Alumina/Epoxy Nanocomposite Matching Layers for High-Frequency Ultrasound Transducer Application

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***Abstract*—Mismatch of acoustic impedance at the interface between a piezoelectric transducer and the medium to be probed will substantially reduce the amount of ultrasound energy being transmitted into the medium. Therefore, matching layer is a critical component of an ultrasonic transducer. A spin-coating process was used to fabricate alumina/polymer nanocomposite films with alumina volume fractions ranging from 14 to 32%. The particle size of alumina is in the range of 10 to 40 nm. The thicknesses of the matching layer can be controlled by the spinning speed and the concentration of solution. Acoustic impedances of these nanocomposite matching layers are in the range of 2.8 to 5.1 MRayls with different alumina contents, which meet the matching layer requirement. The attenuation of a nanocomposite matching layer with smooth surface is about 15 dB/mm at 40 MHz. The pulse-echo spectrum and frequency spectrum of a high-frequency transducer using this nanocomposite matching layer are reported.**

I. Introduction

Quarter-wavelength acoustic matching layers between the piezoelectric ceramic and the propagating medium are critical components that will affect to a great deal the overall performance of ultrasonic transducers, including sensitivity and bandwidth. To improve the resolution of ultrasonic imaging, there have been continuous efforts to push for higher and higher operating frequencies because the resolution is roughly inversely proportional to the wavelength of the ultrasonic wave. In recent years, ultrasonic imaging using frequencies higher than 50 MHz has found applications in eye and skin imaging [1], [2] However, because the piezoelectric transducer elements usually have very large acoustic impedance (>30 MRayls) while the bodies to be imaged have very low acoustic impedance (<1.6 MRayls), the acoustic energy transmission efficiency is rather low.

The acoustic mismatching problem can be resolved using 1 or 2 quarter-wavelength matching layers, and the required matching layer materials usually should have acoustic impedance in the range of 3 to 14 MRayls. There is no such single-phase material in nature. Typically, these matching layers are made of metal particle-polymer composites for lower frequency transducers. Unfortunately, they cannot be used for high-frequency transducers because the thickness of the quarter-wavelength matching layer becomes only a few microns [3]. For a 100 MHz PZT ultrasound transducer, the matching layer thickness is less than 8 μm, while the particle materials available are usually greater than 1 micron in diameter so that there will be only a few particles across the thickness dimension, making it impossible to obtain a uniform material to serve the purpose. In addition, relatively large particle size will cause severe attenuation due to scattering. It is very difficult to obtain a homogeneous composite thin matching layer with high powder loading using submicron powders, and it is also very difficult to lap down the material to a desired thickness. Therefore, the conventional mixing process using metal particles dispersed in a polymer matrix cannot be used for the fabrication of high-frequency composite matching layers with homogeneous properties.

In a previous work, the acoustic properties of silicon oxide colloidal/polymer composite films and alumina colloidal/polymer nanocomposite films have been briefly reported in [4] and [5]. In this work, we will report the spin-coating process that has been used to fabricate the alumina/polymer nanocomposite films with alumina volume ratios ranging from 14 to 32%. The thickness of the matching layers is controlled by the spinning speed and the concentration of solution. Characterized properties of a $LiNbO_3$ high-frequency transducer over 40 MHz using only one nanocomposite matching layer are also reported.

II. Materials Preparation

High-purity gamma alumina powder (CR125) was obtained from Baikowski International Corporation (Charlotte, NC). The average particle size of nano alumina powder is about 30 nm. However, the nano powders are usually agglomerated to a porous mosaic. To disperse nano-Al_2O_3 powder in matrix polymer, which is epoxy with part A and part B, powders were ball-milled first in the presence of a surface coupling agent, 3-aminopropylsilane (3-APS), which modifies the surface of Al_2O_3 powders and couples the powders and matrix polymer. In this system, the weight ratio of part A to part B is 4 to 1 except as noted.

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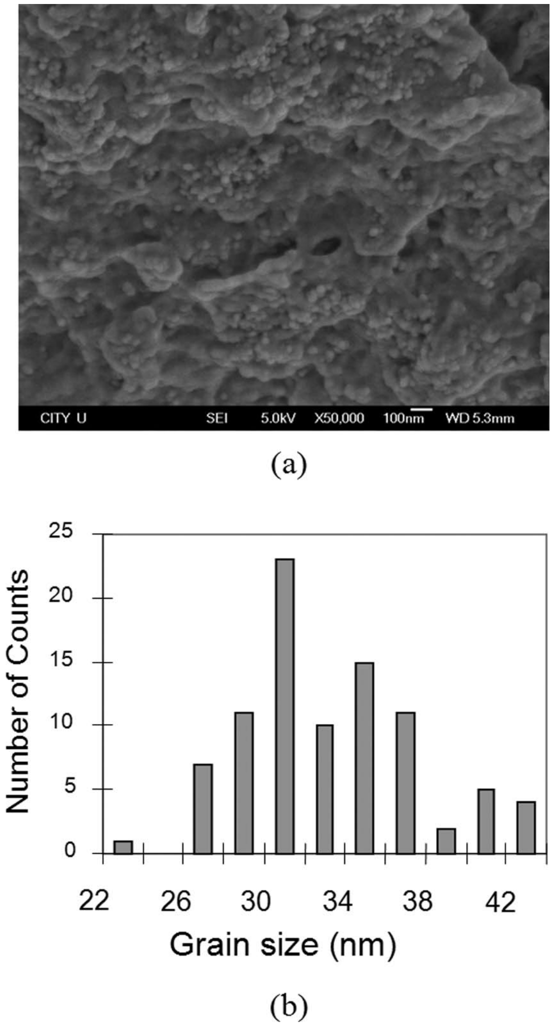

Fig. 1. (a) A cross-sectional SEM micrograph of alumina/epoxy nanocomposite matching layer and (b) the particle size distribution of the nanocomposite.

Ethanol was used as solvent, and zirconia balls were used as milling media. The weight ratio between powders and zirconia balls ranged from 1:2 to 1:5. The particle size distributions before and after 2 d of milling were examined using a Nicoma Particle Size System. The result showed that the size of the agglomerate particles was evidently reduced to nanometer scale; many soft-bonded agglomerate particles were broken after the milling process. Hybrid coating solutions are formulated with colloidal sol and matrix polymer. In this work, the alumina concentration of the matching layer was prepared to have different volume fraction ratios. Epoxy (EPO-TEK 301, Epoxy Technology, Billerica, MA) was chosen as the matrix polymer because it provides easy solidification. It contains reactive groups, such as hydroxyl and glycerol. These groups may react with the hydroxyl group on the surface of alumina oxide powder to form a chemical bond. The hydroxyl group on the surface of nano-alumina condensed with polymer to form a homogeneous nanocomposite. An adhesion promoter (3-aminopropylsilane) was added to ensure good adhesion to the substrate. Finally, ethanol was added into the solution to achieve the desired concentration.

The matching layers were deposited by a spin-coating process. The coating solution was applied onto silicon substrate by spin-coating at different speeds from 30 s up to 1 min. The sample was then placed into an oven to evaporate the solvent (ethanol). The process was repeated to achieve the desired thickness after placing the sample into oven for aging in a desired period in each coating layer process. The annealing temperatures were tested from 40 to 90°C. Fig. 1(a) shows a cross-sectional SEM micrograph of alumina colloidal with epoxy nanocomposite films. It was revealed that the alumina colloidal particles were homogeneously distributed in the composite, which was impossible to obtain by using conventional composite fabrication methods. Fig. 1(b) shows the particle size distribution of nanocomposites. The average particle size is about 30 nm in the nanocomposite.

For testing the performance of the matching layer, a high-frequency (over 40 MHz) single-element transducer was fabricated using $LiNbO_3$ (LNO) single crystal disk [6]–[8]. A 115 μm thick original piece was lapped to 75 μm in thickness, E-solder 3022 (VonRoll Isola, New Haven, CT) was cast on one side of the LNO as backing material, and the backing layer was cured at 40°C for 3 h then lapped down to under 3 mm thick. The E-solder also serves as the bottom electrode. Active element plugs were diced into circular shapes as the aperture of the transducer and the whole assembly was housed using Epotek 301 (Epoxy Technology Inc., Billerica, MA) with copper tubing. An electrical connector was fixed to the conductive backing using a conductive epoxy. The top electrode was sputtered on after the transducer was put into the housing. A nano-matching layer with 32% alumina powder/epoxy was deposited on the surface of the transducer by using spin-coating with suitable thickness. The transducer was then put into an oven to cure at a temperature of 60°C.

A single-element transducer was modeled using PiezoCAD (Woodinville, WA), a commercial transducer modeling software. The electrical impedance of the sample was measured using the HP4194A impedance analyzer (Agilent Technologies, Englewood, CO). The transducers were tested in a deionized water bath in pulse/echo mode by reflecting the signal off an X-cut quartz target placed at the focal point. For pulse/echo measurements, transducer excitation was achieved using a Panametrics (Waltham, MA) model 5900 PR pulser/receiver. The reflected waveforms were received and digitized by a 500-MHz LC534 Lecroy (Chestnut Ridge, NY) oscilloscope.

The experimental setup for the ultrasonic measurement of the matching layer is shown in Fig. 2(a) and (b). Two immersion-type broadband transducers (Panametrics V358) with center frequency of 40 MHz [see Fig. 2(b)] and a one-way −6 dB bandwidth of 75% were used. The transmitting transducer was driven by a 200 MHz computer-controlled pulser (Panametrics 5072PR), and the signal from the receiving transducer was sampled using a digital oscilloscope (TDS 460A; Tektronix, Inc., Beaverton, OR).

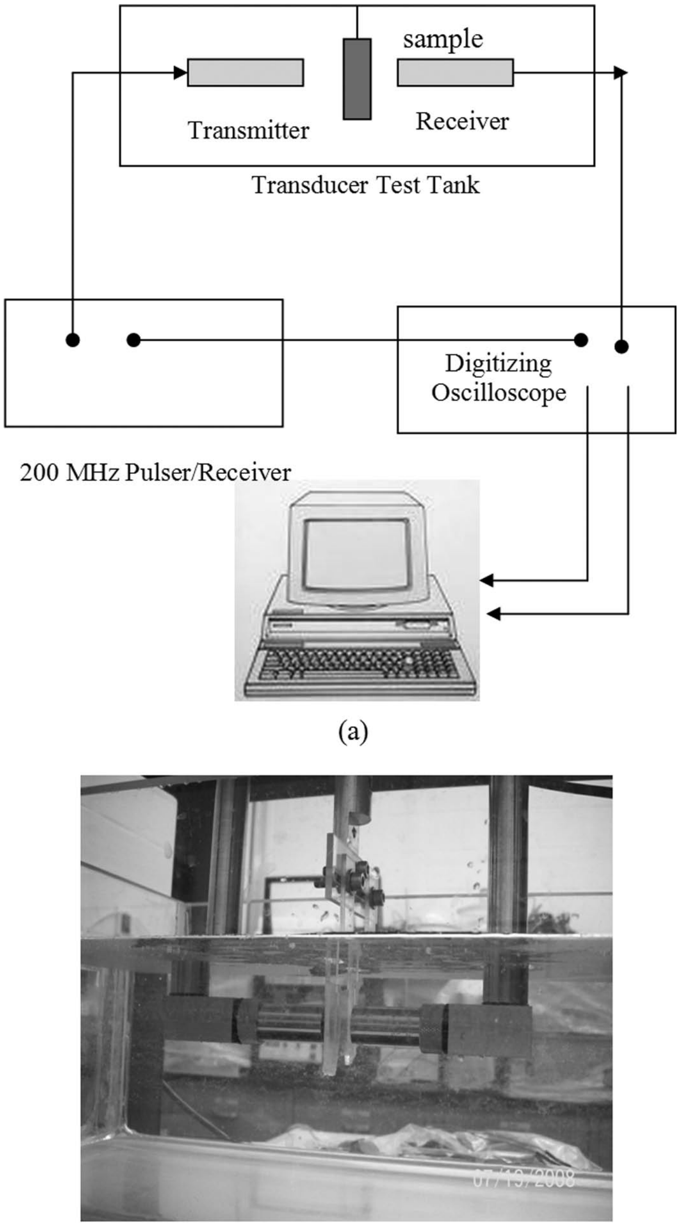

Fig. 2. Experimental setup for ultrasonic testing.

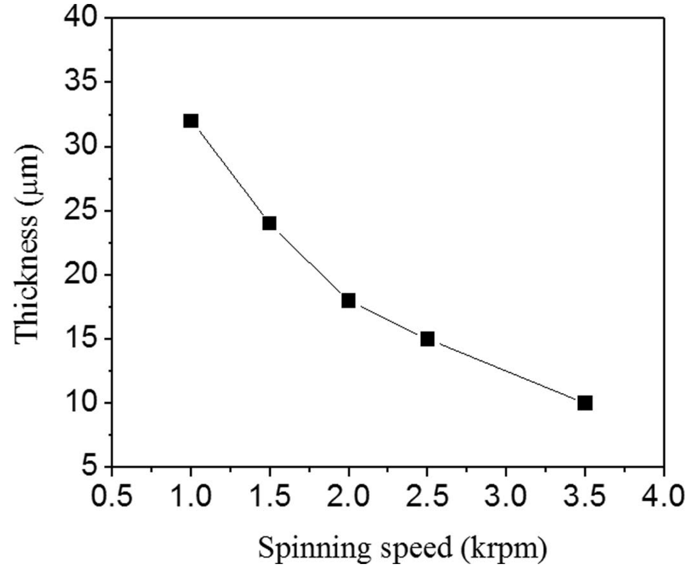

Fig. 3. The thickness of films with different spinning speeds for 32% Al_2O_3 nanopowder/epoxy composites deposited on silicon substrate.

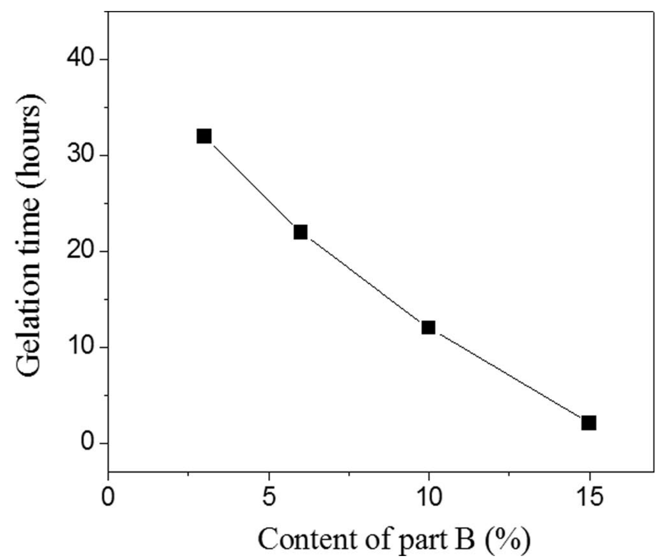

Fig. 4. The solidifying time of films with different amounts of part B cured at 80°C.

The sampling rate was set to 10 Gs/s to avoid aliasing. Each sampling window contained 2500 data points [7]. To reduce ubiquitous random errors, each measurement was averaged 64 times. The data was transmitted to a personal computer where the fast Fourier transform was performed. To test the effectiveness of the matching layer, a 42 MHz single-crystal lithium niobate transducer with a nano-matching layer was fabricated, and properties were tested and compared [8], [9].

III. Experimental Results

As described above, the key technology of the nanocomposite matching layer is to prepare low-temperature-cured coating solutions that contain nanoparticles and polymer phase. The low-temperature curing process is very important to avoid any grain growth and de-poling of the transducer. It is also very important to control the viscosity during the mixing so that the coatings can be made uniform, both in terms of particle distribution and in layer thickness. The spin-on coating process is a well-established fabrication technique that has been used for many other applications. By using a stable hybrid solution and maintaining constant coating parameters, coating thickness and thickness uniformity can be controlled by the spinning speed on a flat substrate. In general, if other parameters are fixed, the coating thickness is inversely proportional to the spin speed. Of course, the properties

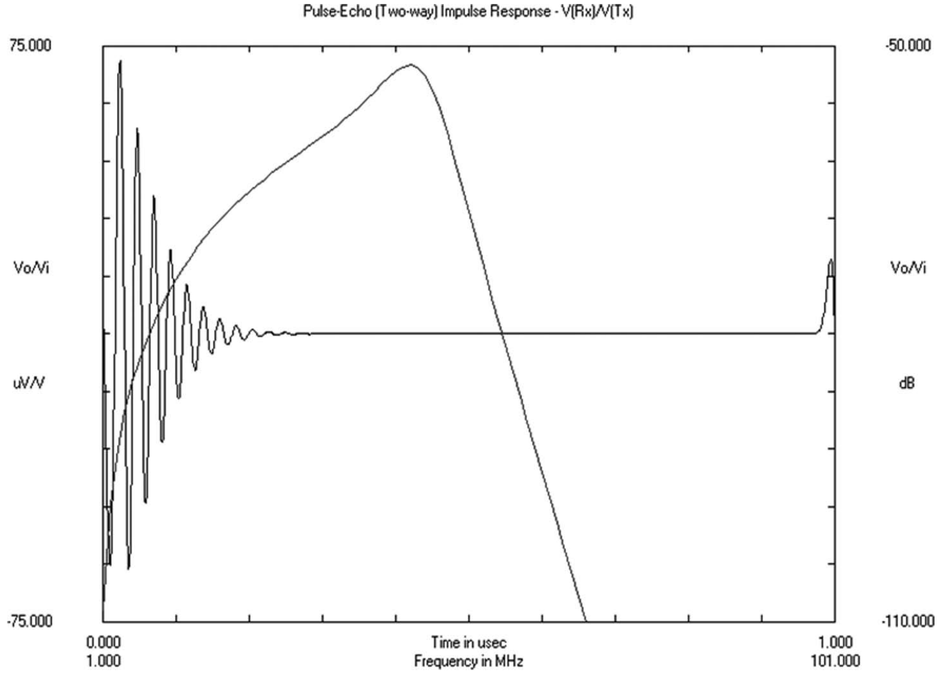

Fig. 5. Pulse-echo waveform (voltage) with times (μs) and magnitude spectrum (dB) with frequencies (MHz) for a high-frequency LNO transducer by PiezoCAD modeling.

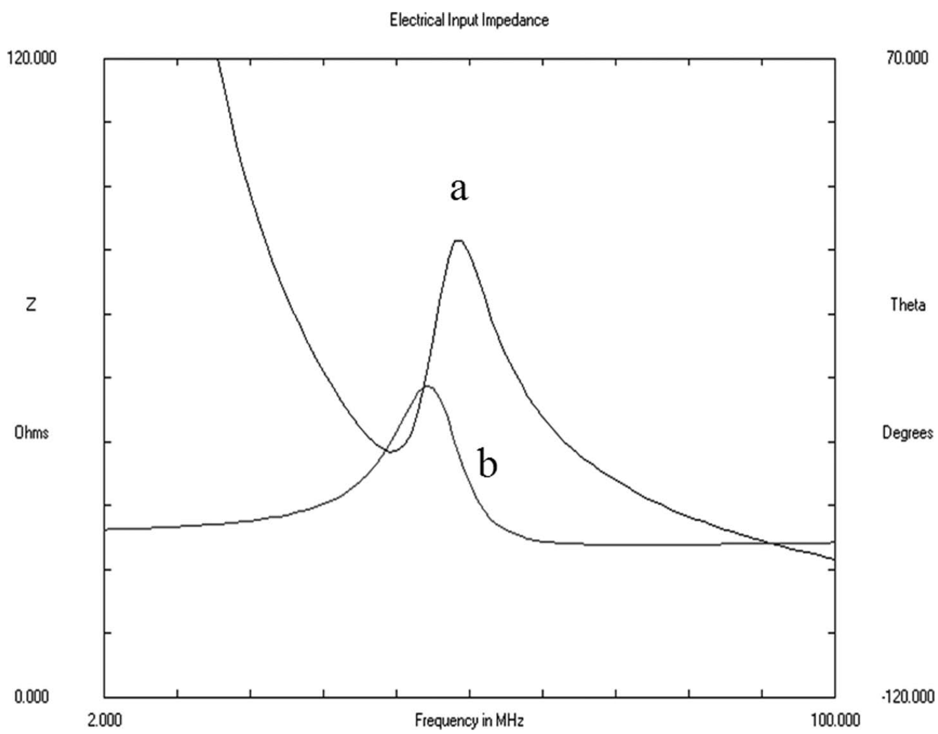

Fig. 6. (a) The electrical impedance and (b) phase spectrum of the transducer with different frequencies.

of coating solutions, i.e., the viscosity and density, also have a strong influence on the coating thickness control. Fig. 3 shows the thickness of films with different spinning speeds for 32% (volume ratio) Al_2O_3 nanopowder/epoxy composite deposited on a silicon substrate. It shows that the thickness decreases with increasing speed at same concentrations. For example, a thin matching layer around 10 μm thick can be achieved by using high spinning speed at 3500 rpm. Therefore, these results suggest that the thickness can be controlled by spinning speed of the coating machine. Zheng *et al.* have successfully controlled the thin films thickness for optical reflect layers of glasses using spin-coating technology [10]. Fig. 4 shows the solidifying time of films with different doping amount of part B in

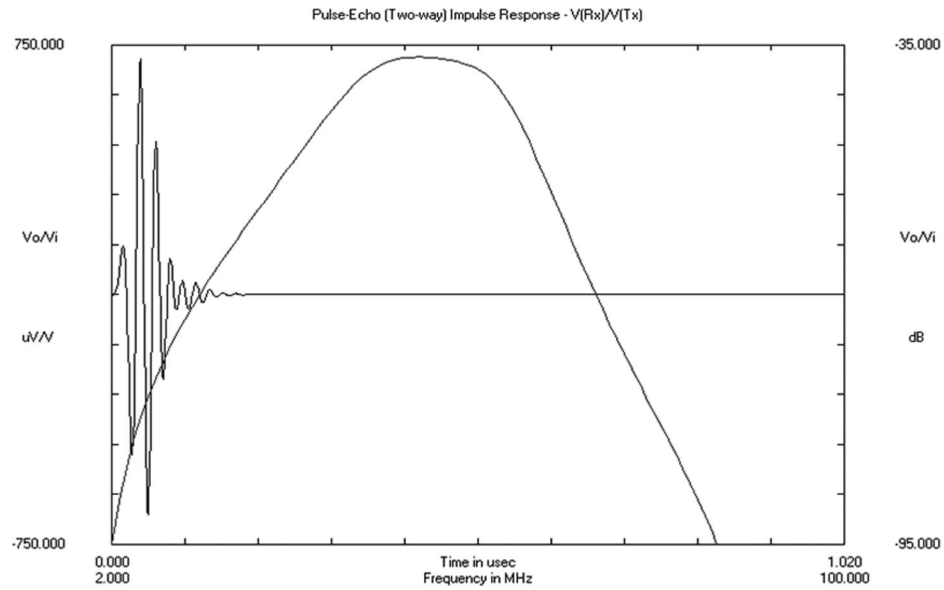

Fig. 7. PiezoCAD modeling of the pulse-echo waveform (voltage) with times (μs) and magnitude spectrum (dB) with frequencies (MHz) for the transducer with nanocomposite layer and parylene.

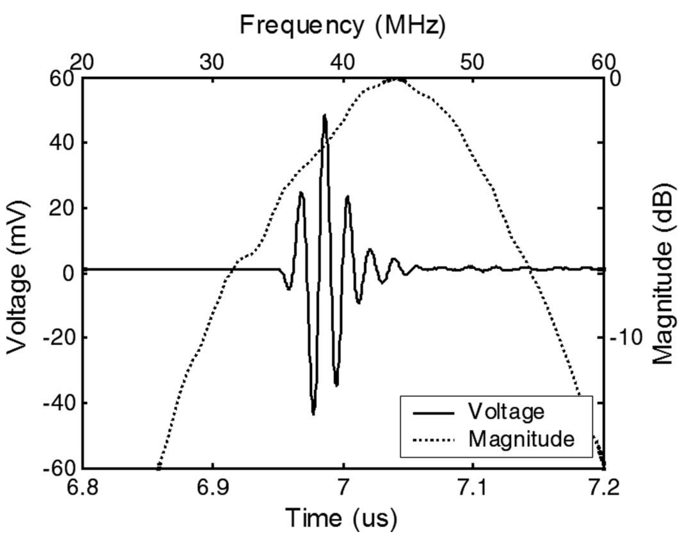

Fig. 8. The pulse-echo waveform (voltage) with times (μs) and magnitude spectrum (dB) with frequencies (MHz) for LNO transducer with nanocomposite layer and parylene.

epoxy cured at 80°C (sample thickness is 13 μm). The gelation time of composite films decreases with increasing content of part B of the curing agent.

The density and acoustic phase velocity of nanocomposite films as a function of the alumina volume ratios were measured. It was found that the film density increases from 1150 Kg/m^3 to 1630 Kg/m^3, and acoustic velocity increases from 2500 m/s to 3200 m/s as the alumina volume ratio increases from 14% to 32%. In the volume fraction range of 14 to 32%, the acoustic impedance appears to be linearly dependent on the particle volume fraction. The acoustic impedance of nanocomposite films increases nearly 82% from 2.8 MRayls to 5.1 MRayls by increasing the alumina ratios from 14% to 32% [5]. In addition, the attenuation of the nanocomposite matching layer was also measured, which is more dependent on the surface smoothness of the sample and quality of the coating layer. The sample with a smoother surface has lower attenuation coefficient of about 15 dB/mm at the frequency of 40 MHz, which is much lower than that of conventional ceramic composites [7]

Modeling results of the $LiNbO_3$ high-frequency single-element transducer by using PiezoCAD are shown in Fig. 5 for the case without matching layer. E-solder was used as the backing material. The center frequency of the transducer without matching layer is around 42 MHz with a fractional bandwidth of 35% at −6 dB. For this design, the aperture size of transducer is about 4 mm. Fig. 6 shows the electrical impedance spectrum (modeling) with frequencies. The electrical impedance of transducer is about 55 Ohms at resonant frequency, which matches electric circuit requirement (50 Ohms). Fig. 7 shows the piezoCAD modeling result of the pulse-echo spectrum for the transducer with quarter wavelength nanocomposite layer and parylene. The bandwidth of the transducer is about 54% at −6 dB. Fig. 8 shows the experimental results of a $LiNbO_3$ transducer with the nanocomposite matching layer deposited by spin-coating and parylene. The bandwidth of the transducer at center frequency is about 51% at −6 dB. The experimental result of bandwidth is a little lower than that of modeling, which may be caused by the variation of matching layer thickness. By adding a nano-matching layer fabricated using the process described in this work, the bandwidth has been evidently increased to 51% from 35%. The work is ongoing to further increase the acoustic impedance up to 7 MRayls of the nanocomposite matching layer to better match $LiNbO_3$ single crystal to achieve even broad bandwidth.

IV. Conclusion

A spin-coating method is developed to fabricate thin alumina colloidal/polymer nanocomposite films, which can be used as quarter-wavelength matching layers for high-frequency ultrasonic transducers. A few 13-μm-thick film samples were prepared with the average particle size of alumina colloidal particles of approximately 30 nm. The measured results showed that the acoustic impedance of nanocomposite can be increased from 2.8 MRayls to 5.1 MRayls when the alumina volume fraction was increased from 14% to 32%. This nanocomposite fabrication technology may provide a solution to resolve the poor matching problems currently existing in high-frequency medical ultrasonic transducers.

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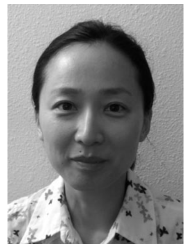

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