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Filled and Unfilled Temperature-Dependent Epoxy Resin Blends for Lossy Transducer Substrates

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Abstract

In the context of our ongoing investigation of low-cost 2-dimensional (2-D) arrays, we studied the temperature-dependent acoustic properties of epoxy blends that could serve as an acoustically lossy backing material in compact 2-D array-based devices. This material should be capable of being machined during array manufacture, while also providing adequate signal attenuation to mitigate backing block reverberation artifacts. The acoustic impedance and attenuation of 5 unfilled epoxy blends and 2 filled epoxy blends—tungsten and fiberglass fillers—were analyzed across a 35°C temperature range in 5°C increments. Unfilled epoxy materials possessed an approximately linear variation of impedance and sigmoidal variation of attenuation properties over the range of temperatures of interest. An intermediate epoxy blend was fitted to a quadratic trend line with R^2 values of 0.94 and 0.99 for attenuation and impedance, respectively. It was observed that a fiberglass filler induces a strong quadratic trend in the impedance data with temperature, which results in increased error in the characterization of attenuation and impedance. The tungsten-filled epoxy was not susceptible to such problems because a different method of fabrication was required. At body temperature, the tungsten-filled epoxy could provide a 44 dB attenuation of the round-trip backing block echo in our application, in which the center frequency is 5 MHz and the backing material is 1.1 mm thick. This is an 11 dB increase in attenuation compared with the fiberglass-filled epoxy in the context of our application. This work provides motivation for exploring the use of custom-made tungsten-filled epoxy materials as a substitute PCB-based substrate to provide electrical signal interconnect.

I. Introduction

For several years, researchers at the University of Virginia have been developing low-cost pocket-sized ultrasound imaging systems [1]–[5]. The ultimate objective is to produce a C-scan imaging device using a fully sampled 2-D array with an estimated final sales price of less than \$5000. This objective drives the need for an extremely low-cost 2-D array structure. Central to the low-cost 2-D array is a specialized substrate that simultaneously provides the requisite electrical conductivity to the signal lines of each of the 60×60 transducer elements and provides sufficient acoustic absorption to dissipate rearward propagating ultrasound waves. In the interests of maintaining a low-cost characteristic, we have chosen to base our design on printed circuit board (PCB) technology. Currently, the transducer array is built on a 1.1-mm-thick FR-4 glass-reinforced plastic (GRP) PCB

(Coastal Circuits, Redwood City, CA). Consequently, there is a small but measurable backing block origin echo superimposed on pulse echo signals. This echo is not fully characterized but appears to include both distributed echoes from the woven glass fiber planes and one echo from the discontinuity at the rear surface of the PCB. (Currently, in prototyping, the PCB is air backed, though in the final configuration silicon IC die will be attached directly to the PCB, which will be subsequently potted in epoxy or a similar polymer.) To reduce this “ringing,” a more attenuative material should ideally be substituted for the GRP PCB. It is not practical to solve this problem by simply increasing the thickness of the PCB because of fabrication challenges. Through-hole vias, which allow electrical access to the elements in the current array, are drilled at a standard PCB thickness-to-drill diameter ratio of 10:1 [6]. Therefore, a thicker PCB requires a larger drill to satisfy this requirement. This aspect ratio is in place to minimize the degree to which the drill bit wanders through the PCB thickness. The array pitch constraint in place for this project limits the allowable drill diameter, thereby limiting the maximum PCB thickness to 1.1 mm. This compelled us to find a material with more desirable acoustic properties. Once the material has been finalized, a custom PCB could be fabricated either through collaboration with an existing PCB manufacturer, or through custom machining and photolithography.

Strictly, the material need only be highly attenuative at the transducer operating temperature, which is approximately body temperature due to the transducer’s proximity to the patient’s skin and unavoidable transducer self-heating. Generally materials that are rigid, like a conventional PCB, are relatively low-loss, while pliable materials tend to be more attenuative. However, pliable materials present a challenge to machine because they distort easily under the shear stress imparted by drills, mills, and so on. Therefore, we propose to use a material possessing a low glass transition temperature [7]. Glass transition temperature is the temperature at which a material transitions from behaving like a glass (below the T_g) to behaving like a rubber (above the T_g) [8]. Ideally the T_g should be low enough such that when chilled (in the range 1°C to 20°C), the material may be machined, and when warmed to body temperature, the material becomes significantly more attenuative. While materials that may be machined are of interest, epoxies also lend themselves to molding. A lossy, z -axis conducting material could be assembled, for example, using the method described by Greenstein, which involves filling the gaps in a densely packed array of wires with a lossy polymer [9].

The T_g values of several epoxy resin blends have been previously analyzed in the context of ceramic composite materials [10]. Using these materials as a foundation for our work, similar blends were fabricated and tested. Because the current generation Sonic Window device incorporates a GRP PCB substrate, glass fiber-filled samples were also assembled to emulate the PCB structure. Finally, tungsten particle (10–20 μm powder) filled samples were fabricated because tungsten particle loaded backing blocks are frequently used when a high acoustic impedance, highly lossy backing is desired [11], [12].

We report the dependence of acoustic impedance and attenuation with respect to temperature for all unfilled samples. The unfilled sample determined to possess the most desirable properties was used as a basis for fabricating fiberglass- and tungsten-loaded samples. Acoustic impedance and attenuation data are also presented for these filled samples.

II. Methods

A. Sample Preparation

The epoxy resins DER 332 and DER 736 (Dow Chemical Company, Midland, MI) chosen for these trials were selected based on their temperature-dependent properties as previously measured by differential scanning calorimetry [10]. The former resin forms a hard epoxy when cured, while the latter forms a pliable epoxy. The epoxies were cross-linked using dodecenyl succinic anhydride (DDSA) and the reaction was accelerated using 2,4,6-Tri(dimethylaminomethyl) phenol (DMP-30, Electron Microscopy Sciences, Hatfield, PA). Five different blend ratios were made according to the quantities given in Table I, which also lists the T_g for each blend calculated using the Fox equation [7]. Unfilled epoxy blends will be referred to by their calculated T_g for simplicity (e.g., T_{g110}). For each blend, 3 samples of differing thickness were formed by pouring differing quantities of the epoxy into 3 identical cylindrical molds. The samples were then cross-linked at 60°C on a horizontal surface for 3 h.

To select an unfilled epoxy with which to form filled epoxies, quadratic trend lines were fitted to the attenuation and impedance data. Epoxy T_{g25} was chosen because its attenuation and impedance curves possessed the best fit ($R^2 = 0.94$ and 0.99 , respectively), and because it is the hardest epoxy (at room temperature) that possesses a T_g less than body temperature. Four thicknesses of each filled epoxy were fabricated, yielding samples with the physical properties described in Table II. The fiberglass samples were constructed to emulate a printed circuit board material, and were assembled by layering epoxy-impregnated squares of fiberglass and then applying a pressure of 5 kPa while curing at 70°C for 6 h. The tungsten-filled epoxies were fabricated by mixing 60 g of warmed (60°C) tungsten powder in a 100 mL cylindrical centrifuge tube in 3 equal stages. The powder consisted of 39% 10 to 20 μm particles and 48% 0 to 10 μm particles (with trace quantities of larger particles). Mixing was performed after each tungsten addition using a G-560 Vortex Genie (Scientific Industries Inc., Bohemia, NY) stirrer. The mixture was then cross-linked in an upright position at 70°C for 6 h. Curing in this position allowed the tungsten to settle to the bottom of the container during the early stages of heating, when the epoxy was less viscous due to warming but had not yet started to cross-link. A 1.5-cm-thick cross-section of the resulting tungsten-epoxy mass was sectioned into 4 thin slabs, each of which was then ground to a desired thickness. The ground surfaces were measured to be flat and parallel to $\pm 10 \mu\text{m}$ across each sample's 25.5 mm diameter. The volume fraction of tungsten in the samples may be easily computed using the density of each component—tungsten, unfilled epoxy, and filled epoxy—which was computed from mass and volume measurements. The volume fraction of tungsten was calculated to be 30%. Despite tungsten being electrically conductive, the filled epoxy possessed greater than 1 k Ω impedance magnitude at the frequency of interest (5 MHz) between electrodes separated by 75 μm when compressed under a pressure of 10 MPa (and greater than 5 k Ω when uncompressed), eliminating this as a source of concern. Sample material data are provided in Table II.

B. Experimental Apparatus

All measurements required for attenuation and acoustic impedance calculations were recorded in a temperature-controlled water tank. Prior to recording experimental data, the sound velocity in water was measured at each of the 8 temperatures to be tested, $\pm 1^\circ\text{C}$, thus yielding 24 measurements. These data correlated well with previous work in this area that estimated the sound velocity as

$$c(t) = 1404.3 + 4.7t - 0.04t^2, \quad (1)$$

where t is temperature and c is the speed of sound [13].

Material samples were mounted in an adapter plate press-fit into a mounting bracket, which positioned the sample between and in coaxial alignment with an unfocused 5-MHz piston transducer (V310, Panametrics, Waltham, MA) and a hydrophone (GL-0200, Onda Corp., Sunnyvale, CA). This transducer was chosen based on previous work in the area of acoustic measurements of material properties [14]. The coaxial alignment was achieved by fixing each component of the apparatus—transducer, mounting bracket, and hydrophone—along an optical rail (9731, New Focus, San Jose, CA), which also allowed the distance between components to be set arbitrarily while maintaining alignment. The sample was placed in the far field zone of the transducer. Prior to recording data, each epoxy sample was maintained in the water tank for 5 min to allow its temperature to equilibrate with that of the water bath. Measurements were recorded at 5°C increments over the range 5 to 40°C . A 50-cycle 5 MHz waveform was programmed on an arbitrary waveform generator (AWG2021, Sony/Tektronix, Beaverton, OR). This waveform was used to determine both speed of sound and attenuation. The signal output of the AWG was connected to a 55 dB RF amplifier (A150, ENI, Rochester, NY), which drove the piston transducer at 100 V peak amplitude. The signal received by the hydrophone was first amplified 17 dB (A17dB, Specialty Engineering Associates, Sunnyvale, CA) followed by a further 45 dB (5676, Panametrics). The resulting signal was sampled at 500 MHz and averaged 1000 times by the digitizing oscilloscope (LC334A, LeCroy, Chestnut Ridge, NY) to increase final signal SNR.

C. Data Analysis

Fig. 1 illustrates schematically the signal pathways through 2 epoxy samples differing only in thickness, helping to describe the method employed to derive acoustic impedance and attenuation. It is assumed that any given epoxy will have a consistent speed of sound across its sample thicknesses. This speed of sound differs from that of the surrounding water, and therefore each thickness sample causes a temporal shift in the received signal that is a function of its thickness and speed of sound. This is illustrated in Fig. 1 by the spatial shift of the second pressure wave (i.e., advancement of wave due to substitution of water path length by higher velocity sample path length).

Acoustic impedance was computed by first cross-correlating the received signals for each of the thicknesses of a given material to determine the time delay associated with a measured difference in thickness. It was then feasible to generate an estimate of the speed of sound

through the material sample. Using this technique, $\sum_{x=1}^N (x - 1)$ estimates of sound velocity, where N is the number of thickness samples, may be computed. Once speed of sound was determined, the density of the sample was measured and the acoustic impedance was calculated.

Similarly, 3 attenuation measurements were computed. This calculation is based on the differential received signal amplitude between sample thicknesses. This amplitude difference is illustrated in Fig. 1 by the line widths of the received echo pulses. Attenuation may be measured separately from interface reflections using this method because the reflections are constant. This is because only material thickness is altered while acoustic properties are held constant. Therefore, the differential calculation causes the constant reflection term to be eliminated in the final calculation. It is assumed that the attenuation of water is 0.05 dB/cm at the frequency of interest [15]. Though, to a small degree, standing waves may be generated within the sample, this does not impact the experiment as it is only forward-propagating echoes that are measured. Interference from multipath echoes may also be neglected due to attenuation within the samples and the energy loss resulting from the 2 material interface reflections. Signal amplitude was derived from the steady-state (i.e., midpulse) peak-to-peak amplitude.

III. Results

Fig. 2 illustrates the relationship of impedance with respect to temperature for each of the 5 unfilled epoxy samples fabricated. The mean impedance amplitude for each temperature is illustrated with circles, each with an accompanying error bar of $\pm$ (standard deviation). The trend line drawn on each plot was fitted to the complete set of data, comprising 3 impedance estimates at each temperature. It is observed that, while the trend line associated with each data set is nearly linear, the impedance of epoxy T_g25 possesses the minimum quadratic component among the epoxies, and the maximum R^2 value of 0.99. The acoustic impedance of all blends illustrated here is an order of magnitude less than that of the PZT5H used in the Sonic Window, which is approximately 30 MRayl. This mismatch in impedance will cause a large reflection to be generated from the PZT-substrate interface, resulting in a transmit pulse with ringing.

Fig. 3 illustrates the attenuation results for the 5 unfilled epoxies. The data are presented in the same format as those above. Again, epoxy T_g25 possesses the best match to the quadratic trend line, with an $R^2 = 0.94$. The quadratic coefficient term decreases, becoming negative, with decreasing T_g . This correlates well with previous findings that, over a wider temperature range, a sigmoid-shaped attenuation curve is observed and is centered around the material's T_g . To explain further, the attenuation data (Fig. 3) are plotted over the temperature range 0 to 40°C. Therefore, the temperature range shown for the T_g110 epoxy data are much below its T_g value. As epoxies of decreasing T_g are considered, the range of plotted temperatures becomes closer to the material's T_g , and eventually spans the T_g . The decreasing quadratic coefficient term with decreasing T_g indicates that the curves transition from concave-up to concave-down, implying a sigmoid shape, as found in previous work [16]. Fig. 4 illustrates this comparison by plotting, on a normalized scale, the previously

published data (adjusted for frequency of measurement using the William-Landel-Ferry equation [8]) alongside the data represented in Fig. 3.

Fig. 5 illustrates the acoustic impedance results for the filled epoxy materials, while Fig. 6 illustrates the attenuation data for the same material. The large error bars on both the impedance and attenuation plots for the fiberglass-loaded epoxy are most likely attributed to heterogeneity within the sample composition. This material possesses an inherent surface and interlayer texture that arises from the roughness of the fiberglass weave. Additionally, because of the weave, there is some potential for trapping of air within the sample. These are the possible causes of nonlinearity in the impedance performance of the fiberglass sample, since the unfilled epoxy used to form these samples has a very linear relationship between impedance and temperature. Additionally, the impedance plot for the tungsten sample is much more linear than that for the fiberglass sample and exhibits better agreement between the measurement estimates. The 10 to 20 μm particle size of this metal powder is an order of magnitude smaller than the wavelength of the 5 MHz pressure wave, and therefore can be considered as a Rayleigh scatterer. Also, the tungsten fabrication process allows less room for trapping air, as sedimentation of the tungsten powder during epoxy heating results in a densely-packed tungsten-epoxy mass after cross-linking.

The tungsten-filled sample yields promising results. The acoustic impedance in the range of body temperature (37°C) for this material is approximately 15 MRayl, the highest of any of the tested materials. Attenuation at body temperature is approximately 20 dB/mm. In the intended application, in which the substrate material is 1.1 mm thick, this would provide 44 dB round-trip attenuation of the backing block echo at 5 MHz. This represents an increase in attenuation of 27.5 dB at 37°C compared with 5°C, 26.4 dB compared with unfilled epoxy at 37°C, and 11 dB compared with the fiberglass-filled epoxy at 37°C in the context of a 1.1 mm backing material.

IV. Conclusion

We have successfully demonstrated that formulating blends of DER 332 and DER 736 epoxies with low T_g values results in materials whose acoustic impedance and attenuation vary significantly over the range of temperatures of interest. These materials may either be machined at chilled temperatures when they are more rigid, or may be set in a mold to form a z -axis conductive backing material. The former technique is closely related to the existing method of establishing electrical interconnect using a GRP PCB.

Loaded epoxies were fabricated using fiberglass and tungsten as filler materials. The addition of fiberglass to the original epoxy material disrupts the approximately linear relationship to temperature possessed by the unfilled epoxies in terms of both impedance and attenuation. The error observed in the data obtained for the fiberglassfilled sample may be related to surface roughness of the material sample caused by the fiberglass weave and small quantities of trapped air. These results correlate well with our previous experience with the use of GRP PCB materials as transducer substrates, in which we observe a measurable echo originating from the backing material.

Conversely, the addition of tungsten does not have such an effect, and also serves to greatly increase the material's attenuation and acoustic impedance. This will reduce the round-trip backing block echo by an estimated 26.4 dB compared with unfilled epoxy and 11 db compared with fiberglass-filled epoxy, and improve the acoustic match to PZT, respectively, indicating that it may be a superior choice of filler material for our application. The findings in this work provide motivation for exploring the use of custom-made tungsten-filled epoxy materials as a substitute PCB substrate.

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Fig. 1.

Schematic of experimental measurement of acoustic impedance and attenuation. The thicker sample (2) will attenuate the signal more, and cause a greater time shift than the thinner sample (1).

Fig. 2.
Impedance of unfilled epoxies versus temperature.

Fig. 3. Attenuation versus temperature of unfilled epoxies. Plots A–E illustrate results for T_g110 , T_g43 , T_g25 , T_g17 , and T_g12 epoxies, respectively.

Fig. 4. Comparison of experimental data with published data. All plots are adjusted by the appropriate T_g .

Fig. 5.
Impedance data for A) fiberglass- and B) tungsten-filled epoxy materials.

Fig. 6.
Attenuation data for A) fiberglass- and B) tungsten-filled materials.

TABLE I

Unfilled Epoxy Composition.

Calculated T_g (°C)	Chemical Quantities (g.)			
	DER 332	DER 736	DDSA	DMP-30
110	14.0	0.0	6.0	0.27
42.8	10.5	2.5	6.0	0.27
25.0	7.0	5.0	6.0	0.27
16.7	3.5	7.5	6.0	0.27
12.0	0.0	10.0	6.0	0.27

TABLE II

Filled Epoxy Sample Geometry.

Quantity	Epoxy sample filler material		
	Unfilled	Fiberglass	Tungsten
Thicknesses (mm)	2.06	0.35	0.91
	3.65	0.54	1.49
	4.95	0.70	1.95
	6.47	1.39	2.22
Mass (thickest only) (g)	5.75	1.75	7.79
Diameter (mean) (mm)	31.2	30.5	25.6
Density (mean) (mm)	1205	1714	6674