be easily peeled off. Roughening of the surfaces was attempted to promote mechanical interlocking of the adhesive with the adhered, but no substantial improvement of interfacial strength was achieved.

As an alternative to adhesives, the high temperature brazing alloys listed in Table 5.1 were tried next as bonding agents. For the case of the Active-braze™ No.10 paste, the same sequence of application steps as used for the adhesive pastes was employed. For the brazing foils listed in
Table 5.1, a disc of 12 mm diameter was cut from the foil and placed between the two transducer components. The entire assembly jig of Fig. 5.1 with foil and adherents was placed in a vacuum furnace and the temperature cycle specified by the manufacturer was followed at the recommended absolute pressure of $3 \times 10^{-5}$ Torr; the near vacuum condition inhibits oxidation of the brazing alloy during high-temperature brazing cycle.

Each of the three brazing alloys listed in Table 5.1 was tried for bonding a LiNbO₃ piezoelement to both a porous zirconia backing and an alumina matching layer. The first candidate was silver-based active-braze™ No.10 paste. Despite trying various heating and cooling rates, large spatial temperature gradients generated during the brazing operation between porous zirconia and the LiNbO₃ crystal led to internal stresses that caused multiple cracks in the piezoelement; these cracks were observed by examining a sectioned interface with a SEM microscope.

The second bonding agent was the TiBraze®800 foil with titanium as the active element. Several trials did not yield successful bonding, as the brazing alloy would not properly wet and bond to the surface of either the piezoelement or the ceramic backing.

The first successful brazing operation was finally achieved with the aluminum-magnesium-based alloy TiBraze®Al-665: no fractures or bonding failures were observed in several trials of brazing the LiNbO₃ to the porous zirconia backing element and alumina matching layer. To investigate further the quality of this bonding scheme, an assembled transducer system was mounted on a flat 10 mm thick carbon steel block using acoustic couplant. Clear back-wall echoes were captured at room temperature, indicating a functionally acceptable bond among the three transducer layers. Further details and an evaluation of the performance of this transducer assembly at high temperatures are presented in the next section.
The brazing agents as used for the lithium niobate transducers were also used to bond GaPO₄ piezoelements to both a porous mullite backing element and a non-porous mullite quarter-wave matching layer. Several trials and various temperature profiles were attempted. In the case of the TiBraze®800 and TiBraze®Al-665 brazing alloys, a good bond was formed between the mullite backing and the brazing alloy, but the GaPO₄ piezoelement would not adhere properly to the brazing foil. The silver-based active-braze™ No.10 paste resulted in multiple cracks in the GaPO₄ crystal (a similar result to that seen for LiNbO₃ crystal). Further investigation may be warranted for other brazing alloys that can achieve an acceptable bond to GaPO₄ crystal.

5.3. Transducer Performance at High Temperature

In this section, the performance of a LiNbO₃-based transducer bonded with TiBraze®Al-665 is studied at elevated temperature; this is the sole type of transducer assembly that was successfully bonded together as described in Section 5.2. Most of the material presented in this section is adopted from the author’s peer-reviewed paper [95].

A transducer assembly (with no encapsulation) was pressed onto a 20 mm thick flat carbon steel block using a plate and two bolts, as shown in Fig. 5.3. A piece of 50 μm thick silver foil was placed between the transducer and the test block to promote good acoustic coupling at high temperatures. The transducer ground wire was attached to the test piece, which was in contact with the bottom surface of the piezoelement through an extension of the brazing foil used between the piezoelement and the matching layer. For the other piezoelement electrode, Inconel 625 wire was used to link the brazing alloy at the piezoelement/backing interface to a high temperature cable, which led back to the ultrasonic pulser/receiver (Olympus 5077PR). The excitation was in the shape of a rectangular pulse with an amplitude of 200 volts and duration of 0.14 μsec.
The transducer setup was placed in a furnace and heated to 800 °C in steps of 100 °C. Back-wall echoes were captured on an oscilloscope (Agilent Technologies DSO-X 2022A)\(^\text{20}\) at each temperature step. It is noted that our maximum target operating temperature of 700-800 °C exceeded the limit recommended by the manufacturer of the brazing alloy; however no damage to the transducer was observed. This may be related to the generation of chemical compounds in the bond layer with very high melting points during the brazing operation.

![Transducer on steel block](image)

Fig. 5.3. Transducer pressed onto a steel block. The ground wire is connected to the steel fixture, while the high-voltage lead is connected at the side of the transducer at the piezoelement/backing interface.

The first back-wall echoes from the steel block are shown in Fig. 5.4a-i, corresponding to transducer operating temperatures ranging from 27 °C up to 800 °C. It can be seen that the signal strength goes down and the SNR ratio deteriorates as the temperature is increased up to 600 °C.

\(^{20}\) Agilent Technologies, US
(Fig. 5.5). This observation is consistent with the decreasing value of the crystal’s electromechanical coupling coefficient with increasing temperature [96]. More significantly, due to the difference in CTEs of the transducer and the fixture, the acoustic coupling between the transducer and test block could be weakened, but this effect is very difficult to measure.

As the transducer and block temperature was raised above 600 °C, it approached the softening point of the intermediary silver foil (600-640 °C per specifications given by the manufacturer). This softening promoted an increase in acoustic coupling, and more efficient energy transfer to the test block. It is believed that this effect is responsible for the relatively higher values of SNR at very high temperature (Fig. 5.5). The SNR was calculated as follows:

\[
SNR = 20 \log \left( \frac{V_s}{V_N} \right)
\]

in which \(V_s\) and \(V_N\) are the peak value, and the RMS value of the noise of the transducer received signal, respectively.
Fig. 5.4. Received first back-wall echo from the test block at various temperatures: a) $T=27\,^\circ C$, b) $T=100\,^\circ C$, c) $T=200\,^\circ C$, d) $T=300\,^\circ C$, e) $T=400\,^\circ C$, f) $T=500\,^\circ C$, g) $T=600\,^\circ C$, h) $T=700\,^\circ C$, i) $T=800\,^\circ C$.

Fig. 5.5. SNR of the received signals from the test block vs temperature.
To investigate the temperature dependence of the acoustic coupling between transducer and test block, a new set of experiments was performed using a disc spring to keep the pressure approximately constant across the transducer-test block interface. As shown in Fig. 5.6, use of the disc spring kept the acoustic coupling between the transducer and test piece at a constant value at temperatures up to 400 °C; back-wall echo signals were very stable over this temperature range. The resultant SNR values were quite close and around 28-30 dB. However, the temperature limit of the disc springs was approximately 500 °C (per manufacturer’s specifications). Therefore, the acoustic coupling and consequently the signal amplitude weakened gradually as this temperature was approached.

Comparing these results with those shown in Fig. 5.4, it can be concluded that the primary reason behind the variation in the signal amplitude with temperature is the variability in acoustic coupling, not degradation of the piezoelement. Furthermore, signal amplitude recovered when the temperature was returned to ambient levels such that there clearly had been no fundamental changes in piezoelectric properties of the crystal. Similar behavior and conclusions were observed in Ref. [97, 98], where a temperature increase led to a reduction in acoustic coupling and consequently a weaker signal.

Figs 5.7a-i show the magnitude spectra obtained from Fourier transforms of the back-wall echo signals of Fig. 5.4. As expected, the graphs show a minimum in signal amplitude at about 600 °C and then an increase as the temperature is raised towards 800 °C. The amplitude peak occurs at 3.2 MHz at room temperature; this is slightly higher than the design value of 3 MHz. A primary reason for the discrepancy is that the Olympus 5077PR pulser excitation center frequency is set at 3.5 MHz due to limited flexibility in the choice of pulse settings. As the temperature is increased, small changes in piezoelectric properties of the crystal [48] cause the central frequency to down to
2.7 MHz at 800 °C. The 6 dB signal bandwidths corresponding to Figs. 5.7a through 5.7i were
found to be 75%, 70%, 77%, 60%, 70%, 70%, 75%, and 75%, respectively (Fig. 5.8).

Fig. 5.6. Received back-wall echo from the test block at various temperatures. Disc springs were used to
clamp the transducer to the test piece.

The main reason for the difference between the 95% bandwidth predicted by the KLM model
and lower value seen in experiment is believed to be the imperfect interfaces linking individual
transducer components and the test block. In the KLM model, the effect of all bonding or coupling
layers is neglected; it is also assumed that all transducer components and the test block are perfectly
coupled together for ultrasonic transmission. In addition, lateral wall boundary effects (neglected
in the 1D KLM model) may also affect the signal bandwidth. Software for 3D simulation of the
transducer was not available.
Fig. 5.7. Fourier-transform of the received signals from the test block at various temperatures: a) T=27 °C, b) T=100 °C, c) T=200 °C, d) T=300 °C, e) T=400 °C, f) T=500 °C, g) T=600 °C, h) T=700 °C, i) T=800 °C.

Fig. 5.8. 6dB bandwidth of the received signal at various temperatures.
5.4. Concluding remarks

Two different bonding techniques were investigated for assembling the transducer elements: adhesives, and brazing alloys. The selected adhesives yielded either a lack of good mechanical bonding between critical transducer components, or very poor ultrasonic coupling at elevated temperatures for both type of transducers. Neither of the brazing alloys resulted in a satisfactory bond to link components of a GaPO₄ based transducer. However, stable bonding linking a porous zirconia backing layer, LiNbO₃ piezoelement, and alumina quarter-wave matching layer was finally achieved by brazing with TiBraze®Al-665 foil.

Using a prototype transducer based on a LiNbO₄ piezoelement and TiBraze®Al-665 foil, back-wall echoes from a steel reference block were recorded at temperatures ranging from ambient up to 800 °C. Despite the observed temperature dependence of transducer coupling to the steel block, clear echo signals were recorded over the entire temperature range. Signal amplitude decreased as the transducer temperature was raised to 600 °C, and then started to increase as the temperature was raised further up to 800 °C; this variation was found to be due primarily to temperature-dependent coupling between transducer and test block. The center frequency of the signal gradually dropped from 3.2 MHz at room temperature to 2.7 MHz at 800 °C. The signal 6 dB bandwidth showed small changes over the temperature range and its average value was approximately 70%. At 800 °C, the 6 dB bandwidth of the echo signal was 75%, at a center frequency of 2.7 MHz. The transducer was found to retain full functionality after cooling from 800 °C to room temperature.
Chapter 6

6. Conclusions and future work

The purpose of this project was to develop a model-based system for designing and manufacturing ultrasonic transducers, tailored for high-temperature industrial applications. This objective was then extended to the development of two transducer designs, based on two different piezoelements. The following specifications, provided by our industrial partner, were targeted for the final transducer:

- Signal center frequency of 2.7-3 MHz with minimum 3 dB bandwidth of 2.7 MHz (90%).
- Functional at temperatures as high as 700-800 °C.
- Continuous trouble-free operation for at least two years for 90% of transducers.
- Maximum high-volume manufacturing cost of $1,500, compared to the currently-available commercial competitor's price of $5,000.
• Reliable and stable acoustic coupling between transducer components. All interfacial bonds must be capable of accommodating any interfacial stresses originating from differences in CTEs of transducer components, from room temperature up to the operating temperature range of 700-800 °C.

To this purpose, a multistep design sequence was implemented:

1. Piezoelectric materials suitable for high temperature environments were identified. Several features of these materials were examined, including dielectric properties, mechanical coupling coefficient and thermal stability.

2. For a known piezoelement and target test specimen, an appropriate material and thickness for quarter-wave matching layer were determined. The matching layer is expected to be thermally and chemically stable at high temperatures (800 °C), and show CTE close to those of the piezoelement.

3. The 1D KLM transducer model was implemented to estimate the optimal acoustic impedance of the backing element to obtain the desired transducer signal bandwidth.

4. Porous ceramics were introduced as a new generation of backing elements in ultrasonic transducers for high temperature applications. An acoustic model was employed to estimate the required porosity and pore size to obtain desired acoustic properties for the backing element.

5. Suitable ceramics were identified and manufacturing processes were optimized to achieve defect-free backing elements with the prescribed porosity and pore size.

6. Various high temperature adhesives and brazing alloys were tested on prototype, non-encapsulated transducers. The bonding quality was assessed by examination of the bond layer for cracks and lack of adhesion. The best candidate was identified, and the
effects of temperature on signal strength, center frequency and bandwidth were studied at temperatures up to 800 °C.

6.1. Conclusions

- Among several possible piezoelectric materials, 36° Y-cut LiNbO₃ and X-cut GaPO₄ were found to be the most promising, commercially-available candidates for operation at high temperatures.
- The KLM model predicted that a backing element with acoustic impedance range of 20-25 MRayls and 14-15 MRayls for LiNbO₃ and GaPO₄ piezoelements, respectively, would yield the signal with desired bandwidth.
- To operate at high temperatures (800 °C), 3% YSZ and mullite ceramics were found suitable as the backing element materials for LiNbO₃ and GaPO₄ piezoelements, respectively.
- The acoustic model of porous media resulted that porous 3% YSZ ceramic with porosity of 25% and average pore diameter of 200 µm would yield the optimal acoustic properties. The corresponding results for mullite were 30% porosity and average pore diameter of 400µm.
- Porous ceramics were manufactured using PE spheres as the pore forming agent. After optimizing the pressing and sintering procedure for each ceramic material, defect-free samples were obtained. SEM images of cross-sections of the porous samples revealed no evidence of cracks at pore walls or in the bulk ceramic matrix.
- Immersion ultrasonic measurements showed that porous zirconia with 25% porosity and average pore diameter of 130 µm, as well as, porous mullite with porosity of 30% and average pore diameter of 230 µm yielded the desired ultrasonic properties.
- Experimental measurements of attenuation in the porous backing materials yielded values considerably higher than those predicted by the numerical model. The reason is that the model
neglects the hysteresis damping of the ceramic matrix. Also, it does not account for the effect of pore aggregation, non-spherical pores, and a broad distribution of pore diameters.

- No brazing alloy and adhesive yielded bonding between GaPO4 and porous mullite backing.
- TiBraze®Al-665 brazing foil was found as the sole candidate to bond a LiNbO3 piezoelement to porous zirconia backing and alumina matching layer. Strong, repeatable back-wall echoes from a steel reference block were recorded with a prototype non-encapsulated transducer at temperatures ranging from ambient up to 800 °C.
- After cooling to room temperature, the interfacial bonds between the transducer components were found undamaged as the transducer was performing properly.

6.2. Contributions

- A multi-step sequence for designing and manufacturing transducers suitable for operation at high temperatures has been developed. For any desired signal center frequency and bandwidth, the design parameters can be adjusted accordingly.
- The application of porous ceramics as backing elements in high temperature transducers has been proposed and tested. They can be adapted to produce backing elements with a variety of values of acoustic impedance and attenuation.
- The manufacturing of porous ceramics was optimized to obtain high thickness-to-radius ratio porous samples with no defects.
- High temperature brazing alloys suitable to bond piezocrystals to ceramic parts have been identified and evaluated.
- A new ultrasonic transducer for operation at high temperatures up to 800 °C was successfully prototyped and tested. Compared to other type of transducers, e.g, electromagnetic acoustic
transducers (EMATs) and magneto-restrictive transducers, the proposed transducer obtains higher SNR and requires remarkably less power to run.

6.3. Future work

- The KLM model considers only one dimensional wave propagation along the transducer axis. It also cannot account for the imperfect flatness of the surfaces at bonding interfaces. A 3D finite element model would reveal the effects of waves with radial components, and possibly the generation of extra, unwanted pulses from internal sidewall reflections.

- Attempts to bond GaPO₄ to porous mullite backing material using a selection of conductive, high-temperature adhesives and brazing alloys were not successful. A more extensive study is required to investigate other possible bonding candidates, e.g. silver or gold brazing alloys.

- Due to time constraints, the long-term durability of the transducer components and bond layers in a high temperature environment was not investigated. High temperature and excessive mechanical stress can gradually depolarize the piezoelement [99], and may cause chemical degradation at the brazing interfaces. Both thermal cycling, and sustained operation at high temperature should be considered in endurance tests.

- The capability of the transducer to withstand thermal shock in the case of very sudden temperature changes needs to be investigated. Although finite element modeling could be of assistance, this evaluation will ultimately need to be performed experimentally.

- Alternative materials to acoustically couple a transducer to a test piece need to be investigated. Consistent coupling is required over a large temperature range. For this study, a 50 µm thick silver foil was used, placed between the transducer and the steel test piece. Good coupling was obtained when temperature reached the softening point of the silver, but other soft materials
might provide more consistent coupling. The optimal thickness of the foil and a fixture to apply an appropriate pressure to obtain good coupling must be investigated.

- A transducer housing is composed of several components that together encapsulate the multilayer backing-piezoelement-matching structure and provide electrical connections to the two piezoelement electrodes. The key housing components are: (i) the casing, (ii) wires and leads, and (iii) electrical insulators. All these elements must maintain their mechanical and mechanical properties at high temperatures, while resisting corrosion. The housing must accommodate thermal expansion of the individual elements.

- A proper design of clamping fixture is required to press the transducer on the test piece, while keeping the clamping pressure constant over the temperature range. Besides, the fixture must keep the transducer aligned when it is mounted on pipes.

In the following section, a conceptual design for a transducer housing is proposed, as a possible guide to future work on a transducer housing.

6.3.1. Proposed Encapsulation System

Three factors are key in the design of an encapsulation system for the transducer: connection of piezoelement electrodes to the wires, electrical insulation, and the effect of temperature variations on thermal expansion of transducer components. Fig.6.1 shows a proposed design for the entire assembled transducer. All components must be secured within the transducer casing for ease of handling and to protect the interior components from damage.

The two electrodes of the piezoelement must be electrically connected to the live and ground wires of the cable. The “live” wire from the pulser which carries the excitation voltage is connected to top electrode of the piezoelement through the center rod of the transducer. The electrical ground

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wire is connected to the casing which is connected to the extension tab of the brazing foil used to bond the piezoelement to the matching layer.

Several electrical insulators are used in the transducer housing to isolate components carrying power from those connected to the ground. Three ceramic insulators are implemented in shapes of cylinder, disc and tube. The purpose is to separate the central thin rod, disc connector, and the backing from the housing.

Each transducer component has a slightly different CTE, despite efforts to select materials that are mutually compatible in this regard. Consequently, a small gap will grow between the metal housing and backing element with increase in temperature. To fill the gap at all temperatures, a preloaded high-temperature wave spring is included in the transducer. In addition, the tolerances used for manufacturing the insulator components must be very fine to avoid the generation of large interfacial stresses at high temperatures.
Fig. 6.1. Section view of transducer assembly.
References:


[34] Baba A, Searfass CT, and Tittmann BR, High temperature ultrasonic transducer up to 1000 °C using lithium niobate single crystal. Applied Physics Letter 2010; 97: 1–3.


