Letters

Acoustic Properties of Alumina Colloidal/Polymer Nano-Composite Film on Silicon

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***Abstract*—Alumina colloidal/polymer composite films on silicon substrates have been successfully fabricated using the sol-gel method, in which the crystallite sizes of alumina are between 20 and 50 nm. The density and ultrasonic phase velocities in these films with different alumina ratios from 14% to 32% were measured at the desired operating frequency. We have proved that the density, acoustic phase velocities, and hence the acoustic impedance of the nano-composite films increase with the alumina content, which gives us another option of tailoring the acoustic impedance of the nano-composite film for making the matching layer of high-frequency medical ultrasonic transducers.**

QUARTER-WAVELENGTH acoustic matching layers between the piezoelectric material, whose acoustic impedance is usually ∼30 Mrayls, and the propagating medium, whose acoustic impedance is ∼1.5 Mrayls, are critical for the performance of broadband biomedical ultrasonic transducers. Nowadays, most of the matching layers are made of composite materials, because single-phase materials that meet the need of the acoustic impedance requirements do not exist in nature. By mixing the high-impedance powder and low-impedance polymer, the acoustic impedance of the composite can be easily tailored by varying the volume fractions of the components in the composite to meet the design requirements [1].

Compared to those transducers with lower center frequency, high-frequency ultrasonic transducers can provide higher resolution in both axial and lateral directions, resulting in improved imaging quality for the diagnosis of many diseases and in better monitoring of medical treatments [2], [3]. The quarter-wavelength acoustic matching layers become very thin at high frequencies. For a 100-MHz transducer made of $Pb(Zr,Ti)O_3$ (PZT) piezoceramics, the matching layer thickness is less than 5 μm. Generally, silver/epoxy composites are used as the first matching layer by lapping down the material to a desired thickness [4]; however, it is difficult to obtain a homogeneous thin matching layer using 1.5-μm silver particles. Therefore, the conventional processing method of hand mixing metal particles with polymer matrix becomes very challenging for the fabrication of high-frequency composite matching layers with homogeneous properties.

In this letter, we report the successful fabrication of an alumina colloidal/polymer composite film using the sol-gel coating process [5]. The particle size in the film is less than 50 nm and the uniformity of the properties is guaranteed even for submicron-thickness film. The acoustic properties of the nano-composite films with alumina content from 14% up to 32 wt % were measured at the operating frequency of the transducers, and it was found that the acoustic impedance is in the desired range.

The fabrication process is as follows. Alumina colloidal was mixed with polymer in the presence of a coupling reagent and water. The hydroxyl group on the surface of the nano-alumina condensed with polymer to form a homogeneous nano-composite. Adhesion promoter was added to ensure good adhesion to the substrate. Ethanol was added to achieve the desired concentration of adhesion promoter. The desired concentration was determined by the ratio between the adhesive and ethanol. The mixture solution was deposited onto silicon substrate by spin coating at 2000 rpm for 30 seconds. The substrate was then placed on a hot plate to evaporate the solvent. Each film layer thickness was about 6–8 μm, and the sample was placed in an oven to age for a time period after each coating. The coating process was repeated until the desired thickness was achieved. Five samples with a thickness of 24 μm were prepared, and the alumina contents in these samples were 14%, 18%, 28%, 30%, and 32%, respectively.

The cross-section scanning electron micrograph of the film shown in Fig. 1(a) revealed that alumina colloidal crystallites were homogeneously distributed in the composite, which was impossible to obtain using conventional mixing methods. The film surface is perpendicular to the white arrow in this figure. There were no observable layer interfaces on the micrograph between different coatings. The average alumina crystallite size is around 32 nm, with distribution primarily between 20 and 50 nm as shown in Fig. 1(b).

The thickness d at the corresponding operating frequency f of the polymer nano-composite films can be determined by using the method described in [6]. The experimental setup for the ultrasonic measurement is shown in Fig. 2. Two immersion-type broadband ultrasonic transducers (Panamatrix V358; Panametrics, Waltham, MA) with a center frequency of 50 MHz and a one-way −6 dB bandwidth of 75% were used for the measurements. The transmitting transducer was driven by a 200-MHz computer-controlled pulser (Panametrics 5052PR),

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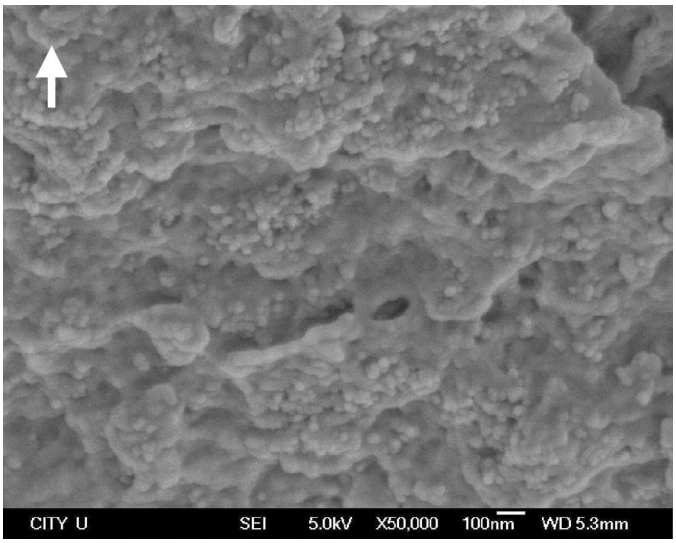
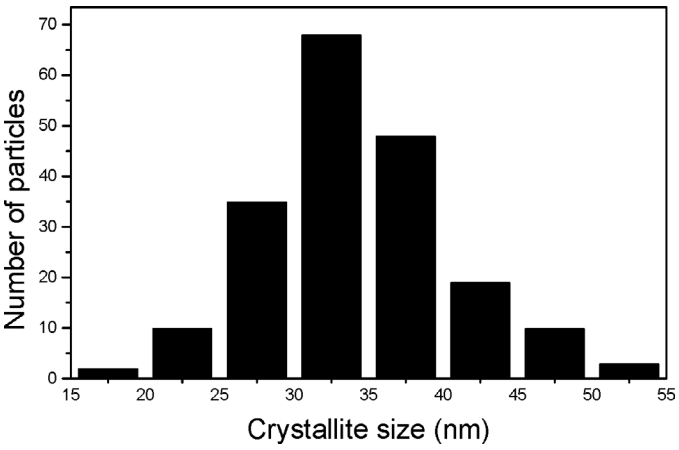


(a)



(b)

Fig. 1. (a) Cross-section SEM micrograph of alumina colloidal/polymer nano-composite films and (b) crystallite size distribution.

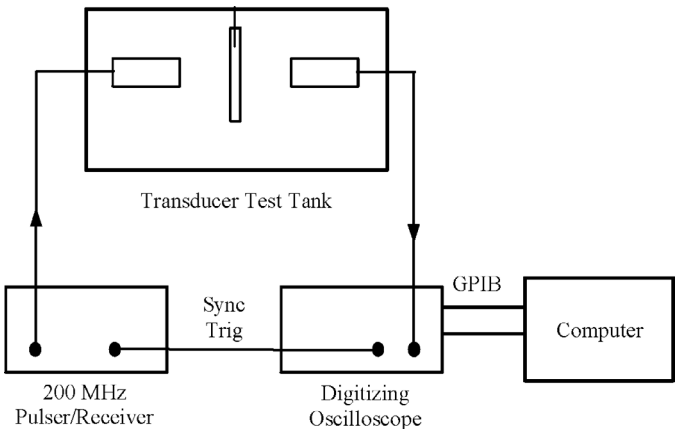


Fig. 2. Experimental setup for ultrasonic measurements.

and the signal from the receiving transducer was sampled using a digital oscilloscope (Tektronix TDS 460A; Tektronix, Inc., Beaverton, OR) with a sampling rate of 10 Gs/s to avoid aliasing. Each sampling window contained 2500 data points. Each measurement was averaged 64 times in order to reduce ubiquitous random errors. The data were transmitted to a personal computer where fast Fourier transform (FFT) was performed.

Two signals, $g(t)$ (with the nano-composite film on the Si substrate in between the two transducers) and $h(t)$ (with only the Si substrate in between the transducers), were recorded. The ratio of corresponding Fourier amplitude spectra were obtained, which is related to the material properties as follows:

$$\left|\frac{G^*(f)}{H^*(f)}\right| = \frac{T_{23}T_{31}}{T_{21}} \frac{e^{-(\alpha-\alpha_1)d}}{\left|1 - R_{31}R_{32}e^{-2\alpha d}e^{-i2\beta d}\right|}, \quad (1)$$

where $\beta = 2\pi f/\nu$ is the wave number in the film, α and α_1 are the attenuation coefficients in the film and in water, respectively, and d is the thickness of the film. $R_{ij} = (Z_j - Z_i)/(Z_j + Z_i)$ is the reflection coefficient in medium i from medium j, $T_{ij} = 2Z_j/(Z_j + Z_i)$ is the transmission coefficient from medium i into medium j, and Z_i is the acoustic impedance of medium i. In our experiments, media 1, 2, and 3 were water, substrate, and nano-composite film layer, respectively. Since $R_{31}R_{32}$ is negative when the acoustic impedance Z_i satisfies $Z_1 < Z_3 < Z_2$, the maximum value of $|G^*(f)/H^*(f)|$ occurs when

$$\beta d = (n + 1/2)\pi, \quad n = 0, 1, 2, \ldots \quad (2)$$

From (2), we get the phase velocity of the thin layer at the corresponding frequency $f_{\max,0}$ when $n = 0$:

$$\nu(f_{\max,0}) = 4f_{\max,0}d. \quad (3)$$

The frequency $f_{\max,0}$ is exactly the designed operating frequency of the transducer because the thickness d equals $\lambda/4$ of the wave propagating in the film at this frequency.

The density of alumina colloidal/polymer composite film ρ_{film} can be calculated straightforwardly by

$$\rho_{\text{film}} = \frac{M - \rho_{si} * S * h_{si}}{(G/\rho_f) - S * h_{si}}, \quad (4)$$

where M and S are the total mass and area of the film and silicon substrate, respectively; ρ_{si} and ρ_f are the densities of silicon substrate and immersion fluid, respectively; h_{si} is the thickness of silicon substrate; and G is the buoyant mass of the whole sample when it is totally immersed in the fluid.

The measured density and acoustic phase velocity of nano-composite films as a function of the alumina volume ratio are shown in Fig. 3. It was found that the film density increases from 1150 Kg/m^3 to 1680 Kg/m^3, and the acoustic velocity increases from 2500 m/s to 3020 m/s when the alumina volume ratio increases from 14% to 32%. Hence, the acoustic impedance, which is the product of the density and phase velocity, of the nano-composite film

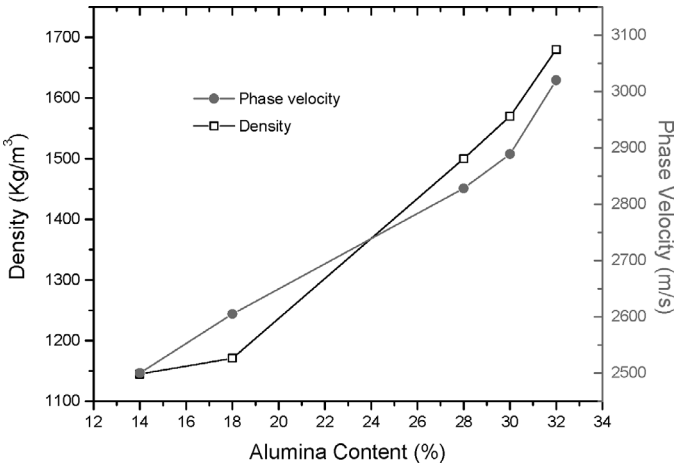


Fig. 3. The density and the acoustic velocities of alumina colloidal/polymer nano-composite films vs. alumina volume ratio.

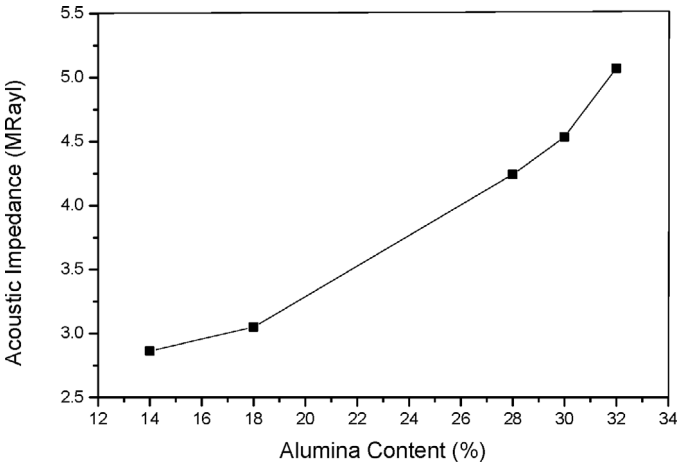


Fig. 4. Acoustic impedance of alumina colloidal/polymer nano-composite films vs. alumina volume ratio.

also increases with the alumina volume ratio, as shown in Fig. 4. The acoustic impedance of the nano-composite film increases from 2.9 MRayls to 5.1 MRayls when the alumina volume ratio increases from 14% to 32%, which is a nearly 70% change. This variation is much larger than that caused by increasing the aging temperature alone, which brings about 31% increase of acoustic impedance [7]. Combining the aging temperature and volume ratio controls, the acoustic impedance of nano-composite film can be tailored in a rather large range, making the composite material ideal as the matching layer of high-frequency transducers.

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