ULTRASONIC CHARACTERIZATION OF ALUMINA/SILICONE-RUBBER COMPOSITES FOR ACOUSTICAL APPLICATIONS

Shiro Biwa, Hideyasu Iwasaki
Kyoto University, Kyoto, Japan
email: biwa@kuaero.kyoto-u.ac.jp

Silicone rubber has been used commonly as a material for an acoustic lens in medical ultrasonic probes. In order to achieve the desired properties for an acoustic lens, i.e., acoustic impedance close that of, and wave velocity lower than that of, human body, different materials have been considered as fillers or dopants to be dispersed in silicone rubber. A micromechanics-based analysis has revealed that silicone-rubber composites can have the desired acoustical properties with a certain amount of dispersed alumina particles. In this study, the ultrasonic wave propagation characteristics of alumina-particle-dispersed silicone-rubber composites fabricated with different particle concentrations and average particle radii were investigated experimentally. Namely, the velocity and attenuation coefficient of longitudinal wave in the alumina/silicone-rubber composites were evaluated at the frequency of 5 MHz by the spectral analysis of the reflected waves obtained in the immersion measurement. The experimental results have shown that the acoustic impedance and the wave velocity desired for an acoustic lens can be achieved by the alumina particle concentration as predicted by the micromechanics-based analysis. The influence of the particle radius on the acoustical properties of the composite has also been examined.
Keywords: ultrasonic waves; acoustic lens; silicone rubber; composite materials; sound velocity

1. Introduction

Ultrasonic waves propagating in composite materials exhibit different characteristics depending on the acoustical properties of their constituting phases as well as their concentrations and microscopic arrangements. In polymer-matrix composites, the wave undergoes remarkable attenuation due to their viscoelastic nature as well as wave scattering losses by their microstructure. Different theoretical models [1-5] have been applied to analyse the wave propagation in these composites and to predict their wave velocities and attenuation coefficients. Such theoretical models are also expected to provide an effective tool to design a composite with suitable acoustical properties. This situation may arise in the design of medical ultrasonic probes used to transmit and focus ultrasound in the human body for diagnosis. In particular, the acoustic lens in a medical ultrasonic probe is desired to have an ultrasonic wave velocity lower than that of human body in order to refract the wave and make focusing. Furthermore, it is preferable that the acoustic impedance of the lens is close to that of human body for better ultrasound transmission. Silicone rubber has typically been used for acoustic lenses. While the wave velocity of silicone rubber is lower than that of human body, there is a significant mismatch of acoustic impedance between the two media. To achieve the acoustical properties desired for an acoustic lens, different combinations of silicone rubber and fillers/dopants have been investigated in the foregoing works based on experimental characterization [6-10]. The design of silicone-rubber composites for acoustic-lens applications may be greatly facilitated if their acoustical properties can be predicted by a theoretical model which accounts for the properties of the filler materials and their concentration as well as their size.

A micromechanics-based analysis of the ultrasonic wave propagation in particle-dispersed silicone-rubber composites was recently reported [11] based on the dynamic generalized self-consistent model [12] (referred to as the DGSC model hereafter). The fibre-composite version of the DGSC model [13] was used to successfully reproduce the ultrasonic wave propagation characteristics of carbon-fibre-reinforced epoxy composites [14]. The analysis in Ref. [11] has revealed that an alumina-particle-dispersed silicone-rubber composite with appropriate particle concentration and size can achieve the aforementioned acoustical properties suitable for an acoustic lens. In this paper, an experimental investigation is reported to verify this theoretical finding. Alumina/silicone-rubber composites were fabricated by using alumina particles of different concentrations and size, and their ultrasonic wave propagation characteristics were evaluated experimentally. The measured longitudinal wave velocities, acoustic impedances and attenuation coefficients of the fabricated composites are compared to the theoretical predictions of the DGSC model.

2. Micromechanical analysis

2.1 Dynamic generalized self-consistent model

The DGSC model used in the theoretical analysis of the ultrasonic wave propagation characteristics in particle-dispersed silicone-rubber composites in Ref. [11] is first outlined here. In the formulation of the DGSC model [12], the composite is represented by a simplified model consisting of a single spherical particle (radius a) embedded in a concentric spherical region of the matrix (outer radius b). This composite sphere is further surrounded by an infinitely extended medium which has the effective properties of the composite, as shown in Fig. 1(a). The volume fraction of the particles in the composite is

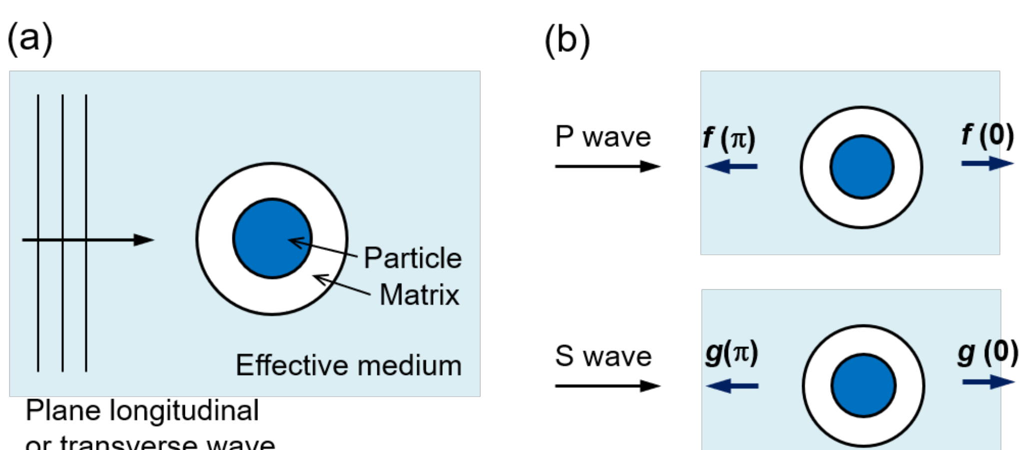

Fig. 1 (a) The DGSC model of a particle-dispersed composite, and (b) the forward- and back-scattered amplitudes for the longitudinal and transverse wave incidence.

given by $\phi = (a/b)^3$. The density, wave velocities and attenuation coefficients of the particle and matrix are denoted by ρ_β, $c_{L\beta}$, $c_{T\beta}$, $\alpha_{L\beta}$, $\alpha_{T\beta}$, respectively, where the index β distinguishes the particle (β = p) and the matrix (β = m), and the indices L and T stand for longitudinal and transverse waves. The density of the composite (effective medium) ρ_{eff} is given by $\rho_{eff} = \phi\rho_p + (1-\phi)\rho_m$.

When the above three-phase composite medium is subjected to the incidence of plane longitudinal or transverse harmonic waves of angular frequency ω, the displacement field can be expressed in terms of the eigenfunction expansions involving the spherical vector wave functions [12]. The expansion coefficients are determined by the stress and displacement continuities at the particle-matrix and the matrix-effective medium interfaces. It then yields the forward- and back-scattering amplitudes, $f(0)$ and $f(\pi)$ for the longitudinal wave incidence and $g(0)$ and $g(\pi)$ for the transverse wave incidence (Fig. 1(b)). The effective wavenumbers of the composite, k_{Leff} and k_{Teff} for longitudinal and transverse waves, respectively, are given by the formula of the Waterman-Truell multiple scattering theory [15] in a self-consistent manner as

$$\left\{1+\frac{2\pi n_0 f(0)}{(k_{Leff})^2}\right\}^2 - \left\{\frac{2\pi n_0 f(\pi)}{(k_{Leff})^2}\right\}^2 = 1, \quad \left\{1+\frac{2\pi n_0 g(0)}{(k_{Teff})^2}\right\}^2 - \left\{\frac{2\pi n_0 g(\pi)}{(k_{Teff})^2}\right\}^2 = 1, \tag{1}$$

where $n_0 = 3\phi/(4\pi a^3)$ represents the number of particles per unit volume. The wavenumbers k_{Leff} and k_{Teff} satisfying Eq. (1) can be obtained by an iterative numerical analysis. Then the wave velocities and attenuation coefficients of the composite are determined from the following relations.

$$k_{Leff} = \frac{\omega}{c_{Leff}} + i\alpha_{Leff}, \quad k_{Teff} = \frac{\omega}{c_{Teff}} + i\alpha_{Teff}. \tag{2}$$

2.2 Analysis of alumina-particle-dispersed silicone-rubber composites

In Ref. [11], the longitudinal wave velocity and attenuation coefficient of particle-dispersed silicone-rubber composites were calculated based on the DGSC model for a wide range of the density, stiffness, volume fraction and radius of spherical particles. As a result, alumina was suggested as a candidate for the particle material to be dispersed in silicone rubber to achieve the wave velocity and acoustic impedance desirable for an acoustic lens. Furthermore, sufficiently low attenuation coefficient was expected when the particle radius was chosen small enough.

In order to compare the experimental data to be shown below with the theoretical analysis, the wave velocity and attenuation coefficient of alumina-particle-dispersed silicone-rubber composites were calculated based on the same model. For this purpose, silicone rubber was assumed to be an isotropic viscoelastic solid (density ρ_m = 1020 kg/m^3) and its acoustical properties in the ultrasonic frequency range were evaluated experimentally. The wave velocity c_{Lm} = 1.03 km/s and the attenuation coefficient α_{LM} = 1.73 cm^{-1} were obtained for the longitudinal wave at the frequency of 5 MHz from the immersion measurement described in the next section (the values of c_{Lm} and α_{LM} different from these were used in Ref. [11] as another set of measurements was used). The velocity of the transverse wave was evaluated as c_{Tm} = 0.659 km/s at the same frequency from the contact method [16, 17], but its attenuation was very high and difficult to quantify. Therefore, the transverse wave attenuation coefficient of 15.6 cm^{-1} measured for epoxy resin was used as α_{Tm} in the analysis. The corresponding coefficient of silicone rubber was likely much higher than this, so approximately the ten times as high value, 150 cm^{-1}, was also used in the analysis to see its effect on the predicted properties. Alumina particles were modelled as an isotropic elastic solid and their properties were set as ρ_p = 3980 kg/m^3, c_{Lp} = 11.73 km/s, c_{Tp} = 6.93 km/s and $\alpha_{Lp} = \alpha_{Tp} = 0$. It is assumed in the theoretical model that the particles have the common radius although the samples used in the experiment had a distribution of particle size. To compare the theoretical results to the experimental data, the average radius of the particles was used for the analysis.

3. Experiment

3.1 Specimens

Alumina/silicone rubber composite specimens used in this study were fabricated using alumina particles (Alunabeads/CB, Showa Denko K.K.) and silicone rubber (TSE3032, Momentive Performance Materials Japan L.L.C.) by the compression moulding at 323 K for 90 minutes. Silicone rubber and alumina particles were mixed with the assigned particle weight fractions of about 30, 50, 70 and 80 %. The specimens were square sheets of thickness 0.8 mm and lateral size 55 mm, and had the volume fractions and average radii of alumina particles as shown in Table 1. The particle volume fractions were calculated from the weight fractions and the densities of silicone rubber and alumina particles. The average radii are the values provided by the supplier of the alumina particles.

From the scanning electron micrographs of the specimen cross-sections, no remarkable aggregation of particles was found in the specimens with average particle radius of 5 microns (Fig. 2). For the specimens with smaller average particle radii, however, some aggregation was found. For the specimens with larger particle radii, the particles tended to cluster near the lower surface of the specimen, likely due to the settling due to their weight during the moulding.

3.2 Ultrasonic measurement

In the ultrasonic measurement, each specimen was immersed in water and the longitudinal wave was

Table 1 Specification of fabricated alumina/silicone-rubber composites

Specimen No.	1	2	3	4	5	6	7	8	9	10
Particle weight fraction [%]	0	29.9	50.0	50.0	50.0	50.0	50.0	60.0	70.0	80.0
Particle volume fraction [%]	0	9.9	20.4	20.4	20.4	20.4	20.4	27.8	37.4	50.6
Particle average radius [μm]	-	5.0	1.5	2.0	5.0	11.0	21.5	5.0	5.0	5.0
Density [kg/m^3]	1020	1330	1560	1570	1630	1630	1560	1820	2090	2380

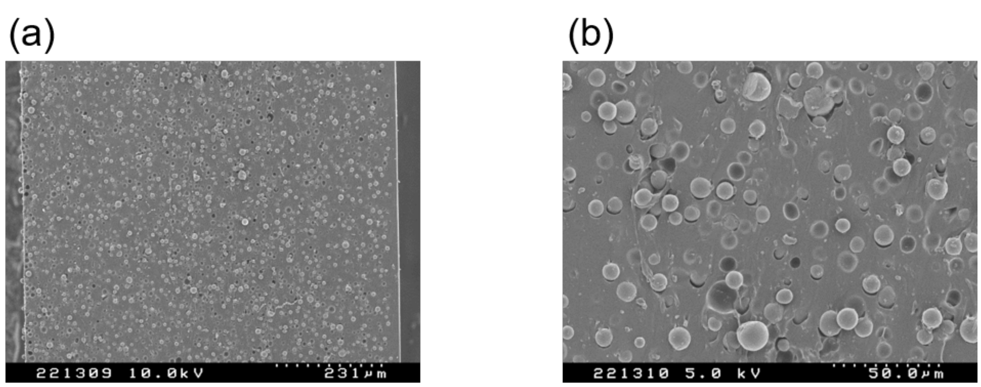

Fig. 2 SEM images of the cross-section of a composite specimen (particle volume fraction 20.4 %, average particle radius 5 mm), showing (a) the whole thickness and (b) an enlarged area.

sent to the specimen in the normal direction and the surface and bottom echoes were recorded using a Panametrics pulser-receiver 5072PR and a piezoelectric transducer (Panametrics C309, nominal centre frequency 5 MHz). From the recorded digital reflection waveforms, the wave velocity was evaluated by the phase spectrum method [16]. The attenuation coefficient was evaluated from the amplitude spectra by accounting for the reflection at the water-specimen interface. The diffraction correction was also made although its effect was negligible as the specimen was sufficiently thin [17]. These methods yield the wave velocity and the attenuation coefficient as functions of the frequency. Below, their values at the frequency 5 MHz of the centre frequency of the transducer used in the experiment. The measurements were made at different points and from both sides of each specimen.

4. Results and discussion

The longitudinal wave velocity of the alumina/silicone-rubber composite at the frequency of 5 MHz was obtained by the theoretical analysis as a function of the volume fraction and radius of particles, as shown in Fig. 3. In Fig. 3(a), the wave velocity calculated for the particle radius of 5 microns is shown

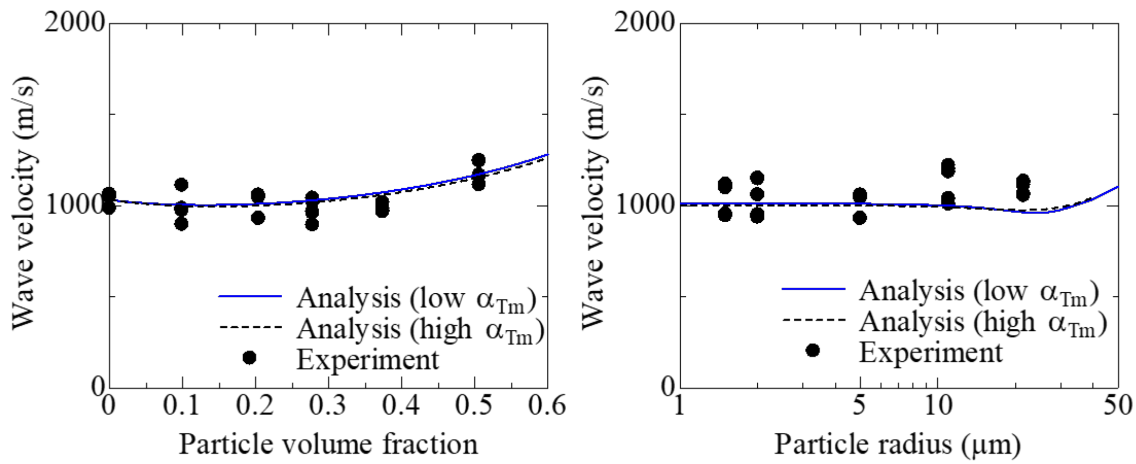

Fig. 3 Variation of the wave velocity with (a) the particle volume fraction (when a = 5 μm), and (b) with the particle radius (when $\phi = 0.204$).

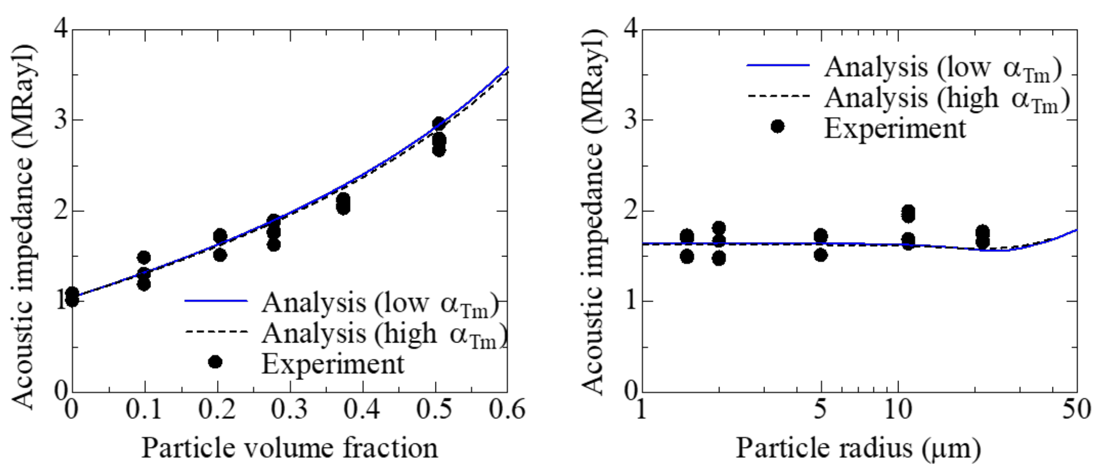

Fig. 4 Variation of the acoustic impedance with (a) the particle volume fraction (when a = 5 μm), and (b) with the particle radius (when $\phi = 0.204$).

against the particle volume fraction. The velocity first decreases slightly and turns to increase with the particle volume fraction. Two theoretical curves are shown for different values of the transverse wave attenuation of silicone rubber (α_{Tm} = 15.6 cm^{-1} and 150 cm^{-1}), but its effect is negligibly small. The experimental results for the average particle radius of 5 microns are also shown in Fig. 3(a), which have some scatters but follow the same trend as the theoretical results. In Fig. 3(b), the theoretical wave velocity, calculated for the particle volume fraction of 20.4 %, is almost constant against the particle radius up to 20 μm. The experimental results for the same volume fraction do not show any clear dependence on the average particle radius. From Fig. 3(a) and (b), it is found that the wave velocity of the composite remains lower than that of human body, which may be represented by the wave velocity in water (1540 m/s), for a wide range of particle volume fraction and radius.

With respect to the application to an acoustic lens, it is of interest to design the composite which has the acoustic impedance close to that of human body. The acoustic impedance is the product of density and wave velocity. The human body is known to have the impedance in the range of 1.5 - 1.6 MRayl (1 MRayl is 1×10^6 kg/(m^{2}s)), which is higher than that of silicone rubber (about 1.0 MRayl). The acoustic impedance of the alumina/silicone-rubber composite can be obtained theoretically using the wave velocity calculated by the micromechanical model and the density by the rule of mixture. The composite acoustic impedance is shown as a function of the particle volume fraction in Fig. 4(a) for the particle radius 5 microns, and as a function of the particle radius in Fig. 4(b) for the particle volume fraction of 20.4 %. Again, the effect of the transverse wave attenuation of silicone rubber is negligible. In Fig. 4(a), it is shown that the composite acoustic impedance is close to that of human body when the particle volume fraction is as about 20 %. It is shown in Fig. 4(b) that this choice is appropriate for the desired value of the impedance for a wide range of particle radius. The experimental results show a reasonable agreement with the theoretical predictions.

Finally, the wave attenuation in the composite is examined as it also affects the performance as an acoustic lens. The theoretical and the experimental results are shown in Fig. 5. In Fig. 5(a), the theoretically calculated attenuation coefficient first increases with the particle volume fraction and then turns to decrease for higher volume fractions. It is clearly shown that the transverse wave attenuation of silicone rubber makes significant effect on the theoretical results. The higher value α_{TM} = 150 cm^{-1} appears to give the theoretical results closer to the experimental results. It should be noted, however, that the experimental results contain the influence of the spatial inhomogeneity of particle distribution while the theoretical model assumes a uniform particle distribution. It is known that the wave attenuation in

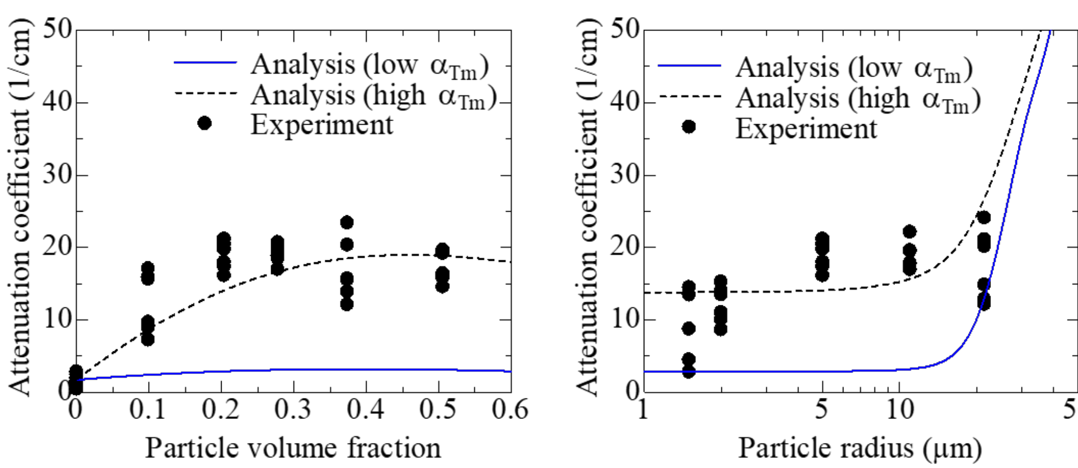

Fig. 5 Variation of the attenuation coefficient with (a) the particle volume fraction (when a = 5 μm), and (b) with the particle radius (when ϕ = 0.204).

polymer-based composites is mainly brought about by the scattering loss and the viscoelastic absorption [3-5]. The scattering loss dominates when the size of particles is comparable to the wavelength, but becomes negligible for sufficiently small particles. In Fig. 5(b), the theoretical attenuation coefficient of the composite shows almost constant values up to the particle radius of 10 microns, and starts to increase for larger particle radii where the scattering loss is expected to dominate. The experimental results show no clear dependence on the average particle radius, but they are significantly high as compared to the value of silicone rubber itself. The attenuation is preferably as low as possible in the acoustic-lens application. This may be achieved by using even smaller particles as has been attempted by other investigators [6-10], and/or making the spatial particle distribution more uniform, without affecting the wave velocity and acoustic impedance.

5. Summary

The propagation characteristics of ultrasonic waves in alumina-particle-dispersed silicone-rubber composites have been investigated theoretically and experimentally. The dynamic generalized self-consistent model has been used to calculate the wave velocity and attenuation coefficient of the composites. The theoretical results are in reasonable agreement with the experimental ones in terms of the ultrasonic wave velocity and acoustic impedance. The micromechanical model can thus be used to choose the material, concentration and size of fillers which give the acoustical properties of particle-dispersed silicone-rubber composites suitable for acoustic lens applications. Micromechanics-based analysis is expected to be useful for the design of other acoustical composites, e.g., backing materials in ultrasonic probes.

Acknowledgments

The authors would like to express their gratitude to Showa Denko K.K. for providing alumina particles and for TANAC Co., Ltd. for fabricating the composite specimens. The authors also thank Mr. S. Kinoshita for his assistance in SEM observations.

REFERENCES

1 Sabina, F. J. and Willis, J. R. A simple self-consistent analysis of wave propagation in particulate composites, *Wave Motion*, **10**, 127-142, (1988).

2 Kim, J.-Y., Ih, J.-G. and Lee, B.-H. Dispersion of elastic waves in random particulate composites, *Journal of the Acoustical Society of America*, **97**, 1380-1388, (1995).

3 Biwa, S., Idekoba, S. and Ohno, N. Wave attenuation in particulate polymer composites: independent scattering/absorption analysis and comparison to measurements, *Mechanics of Materials*, **34**, 671-682, (2002).

4 Biwa, S., Watanabe, Y. and Ohno, N. Analysis of wave attenuation in unidirectional viscoelastic composites by a differential scheme, *Composites Science and Technology*, **63**, 237-247, (2003).

5 Biwa, S., Watanabe, Y., Motogi, S. and Ohno, N. Analysis of ultrasonic attenuation in particle-reinforced plastics by a differential scheme, *Ultrasonics*, **43**, 5-12, (2004).

6 Yamashita, Y., Hosono, Y. and Itsumi, K. Effects of metal particle dopant on acoustic attenuation properties of silicone rubber lens for medical echo array probe, *Japanese Journal of Applied Physics*, **44** (6B), 4558-4560, (2005).

7 Yamashita, Y., Hosono, Y. and Itsumi, K. Effects of metal particle dopant on acoustic attenuation properties of silicone rubber lens for medical echo array probe, *Japanese Journal of Applied Physics*, **44** (6B), 4558-4560, (2005).

8 Yamashita, Y., Hosono, Y. and Itsumi, K. Effects of sub-100 nm platinum metal particle on the acoustic attenuation properties of silicone rubber lens for medical array probe, *Japanese Journal of Applied Physics*, **45** (5B), 4684-4687, (2006).

9 Yamashita, Y., Hosono, Y. and Itsumi, K. Effects of ceramic nanopowder dopants on acoustic attenuation properties of silicone rubber lens for medical echo probe, *Japanese Journal of Applied Physics*, **46** (7B), 4784-4789, (2007).

10 Yamashita, Y., Honoso, Y., Yamamoto, N. and Itsumi, K. Effects of ytterbium oxide nanopowder particle size on the acoustic properties of silicone rubber lens for medical echo array probe, *Japanese Journal of Applied Physics*, **47** (5), 4132-4136, (2008).

11 Biwa, S. and Iwasaki, H. Micromechanical analysis of particle-dispersed silicone-rubber composites for acoustical applications, *Proceedings of the 7th Asian Conference on Mechanics of Functional Materials and Structures*, on-line, 13-15 March, (2021).

12 Yang, R.-B. A dynamic generalized self-consistent model for wave propagation in particulate composites, *Transactions of the ASME Journal of Applied Mechanics*, **70**, 575-582, (2003).

13 Yang, R.-B. and Mal, A. K. Multiple scattering of elastic waves in a fiber-reinforced composite. *Journal of the Mechanics and Physics of Solids*, **42**, 1945–1968, (1994).

14 Lonné, S., Lhémery, A., Calmon, P., Biwa, S. and Thévenot, F. Modeling of ultrasonic attenuation in unidirectional fiber reinforced composites combining multiple-scattering and viscoelastic losses, *AIP Conference Proceedings*, **700**, 875-882, (2004).

15 Waterman, P. C. and Truell, R. Multiple scattering of waves, *Journal of Mathematical Physics*, **2**, 512-537, (1961).

16 Papadakis, E. P., Fowler, K. A. and Lynnworth, L. C. Ultrasonic attenuation by spectrum analysis of pulses in buffer rods: method and diffraction correction, *Journal of the Acoustical Society of America*, **53**, 1336-1343, (1973).

17 Biwa, S., Ito, N. and Ohno, N. Elastic properties of rubber particles in toughened PMMA: ultrasonic and micromechanical evaluation, *Mechanics of Materials*, **33**, 717-728, (2001).