# Development of Polymer-Ceramic-Metal Graded Acoustic Matching Layers via Cold Sintering

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Abstract—A family of three phase, polymer-ceramicmetal (Poly-cer-met) electrically conducting composites was developed via cold sintering for acoustic matching application in medical ultrasound transducers. A range of acoustic impedance (Z) between 5 < Z < 21 MRayl with low attenuation (<3.5 dB/mm, measured at 10 MHz) was achieved in composites of zinc oxide, silver, and in thermoplastic polymers like Ultem polyetherimide (PEI) or polytetrafluoroethylene (PTFE) at sintering pressure less than 50 MPa and temperature of 150 °C. Densities exceeding 95% were achieved, with resistivities less than 1 Ω-cm. The acoustic velocity was homogeneous across the part (variations <5%). The acoustic velocities exceeded 2500 m/s for Z above 12 MRayl. The experimentally measured acoustic impedance of ZnO/Ag/PEI composites was observed to be in close agreement with the theoretical logarithmic model developed for different volume fractions of individual phases at the percolation limit for Ag. Thus, the acoustic properties of this family of matching layers (MLs) can be predicted to a good approximation before experimental realization. Additionally, a non-conducting low Z (5 MRayl < Z < 12 MRayl) with acoustic velocities exceeding 2000 m/s was achieved using hydrozincite as the ceramic component. Scaling of the composites to 2" diameter was demonstrated. A –6 dB bandwidth greater than 85% was measured for a three ML ultrasound transducer, fabricated using a single cold sintered layer (Z = 19 MRayl) and two other commercial layers in the stack. Finally, a co-cold sintered graded prototype consisting of three tape-casted formulations corresponding to Z = 5, 9, and 19 MRayl, while still retaining the correct distributions of the components was demonstrated.

Index Terms—Acoustic impedance, acoustic matching, attenuation, cold sintering, ultrasound transducer.

#### I. INTRODUCTION

ULTRASOUND imaging is a valuable diagnostic imaging modality in medicine due to its noninvasive and

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nonionizing nature, cost effectiveness, and real-time imaging ability [1], [2]. Ultrasonic waves produced by piezoelectric transducers are focused and sent into the human body through one or more acoustic matching layers (MLs) and gel coupling material. Typically, absorptive backing materials dampen acoustic ringing in the transducer, promoting higher bandwidth though at the expense of the overall sensitivity [3].

Acoustic MLs are critical for the performance of highfrequency ultrasound imaging transducers. Without proper MLs, the large acoustic impedance mismatch between the piezoelectric element (>30 MRayl) and human tissue (~1−2 MRayl) will cause the majority of the acoustic energy to be reflected at the transducer-tissue interface, resulting in poor resolution and sensitivity [4]. Transducer designs set the thickness of the ML at a quarter wavelength [5] for the center frequency of a broadband transducer, with multiple MLs having acoustic impedance in the range of 2–14 MRayl, depending on the type of the piezoelectric material [3], [6]-[8]. This composite design with multiple stacked MLs improves the transducer sensitivity due to low transmission loss, low ripple in the bandpass region and wider spectral performance. For example, it has been reported that a transducer with a single ML [8] has 40%-50% 6-dB bandwidth, while a double ML [6], [7] has ~70% fractional bandwidth. Inoue et al. [9] have shown that for a 3.5-MHz air-backed PbTiO<sub>3</sub> disk linear array transducer, switching from a single ML to a triple ML configuration provides low-loss, excellent impulse response, and wider spectral characteristics (88%), with more than a 40% increase in 6-dB fractional bandwidth. For a double ML design, the optimized acoustic impedance values are found to be 8.9 MRayl and 2.3 MRayl for PZT-5H based transducers [7]. For a transducer with more than three MLs, impedances above 14 MRayl are required [3], [6]. Additionally, switching from conventional piezoelectric materials such as lead zirconate titanate (PZT) to high electromechanical performance families of single crystals (such as lead magnesium niobate - lead titanate, PMN-PT) as an ultrasound source would require redesign of current ML schemes to enable >110% fractional operational bandwidth [10]-[12].

Theoretical predictions [13] have shown that an inhomogeneous ML with continuously changing acoustic impedance having an exponential matching profile can enable broadband characteristics. The concept of a graded tapered ML design [14] was first experimentally demonstrated by Haller and Khuri-Yakub [15] and later modified with a metamaterial

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design by Li *et al.* [16], however, the fabrication of the metamaterial design is cumbersome for use on a commercial scale.

Typically, polymer composites with metal fillers are used for the MLs [17]–[21], where the stack of acoustic MLs is bonded to the active piezoelectric element. Low attenuation is desirable to minimize losses and achieve greater transducer sensitivity. Making such composites is challenging for highfrequency transducers (10–100 MHz). Because the thickness of the quarter wavelength is less than 10  $\mu$ m, the composite particle size is required to be much less than 1  $\mu$ m in order for the composite to avoid acoustic scattering and increased attenuation [17], [22]. In addition, high concentration submicrometer metal powder loaded polymer composites have homogeneity issues and are difficult to machine (e.g., lapping or grinding and dicing [23]).

While the primary function of the MLs is an acoustic one, secondary properties are also of importance. For example, electrically conductive ML(s) facilitate electrical access to the individual elements in a phased-arrayed ultrasonic transducer [24], [25]. From a materials perspective, when the requirement for high electrical conductivity is added, the options become quite limited to a range of approximately 6–11 MRayl. While some materials are available outside of this range, they all suffer for other reasons, such as cost or manufacturability [17].

The goal of this work is to demonstrate a methodology that allows graded matching multilayers via fabrication of acoustic MLs with cold sintering [26], a process that enables cosintering of diverse material groups. It is typically challenging to cosinter ceramics with polymers due to incompatibilities at high temperatures. Cold sintering is an emerging methodology [27] for the development of high density (>95%) ceramics, including ferroelectrics [28]-[32], thermoplastic polymer-ceramic composites [33]-[39], thermoset polymer-ceramic composites [40], Li-ion batteries [34], etc., at unusually low temperatures < 300 °C and in reduced times relative to conventional sintering approaches. In cold sintering, densification occurs under uniaxial pressures in conjunction with transient liquid phases [41] that (often) partially dissolve the constituents, rearrange the particle compact, and reprecipitate the dissolved ions at the pores as the liquid phase is vaporized from the open system [42]. Cold sintered polymer-ceramic composites with high ceramic volume fractions (volume fraction >95%) retain the properties of individual phases by engineering ceramic grain boundaries with polymer functionalization.

Cold sintering is a promising route to exploit the diverse properties of dense, heterogeneous composites comprising polymer, ceramic, and metal (Poly-cer-met) families to design graded MLs for a wide range of acoustic impedances. The current work focuses on developing processes for preparing a family of electrically conductive composites using zinc oxide (ZnO) or hydrozincite (Zn<sub>5</sub>(CO<sub>3</sub>)<sub>2</sub>(OH)<sub>6</sub>) as the ceramic phase, polytetrafluoroethylene (PTFE) or polyetherimide (PEI) as the polymer phase and Ag as the conducting phase, targeting a broad range of acoustic impedance values. Zinc oxide is one of the widely studied cold sintered ceramics which can readily cold sintered with lower volume fractions of polymer and shows retention of individual phase properties by engineering ceramic grain boundaries with polymer functionalization [38]. The individual poly-cer-metal composite layers corresponding to Z = 5, 9, and 19 Mrayl have been tape-casted to the desirable thickness and the stack co-cold sintered to achieve a graded ML prototype. This provides an alternate approach to readily fabricate dense MLs with stack thicknesses ranging from 5 to 200  $\mu$ m, without needing to bond and grind a bulk layer. It is also noted that the methodology is general, such that different materials that can be cold sintered could be employed in place of ZnO and hydrozincite.

# II. MODELING OF THE COMPOSITE ACOUSTIC IMPEDANCE

Each component (ZnO or hydrozincite, Ag and PEI or PTFE) in the composite is assumed to be interconnected in three dimensions such that the composite has a 3–3–3 connectivity pattern. The density of a three-phase system is approximated by the linear mixing model

$$\rho_{\text{composite}} = \rho_{\text{ceramic}} * V_{\text{ceramic}} + \rho_{\text{metal}} * V_{\text{metal}} + \rho_{\text{polymer}} * V_{\text{polymer}}$$
(1)

where  $\rho$  is the density and  $V_i$  is the volume fraction of phase *i*. The velocity of the acoustic wave  $(v_P)$  and the acoustic impedance *Z* of a material are related to the material elastic bulk modulus (*K*) and density ( $\rho$ ) through the following expressions:

$$v_P = \left(\frac{K}{\rho}\right)^{1/2} \tag{2}$$

$$Z = (\rho * K)^{1/2}.$$
 (3)

For this work, the logarithmic model was found to be a good fit to estimate the elastic modulus of the three-component composite given by the following relation [43]:

$$\ln(K_{\text{composite}}) = V_{\text{ceramic}} * \ln(K_{\text{ceramic}}) + V_{\text{metal}} * \ln(K_{\text{metal}}) + V_{\text{polymer}} * \ln(K_{\text{polymer}}). \quad (4)$$

The estimated bulk modulus at a particular volume fraction was then multiplied by the composite density to calculate the impedance at that volume fraction. Table I summarizes the material properties used to estimate the volume fractions of each component corresponding to the acoustic impedances in the range 5–19 MRayl while Tables II and III outline the theoretical volume fraction for the desired Z using PEI and PTFE based composites, respectively.

### III. EXPERIMENTAL PROCEDURES

The experimental work focused on: 1) cold sintering of a three-phase component system targeting a range of acoustic impedance and studying the compositional dependence of the acoustic properties and 2) developing graded matching multilayers via tape-casting of the three-phase composite system and subsequent cold sintering of the composite stack.

TABLE I	
MATERIAL PROPERTIES USED IN THE COMPUTATION OF ACO	OUSTIC IMPEDANCES FOR THE COMPOSITE

Material	Density (g/cm <sup>3</sup> )	Acoustic impedance (MRayl)	Elastic modulus (GPa)	v <sub>P</sub> (m/s)
ZnO [50]	5.6	34.6	214	6200
Hydrozincite*	3.4	15.8	73.2	4640
Ag [50]	10.5	37.8	136	3600
PEI [51]	1.27	3.11	7.62	2450
PTFE [52]	2.2	2.97	4.01	1340

\*Estimated empirically

#### TABLE II

PEI BASED COMPOSITE FORMULATION BY % VOLUME FOR VARIOUS Z USED IN THIS STUDY

Z = 18	75.4 % ZnO, 16.6 % Ag, 8 % PEI
Z = 14	63.4 % ZnO, 16.6 % Ag, 20 % PEI
Z = 9	57.4 % ZnO, 16.6 % Ag, 27 % PEI
Z = 5	14 % ZnO, 16.6 % Ag, 69.4 % PEI

# Z (MRayl) Formulation (by vol%)

#### TABLE III

PTFE AND HYDROZINCITE BASED COMPOSITE FORMULATION BY % VOLUME FOR VARIOUS Z USED FOR TAPE-CASTING

## Z Formulation (by vol%) (MRayl)

Z = 19	50.5% ZnO, 18.5% Ag, 31% Hydrozincite
Z = 9	85% Hydrozincite + 15% PTFE
Z = 7	70 % Hydrozincite, 30 % PTFE
Z = 5	50% Hydrozincite + 50% PTFE

#### A. Materials

Pure ZnO powder (Alfa Aesar Nanoarc ZN-0605, 99.9% purity) with a particle size between 40 and 100 nm was chosen as the starting ceramic material. Hydrozincite (zinc carbonate basic) from Sigma Aldrich was another ceramic choice. Polymer materials considered for the study were custom attrition milled PEI powder (SABIC, Ultem<sup>1</sup> F3SP 1000 resin) with particle size  $D_{50} \sim 1.29 \ \mu$ m (where  $D_{50}$ , the median diameter of particle size distribution, was the value of the particle diameter at 50% of the cumulative distribution) and PTFE (Howard Industries) with particle size less than 3  $\mu$ m. Silver

<sup>1</sup>Trademarked.

powder (Sigma Aldrich, flakes, <10  $\mu$ m size, ≥99.9% or Dupont KOO3L) was chosen to achieve resistivity < 1  $\Omega$ -cm in the composite.

#### B. Pellet Preparation via the Cold Sintering Process

For the ZnO-polymer-Ag system, pure ZnO powder and PEI or PTFE powder were separately ball-milled for 24 h with 1-10 mm yttria-stabilized zirconia (YSZ) milling media. To determine the percolation limit in the two-phase ceramicmetal system, ZnO powder or PEI powder was mixed manually with varying volume fractions of Ag (10%-20%) in an agate mortar and pestle for 5 min. Aqueous solution of acetic acid (1.66 M HOAc, Fisher Scientific A38-500, >99.7% w/w) was used as the transient liquid sintering aid to densify the composite. Approximately 4% (by weight) liquid phase was added to the composite powder mixture; the resulting mixture was mixed manually for 15 min. The powder was then sieved through a mesh (#400) and loaded into a 12.7 mm diameter stainless steel die (Wartburg Tool & Die, Inc., Wartburg, TN, USA) and pressed uniaxially in a Carver laboratory press (Model M, 25-ton) to form a cylinder approximately 1.8 mm thick. The temperature was controlled with a Tempco Corporation, Wood Dale, IL, USA, heating band (part #MBH35489), mounted around the perimeter of the die with a thermocouple adhered between the die and band. Pressure was applied within a few seconds to a constant value at 150 MPa for 300 min. For the three-phase system, silver powder (at percolation limit of 16.6% vol. fraction) was added to achieve



Fig. 1. Schematic for tape casting process flow ( $Z_1$ ,  $Z_2$ , and  $Z_3$ : MLs corresponding to three different formulations).

an electrical resistivity  $< 1 \Omega$ -cm in the composite. A similar process was implemented on the three-phase system. For 2" samples, the cold sintering process (CSP) was performed in a commercial MTI die (EQ-Die-75D). The amount of transient liquid phase was increased to 4%–6% by weight. The compositions were slightly modified to account for the loss in material due to imperfect die sealing. All of the composites were hand polished using a series of grit polishing papers to form flat surfaces. To demonstrate a clear bond-line formation for different Z formulations, two different composite powder mixtures corresponding to Z = 19 and Z = 7 MRayl were loaded sequentially into the 2" die and cold sintered at 150 °C at 150 MPa for 300 min.

## C. Tape-Casting Process Followed by Cold Sintering

Formulations corresponding to Z = 5, 9, and 19 MRayl were chosen to demonstrate the feasibility of a graded matching multilayer prototype. Fig. 1 shows the schematic for the developed tape-casting process for the prototype. The ceramic and polymer powder corresponding to each Z was dry milled for 12 h using 1-10 mm YSZ milling media. The tape cast process was facilitated by two vehicles: vehicle A was composed of 95 wt% methyl ethyl ketone (MEK, Alfa Aesar, Tewksbury, MA, USA) and 5 wt% poly (propylene carbonate) (QPAC<sup>1</sup>40, Empower Materials, Inc., New Castle, DE, USA); vehicle B consisted of 65 wt% MEK, 28 wt% QPAC<sup>1</sup>40, and 7 wt% butyl benzyl phthalate (Santicizer-160, Tape Casting Warehouse, Inc., Morrisville, PA, USA). The ceramic/polymer powder was mixed with 12.96 g vehicle A, 10 g MEK, and 11.41 g vehicle B and ball milled for 12 h. Before film casting, silver flakes were added to the slurry and mixed then defoamed in the Thinky mixer (ARE-310) for 30 min. The slurry was tape cast on a Mylar sheet with a blade height at a speed of 8 cm/sec. The as-cast tape was then air-dried at room temperature for 30 min to remove MEK; the

final thickness of the dried tape was ~80  $\mu$ m. The tapes were cut into circular pieces one half-inch in diameter (~12.7 mm) and were laminated to reach the desired thickness. Lamination was done in a half-inch-diameter circular die at <50 MPa, 60 °C for 30 min. The binders were then burned out at 180 °C in a box furnace under ambient conditions with a ramp rate of 0.4 °C/min to 180 °C, a hold time of 360 min, and a cooling rate of 5 °C/min. Following burn out, the samples were wetted using steam introduced by heating 1.66M acetic acid at 80 °C for 24 min to introduce the "liquid phase" for cold sintering. The stack was immediately cold sintered at 150 °C under 150 MPa uniaxial pressure for 180 min. To lubricate the plunges in the die, a PTFE-containing release agent (MR311, Spray-on, Cleveland, OH, USA) was sprayed before loading the samples.

#### D. Material Characterization

The geometric density was calculated from the weight and sample dimensions. The fractional density is given as the ratio between the measured and theoretical density of composite powder based on the volume fraction of each phase. Bulk densities were also measured by the Archimedes method on 2" pieces, using water as the liquid medium at room temperature due to presence of defects on the sample edge from the die used during cold sintering. Phases of sintered pellets were characterized by X-ray diffraction (XRD) using a PANalytical diffractometer, operating at 45 kV and 40 mA with intensities between 15° and 80°, using a step size of 0.013° with Cu Ka radiation. Scanning electron microscopy (SEM) was performed on the FEI Verios G, with accelerating voltages of 5 kV. The grain size measurement was performed using Image J software on more than 150 grains. Energy dispersive X-ray spectroscopy (EDS) mapping was performed to observe the phase distribution in the specimen for different compositions. The percolation limit for Ag in both ZnO and



Fig. 2. Schematic showing the various signals used in the attenuation measurement on the composite sample ( $V_i$ : transducer generated signal,  $V_0^{\text{FW}}$ : first front wall signal,  $V_0^{\text{BW}}$ : first backwall signal and  $V_0^{\text{BW2}}$ : second backwall reflection peak).



Fig. 3. Experimental determination of the Ag percolation limit in (a) ZnO nanoparticles (inset showing the SEM of the silver flakes) and (b) PEI (inset showing the SEM of the PEI particles). The right axis represents the measured fractional on the two-phase composites.



Fig. 4. (a) Measured and fractional density of ZnO-Ag-PEI composites. (b) XRD data for the compositions corresponding to Zranging to 5–18 MRayl.

PEI matrices was determined empirically using bulk resistivity measurements with a custom-built four-point probe system. For this purpose, 100 nm Au was sputter deposited as electrodes to minimize the effect of contact resistance. The acoustic velocity for each composite was calculated from the time of flight through sample thickness, measured using a pulse-echo setup with a pulser-receiver (Panametrics 5052) coupled to a 10-MHz contact transducer

% Volume Ag	Geometric density (g/cm <sup>3</sup> )	Fractional density (%)	Resistivity
10	5.9	97.1	4.7 GΩ-cm
12	5.8	93.7	0.25 GΩ-cm
14	6.0	94.9	0.25 KΩ-cm
15	5.9	93.7	3.6 Ω-cm
17	6.0	93.3	0.44 Ω-cm
20	6.2	94.2	0.22 Ω-cm

TABLE IV DENSITIES AND RESISTIVITY OF THE COLD SINTERED ZnO-Ag COMPOSITE



Fig. 5. Energy dispersive spectroscopy (EDS) data showing distribution of C, Zn, and Ag for the compositions corresponding to (a) Z = 5 MRayl, (b) 9 MRayl, (c) 14 MRayl, and (d) 18 MRayl.

(0.25" diameter, NDT Systems, Huntington Beach, CA, USA, part# PDG 102) for excitation. The acoustic impedance was obtained using the composite densities and the calculated velocities. Acoustic attenuation ( $\alpha$ ) was quantified using the air-backed contact method. The amplitudes of the transducer generated signal ( $V_i$ ), first front wall ( $V_0^{FW}$ ), first backwall ( $V_0^{BW2}$ ) reflection peaks as shown in Fig. 2 were measured using an oscilloscope and attenuation

was calculated using the following relationship:

$$\alpha = \frac{-20}{2 * \text{thickness}} \log_{10} \frac{V_0^{\text{BW}} * V_i}{V_0^{\text{BW}} * V_0^{\text{FW}}}.$$
 (5)

Additionally, velocity scanning maps and C-scans [44]–[46] were obtained by immersion of the composite in water to assess the acoustic homogeneity and detect macro-flaws through the thickness.

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Fig. 6. SEM data showing the densification of grains (with average grain size) in compositions corresponding to (a) Z = 5, (b) Z = 9, (c) Z = 14, and (d) Z = 1.8 MRayl. (e) ZnO-25%PTFE-16.6% Ag.

## IV. RESULTS AND DISCUSSION

#### A. ZnO-Ag-PEI Composite System

The experimental results and analyses are presented in three parts.

- Determination of the percolation limit for conductivity in ceramic and polymer system.
- Analyzing the compositional dependence of individual phases on the acoustic properties of the composite corresponding to each acoustic impedance.
- Lateral scaling of the composite system.

1) Percolation Limit of Ag in ZnO and PEI Matrices: Dense, pure phase half-inch pellets were successfully cold sintered with 10%–20% vol. fraction of Ag in ZnO ceramic and 7%–16.6% vol. fraction of Ag in PEI. Tables IV and V summarize the density, fractional density, and resistivity of the two-phase system for ZnO and PEI, respectively. The electrical percolation threshold for Ag in each of the two-phase systems can be determined from Fig. 3(a) and (b), which illustrate the variation in electrical conductivity and fractional density as a function of vol% Ag. The percolation limit is defined as the minimum concentration of the metallic filler which allows formation of a continuous conductive network, thereby causing an insulator-metal transition and is characterized by an abrupt change of conductivity [47]. The ZnO-Ag composite

% Volume Ag	Geometric density (g/cm <sup>3</sup> )	Fractional density (%)	Resistivity (Ω-cm)
7	1.92	99.5	8.41 G
10	2.18	99.3	2.62 G
11	2.17	95.2	4.1
12	2.38	99.8	0.23
16.6	2.79	99.4	0.09

TABLE V DENSITIES AND RESISTIVITY OF THE COLD SINTERED ZnO-PEI COMPOSITE



Fig. 7. Velocity scans across the 1/2 in sample with composition for Z = 5 MRayl. (a) Lateral dimension. (b) Through-thickness B-scan measured at 10 MHz.

with flake Ag filler starts to percolate at 12% vol Ag and plateaus at 16% vol. Ag while the PEI-Ag composite starts to percolate at 10% vol. Ag and plateaus at 12% vol. Ag. The percolation threshold of the PEI system is at a lower concentration and the conduction onset is sharper than the ZnO system. Although ZnO is smaller in size compared to PEI, the difference in the percolation threshold may be attributed to the easier compressibility of the filler (Ag flakes) in a low modulus polymer matrix compared to a stiff ceramic matrix [47]. Since the acoustic transducer design typically requires the conductivity of the ML to be less than 1  $\Omega$ -cm, the volume fraction of Ag for the three-phase composite system was chosen to be 16.6%.

2) Densification of ZnO-Ag-PEI and the Dependence on the Acoustic Properties of the Composite Composition for Each Target Acoustic Impedance: ZnO-Ag-PEI composites were cold sintered with compositions corresponding to Z varying from 5 to 18 MRayl with formulation by vol% as listed in Table II. As seen in Fig. 4(a), the fractional densities of the composites are >95% of the theoretical density (calculated using (1) for the formulation). As seen in Fig. 4(b), ZnO and Ag are the only crystalline phases. The composites were investigated using low resolution EDS mapping to observe the distribution of the individual phases. As observed in Fig. 5, the overall distribution of PEI and Ag as indicated by the C and Ag maps, respectively, is uniform in all four compositions.



Fig. 8. Influence of pressure on the absolute and fractional density of ZnO-16.6% Ag-8% PEI composite in CSP at 150 °C for 5 h.

However, there is some preferential segregation and densification of ZnO in the Z = 5 MRayl and Z = 9 MRayl compositions. This might be attributed to either incomplete mixing of the individual precursors or preferential densification/agglomeration of ZnO grains with increasing vol% of polymer concentration. Fig. 6(a)–(d) shows the microstructure development in the various ZnO-Ag-PEI composites. From



Fig. 9. Spatial distribution of Z and velocity measured at 10 MHz in a 2" cold sintered pellet for composition targeting Z = 14 MRayl with (a) and (b) uniform pressure distribution and (c) and (d) nonuniform pressure distribution during cold sintering.

previous studies, the average grain size obtained after cold sintering pure ZnO is ~300 nm  $\pm$  100 nm [40], [48]. The addition of increasing vol% of PEI leads to densification of the ZnO matrix, however a reduction in the average grain size occurs, with 215 nm  $\pm$  151 nm, 262 nm  $\pm$  186 nm, 281 nm  $\pm$  205 nm, and 345 nm  $\pm$  191 nm for Z = 5, 9, 14, and 18 MRayl, respectively. This is consistent with findings from previous ZnO-polymer composite work [38], [40], where the polymer is believed to act as a diffusion barrier, preventing grain growth during CSP.

Table VI summarizes the measured acoustic velocity and impedances for the conductive cold sintered samples with compositions targeting Z = 5 MRayl to Z = 18 MRayl. There is good agreement between the modeled and measured Z. All the compositions were conductive with resistivity less than 1  $\Omega$ -cm. The attenuation in all four compositions was less than 3.5 dB/mm, measured at 10 MHz. Previous work [20] has shown that it is challenging to manufacture composites

with both high-Z and low attenuation. However, the threephase high-Z cold sintered composites show low losses.

Additionally, despite ZnO segregation, compositions corresponding to Z = 5 MRayl and Z = 9 MRayl had low attenuation. Previous work by Grewe *et al.* [43] demonstrated that for some material systems (with larger mismatch between the acoustic impedance of the filler and the matrix), increasing the particle size of the filler generally increases the composite attenuation. In this work, it is hypothesized that the length scale of ZnO particle segregation is not large enough to cause significant attenuation. Velocity scans obtained from the time-of-flight measurement showed good uniformity across the 1/2 inch sample for Z = 5 MRayl composition, as illustrated in Fig. 7(a). Furthermore, the pulse-echo B-scan shown in Fig. 7(b) shows dense monolithic structure without any large macro flaws in the part piece.

3) Scaling to 2": Bang et al. [42] demonstrated that several material systems can be cold sintered at pressures

Target Z (MRayl)	Measured density (g/cm³)	Measured Z (MRayl)	Velocity (m/s)	Attenuation (dB/mm) @ 10 MHz	Electrical Resistivity (Ω-cm)
18	5.8 ± 0.09	$18.7 \pm 0.8$	3300 ± 200	$2.2 \pm 0.7$	0.5
14	$5.3 \pm 0.04$	$14.7 \pm 0.7$	$2800\pm100$	$2.8 \pm 1.0$	0.8
9	$4.5 \pm 0.08$	$9.9 \pm 0.9$	2300±200	$2.6 \pm 0.2$	0.6
5	$2.9 \pm 0.12$	$5 \pm 0.5$	1800± 300	$2.7 \pm 1.3$	0.3

TABLE VI TABLE SUMMARIZING THE ACOUSTIC DATA FOR THE COLD SINTERED ZnO-Ag-PEI COMPOSITES



Fig. 10. Frequency spectrum for a three ML transducer (-6 dB bandwidth > 85%).

below 50 MPa, facilitating promise for commercialization. To enable scaling, in this work, a densification study was undertaken to realize the minimum pressure required to cold sinter half inch diameter ZnO-Ag-PEI composite pellets with composition targeting Z = 18 MRayl at 150 °C with acetic acid as the transient liquid phase. As illustrated in Fig. 8, a minimum uniaxial pressure of 40 MPa is required to fully densify this material system. The acoustic impedance for this sample was found to be 17.9 MRayl.

Next a 2" sample with composition targeting Z = 14 MRayl was cold sintered at 100 MPa on a 150-ton press, achieving pellet relative densities > 95%. The spatial distribution measured on the 2" sample is illustrated in Fig. 9(a)–(d). For a uniform pressure distribution and homogenous particle distribution during sintering, the whole pellet conducts acoustically with an average acoustic impedance of 14.9 MRayl and

attenuation < 3 dB/mm, measured at 10 MHz. The attenuation and inhomogeneity increase with nonuniform pressure application during the CSP.

To demonstrate transducer feasibility, a three-ML 64-element phased array ultrasonic transducer was fabricated by Philips using standard manufacturing techniques and equipment utilized in their factory with minimal process optimization [49]. The first ML consisted of a 2" cold sintered material with acoustic impedance of Z = 19 MRayl, while other two MLs of different Z were made of commercial grade materials. It was observed that (>99%) of the elements on the unit performed well. Fig. 10 shows the measured frequency spectrum magnitude on the transducer. Although there is no significant loop sensitivity improvement observed on this unit, the -6 dB bandwidth is above 85%, which is 10% wider than a two-ML design, demonstrating improved quality and functionality.



Fig. 11. Relation between velocity and Z for cold sintered ZnO-Ag-PEI composites.



Fig. 12. (a) Low magnification backscattered SEM image of a bond-line formed between Z = 7 MRayl and Z = 19 MRayl. (b) High magnification backscattered SEM image of a bond-line formed between Z = 7 MRayl and Z = 19 MRayl.

## B. Improving Acoustic Velocity Using Hydrozincite Based Composites

Based on the acoustic data of the ZnO-Ag-PEI system, a generalized velocity-impedance relationship can be developed for designing MLs in ultrasonic transducers. Fig. 11 gives the relation between velocity and Z for cold ZnO-Ag-PEI sintered composites. At low Z values, the low ZnO + Ag volume fraction produced low acoustic velocity composites. The acoustic velocity for compositions with Z less than 12 MRayl (<2500 m/s) can be improved by ZnO with other low-density ceramics like hydrozincite in the case of nonconducting composites or replacing Ag with low density metals like aluminum in the case of conducting composites. Table VII summarizes the acoustic data for low-Z hydrozincite based composites cold sintered with PTFE. It is noteworthy that although PTFE propagates sound at a low velocity compared to PEI, the ZnO-Ag-PTFE composites velocities for the reported Z range is higher than the PEI counterparts.

Formulation (vol%)	Theoretical density (g/cm³)	Archimedes measured density (g/cm <sup>3</sup> )	Fractional Density (%)	Average measured velocity (m/s)	Measured average Z (MRayl)
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TABLE VII	
TABLE SUMMARIZING THE ACOUSTIC DATA FOR THE COLD SINTERED HYDROZINCITE BASED	COMPOSITES

Target Z=11 MRayl: 90% hydrozincite, 10% PTFE	3.28	3.12	95.1%	3440 ± 120	10.7
Target Z=9 MRayl: 85% hydrozincite, 15% PTFE	3.22	2.13	97%	2930±100	9.14
Target Z=8 MRayl: 80% hydrozincite, 20% PTFE	3.16	3.12	98.7%	2640 ± 50	8.3
Target Z=7 MRayl: 68.5 %, hydrozincite, 31.5 % PTFE	3.02	2.95	97.5%	2370 ± 80	7

TABLE VIII TABLE SUMMARIZING THE ACOUSTIC DATA FOR ML-1, ML-2, AND ML-3

	Material	Acoustic impedance (MRayl)	Thickness (μm)
ML-1	50.5% ZnO, 18.5% Ag, Hydrozincite 31 %	19	200
ML-2	85% Hydrozincite + 15% PTFE	9	165
ML-3	50% Hydrozincite + 50% PTFE	5	110



Fig. 13. (a) Scanning electron image of the graded matching multilayer (b)-(d) EDS mapping of zinc, fluorine, and silver in graded stack, respectively.

# C. Co-Sintering of Different Formulations

1) Demonstration of Bond-Line Formed During the Co-Cold Sintering of Two Different Powder Formulations During Cold Sintering: To assess the feasibility of forming a bond-line without a bond agent between different Z formulations, two different powder compositions corresponding to Z = 7 MRayl and Z = 19 MRayl were cocold sintered. Fig. 12(a) and (b) shows the backscattered SEM imaging illustrating the formation of a bond-line without significant diffusion. Note that the powders were manually packed on top of each other during the process, which accounts for the apparent roughness of the bond-line.

2) Tape Casting of Graded Acoustic ML: Since the entire Z range (5-19 MRayl) can be cold sintered using the same liquid sintering aid, a graded acoustic stack was demonstrated via tape casting using three distinct layers with acoustic impedance Z = 19, 9, and 5 MRayl, stacking them to the desired thicknesses (summarized in Table VIII), and cold sintering in a single step. This strategy could, in principle, allow the entire stack to be cold sintered and isostatically laminated to form the desired curvature. Fig. 13(a) shows the cross section of a graded ML tape-casted stack, illustrating three distinct layers of MLs (1-3), while 13b-d shows the elemental grading for zinc, fluorine and silver measured using EDS. The EDS mapping in Fig. 13(d) shows the presence of Ag in ML-1 corresponding to Z = 19 MRayl with higher fluorine concentration in PTFE rich ML-3 layer, corresponding to Z = 5 MRayl as illustrated in Fig. 13(c). The mapping shows that there is no significant interdiffusion between the MLs; each retained their constituent composition during cold sintering.

This work has demonstrated that a family of scalable graded polymer-ceramic-metal composites can be densified via the CSP using the same transient liquid phase. This feasibility demonstration can have profound implications on improvement in bandwidth and sensitivity for ultrasound transducers using high-end piezoelectric single crystals using graded stacks via the CSP. The three ML transducer demonstration shows promise for manufacturing on an industrial scale.

## V. CONCLUSION

MLs play a crucial role in optimizing the sensitivity and bandwidth response of ultrasonic transducers. The emphasis of this study was to broaden the range of polymer-ceramicmetal composite materials used for acoustic matching, via the CSP due to its low thermal budget. The acoustic properties of the composite agree well with a logarithmic model based on the volume fraction and material properties of the individual phases. The process is scalable to 150 mm due to lower pressure and temperature requirements for sintering this threephase material system. This could enable the optimization of commercial MLs utilized in current generation ultrasonic transducers at a reasonable cost.

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