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Effect of particle size of piezoelectric powder phase on sol-gel composites

ゾルゲル複合体における圧電粉体粒径の影響

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1. Introduction

Ultrasonic Biomicroscopy (UBM) has been utilized for micro-structure diagnosis of living tissues such as eye, skin, and blood vessel because this technique is non-invasive and can distinguish tissues by elasticity difference. [1,2] Ultrasonic transducer is considered an important component of UBM. In order to acquire high resolution, it is preferred for ultrasonic transducers to have adequate high frequency, broad frequency bandwidth, and high sensitivity. In the case of mechanical scanning with single ultrasonic transducer with an acoustic lens, low dielectric constant material has better electrical impedance matching.

Sol-gel composites, which were invented by D. Barrow et al for thick film fabrication, could be good candidates for this application, because they have low dielectric constant and broad frequency bandwidth, and it is possible to fabricate onto curved surface such as an acoustic lens directly so that it could improve signal to noise ratio (SNR) [3]. In past studies, ultrasonic transducers with peak frequency higher than 30MHz were accomplished by this method. However, sometimes UBM requires higher frequency. Sol-gel composites were composed by three phases, ferroelectric powder phase, dielectric sol-gel phase, and air phase and it was expected that thin film fabrication by sol-gel composite could be achieved if the particle size of ferroelectric powder phase became smaller. In this study, sol-gel composites with three ferroelectric powders, made from bulk ceramics, sol-gel solution by different powder synthesis processes, were fabricated in order to investigate the effect of ferroelectric phase particle size on sol-gel composites.

2. Material selection and preparation

For the material of ferroelectric powder phase, and dielectric sol-gel phase, lead zirconate titanate (PZT) and PZT were chosen because of high piezoelectric constant and high dielectric constant, respectively. Bulk PZT powders were provided by Murata Corporation. PZT sol-gel source powders were synthesis by two different process. Sol-gel powder A was synthesized based on thin film fabrication process. PZT sol-gel solution was poured into an alumina crucible, then it was dried at 150°C for 1h and fired at 650°C for 2h by a furnace. Then it was pounded in a mortar. Sol-gel powder B, was synthesized based on nano-powder fabrication process. [4] Polyvinylpyrrolidone (PVP) with average molecular weight 1,300,000 were added to PZT sol-gel solution. Stirring was continued for 2h to obtain PZT-PVP gel. Subsequently, PVP with average molecular weight 55,000 was added to PZT-PVP gel. Continuous stirring was operated for 5h. This gel was dried at 70°C for a week and then sintered at 550°C for 24h. PZT sol-gel solution was self-manufactured.

3. Results and discussions

X-ray diffraction (XRD)

X-ray powder diffraction was operated for bulk PZT, sol-gel powder A and sol-gel powder B, and the results were shown in **Fig. 1**. The XRD patterns of both sol-gel powder A and B showed the diffraction peaks indicating the presences of mainly perovskite phases with very little pyrochlore phases (Fig.1b, c). On the other hand, the XRD pattern of bulk PZT showed the first diffraction peaks indicating the presence of perovskite although the positions of high order peaks were different from both perovskite and pyrocholore (Fig.1a). It indicated dopant additive existence, since sol-gel powder A and B were pure PZT.

Scanning electron microscopy (SEM)

To confirm the particle size of each ferroelectric powder phase, SEM images were taken and the results were shown in **Fig. 2**. In SEM images of bulk PZT, spherical particles were observed and the particle size was the order of a few hundred nanometers (Fig.2a and b). On the other hand, in sol-gel powder A, structurally heterogeneous small particles on non-spherical particles were observed and the particle size was the order of tens of micrometers (Fig.2c). In sol-gel powder B, relatively uniform spherical form particles were observed and the particle size was few micrometers (Fig.2d). As a result, three kind particle sizes were prepared.

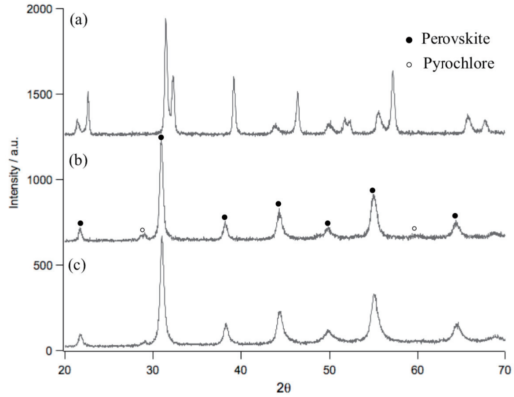

Fig.1 XRD patterns of (a), (b) bulk powder, (c) sol-gel powder A and (d) sol-gel powder B.

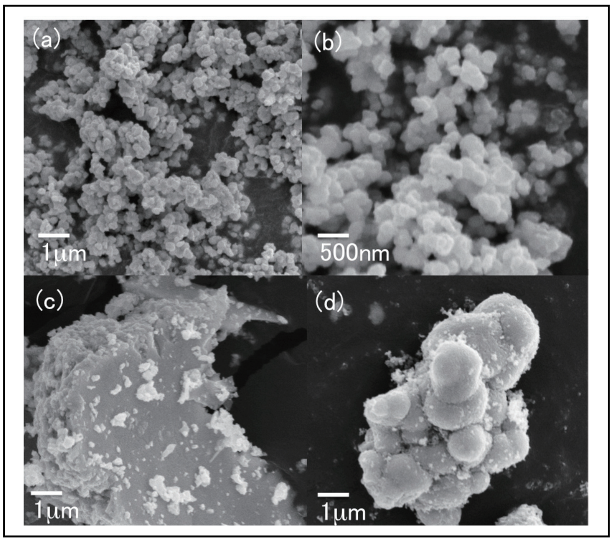

Fig.2 SEM images of (a), (b) bulk powder, (c) sol-gel powder A and (d) sol-gel powder B.

Sol-gel composite film fabrication

Three different kinds of ferroelectric powders, bulk powder, sol-gel powder A, and sol-gel powder B, were mixed with PZT sol-gel solution for sol-gel composite fabrication. However, sol-gel powder B caused some reaction with PZT sol-gel solution and water and PZT sol-gel solution were added in order to obtain appropriate viscosity was obtained. The caused reason is under investigation and chemical residue of sol-gel powder B was suspected. Then each mixture was sprayed onto 30×30×3mm titanium substrate. Titanium substrates were used because the attenuation of high frequency component was relatively small. After spray coating process, thermal treatments, drying at 150°C for 5min and firing at 650°C were operated. Spray coating and thermal treatments were repeated until the surface was completely covered and it was confirmed by optical microscope. Then after electrical poling, 6mm diameter top electrodes were fabricated by silver paste.

Ultrasonic performance

Each ultrasonic transducer made with 3 kind particle size of ferroelectric powder phase was connected with a pulser/receiver and a digital oscilloscope and the reflected echoes from the bottom substrate were measured in pulse-echo mode. The results by bulk powder and sol-gel powder B were shown in **Fig. 3**. Concerning the signal strength, sol-gel powder B was higher than bulk powder, and the signal strength of sol-gel powder A was worst. Fast Fourier Transform (FFT) results of the first reflected echo were shown in **Fig. 4**. The peak frequency of bulk PZT was around 40MHz whereas that of sol-gel B was around 25MHz. The lower signal strength might be resulted from attenuation from the substrate.

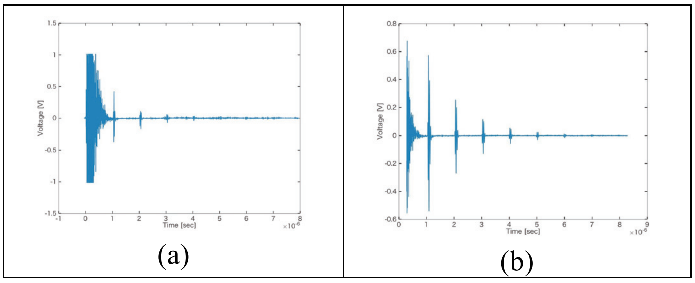

Fig. 3 Pulse-echo measurement results of (a) bulk powder and (b) sol-gel powder B.

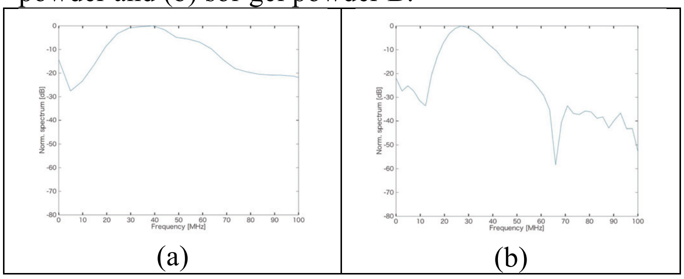

Fig. 4 FFT results of (a) bulk powder and (b) sol-gel powder B.

4. Conclusions

It seemed that smaller particle size of ferroelectric powder phase was suitable to fabricate high frequency ultrasonic transducers. Further investigation for particle control derived by sol-gel solution is required to make a final conclusion.

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