Fundamental Study on Array Type Medical Ultrasound Probe with Hydrothermally Synthesized PZT Poly-Crystalline Film
- Fabrication and Performance Estimation of Miniature 1-Dimensional Array Type Ultrasound Probe -

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Abstract—In this study, in order to apply the hydrothermally synthesized piezoelectric polycrystalline films to the medical miniature 1-dimensional array type ultrasound probes with resonance frequency of 10 MHz, the hydroxyapatite was employed as the material for the substrate. After sputtering of pure titanium on the surface of hydroxyapatite substrate, the titanium film was etched using photo-etching method to form 1-dimensional titanium film electrodes array with element pitch of 75 µm, element width of 40 µm and element length of 4mm in order to scan ultrasound beam electronically by sector scan mode using phased array technique. We tried to deposit PZT poly-crystalline film hydrothermally on the titanium film electrodes. The deposited PZT poly-crystalline film has thickness of about 10 µm on the 1-dimensional array titanium electrode pattern. We could succeed trial fabrication of the miniature 1-dimensional array type ultrasound probe. The sputtered and etched titanium film electrodes were used as the electrodes for signal lines of the 1-dimensional array type ultrasound probe. Furthermore, a gold electrode was deposited as a common ground electrode all over the surface of 1-dimensional hydrothermally synthesized PZT poly-crystalline film vibrators. Transmitted ultrasound pulse from 10 MHz commercial ultrasound probe was received by the fabricated 1-dimensional array type ultrasound probe with hydrothermally synthesized PZT poly-crystalline film vibrators.

Keyword; 1-dimensional array; hydrothermal method; medical ultrasound; photolithography; high frequency; hydroxyapatite substrate; titanium electrode

I. INTRODUCTION

In recent years, it became important to develop high frequency and high resolution medical ultrasonic diagnostic equipments such as the diagnostic equipments for IDUS (Intraductal ultrasonography), for IVUS (Intravascular ultrasonography), and for ultrasonic diagnosis in ophthalmologic field, because the early diagnosis of pathological changes became more significant. Therefore, medical micro ultrasound probes and array type high resolution medical ultrasound probes with wide frequency band in the high frequency range are researched and developed actively. Recently, piezoelectric ceramics like Pb(Ti, Zr)O$_3$ (Lead Zirconate Titanate), and piezoelectric polymer films including PVDF(polyvinylidene-fluoride), P(VDF-TrFE) are mainly used as the piezoelectric material for the array type medical ultrasound probes. However, since piezoelectric ceramics are hard and brittle material, it is difficult to fabricate such tiny array type ultrasound probes. Moreover, ultrasound probes with piezoelectric polymer films show wide frequency band but they have low electromechanical coupling coefficient and small dielectric coefficient.

Therefore, we are studying on synthesis of PZT poly-crystalline film on a titanium substrate by the hydrothermal method. It is thought that hydrothermally synthesized PZT poly-crystalline films are suitable for such tiny ultrasound probes, or hydrophones [1]. Hydrothermally synthesized PZT poly-crystalline films have features as follows. Poling process is not required and they can be synthesized at the low temperature under Curie point [2]. They can be deposited on the tiny or complex shaped titanium substrates [3]. They have low acoustic impedance [4]. The ultrasound probe with hydrothermally synthesized PZT poly-crystalline films have the super-broadband frequency characteristic of sensitivity from 1 to 40 MHz or more [4]. We considered the feasibility to deposit the PZT poly-crystalline films hydrothermally on the one dimensional titanium film electrode array under severe synthesizing condition of strong alkaline solution of 4M and temperature from 120 to 160 $^\circ$C for 24 hours.
II. EXPERIMENTS

A. Fabrication of 1-dimensional array type ultrasound probe

Hydroxyapatite (HAp) pellets with diameter of 13 mm and thickness of 2 mm (Cellyard pellet, Pentax Co.) were employed as substrates in this study. HAp shows stable material properties in an alkaline solution. 1-dimensional titanium film electrodes array was formed by the photolithography method after deposition of titanium films with thickness of about 3 µm on HAp substrate by vapor deposition method. The array pattern of titanium film electrodes with 8 elements was designed for electronic sector scan with directivity wider than steering angle of 70 degree at frequency of 10 MHz. The electrode pattern with element pitch of 75 µm, element width of 35 µm and element length of 4 mm was designed with consideration of electric impedance matching between probe and electronic circuits of a transmitter and a receiver. The specific dielectric constant of hydrothermally synthesized PZT poly-crystalline film is about 500 [5].

Figure 1 shows the structure of the 1-dimensional array ultrasound transducer. Figure 2 shows the deposited and etched one dimensional titanium film electrode array pattern on HAp substrate using the photolithography method with the digital microscope (VH-8000, Keyence). The patterned titanium film electrode array was used as electrode for signal line. PZT poly-crystalline film is deposited with hydrothermal method on the patterned titanium film array as a signal electrode. Then, Au electrode was deposited all over the PZT poly-crystalline film array by vapor deposition method and it was used as a ground electrode. Zirconium chloride oxide octahydrate (ZrOCl₂ · 8H₂O, 99.0 %), Titanium dioxide (TiO₂, 99.0 %), lead nitrate (Pb(NO₃)₂, 99.0 %) were used as source materials, and potassium hydroxide (KOH) was used as mineralizer for hydrothermal synthesis of PZT polycrystalline films. Hydrothermally deposition temperature was 160 °C. Synthesizing time of one cycle was 24 hours and the synthesizing process was repeated 7 times.

B. The experiment of reception and transmission with an element of trially fabricated probe

The received and transmitted ultrasound with an element of 1-dimensional array type ultrasound probe was measured in degassed water.

In this experiment of ultrasonic reception, the output signal (amplitude 300 mV<sub>o-p</sub>, center frequency 10 MHz, tone burst pulse) from a function generator (HP8116A, HP) was amplified with the power amplifier (2100L, ENI) at 55 dB gain factor and applied to a 10 MHz immersion type ultrasound probe (13-1008-R, Harisonic Co.). The detected signal was amplified by a pre-amplifier with gain of 40 dB in a pulser receiver (5800L, Panametrics).

In this ultrasonic wave transmission experiment, an element was driven by a signal (amplitude 150 mV<sub>o-p</sub>, center frequency 10 MHz, tone burst pulse) from a function generator (HP 8116A, HP). The signal was amplified by the power amplifier (2100L, ENI) with a gain factor of 55 dB. The transmitted ultrasonic wave was detected by a needle type hydrophone (NP-1000, NTR Co.). The detected signal was amplified by the pre-amplifier with gain factor of 60 dB in the pulser receiver. These detected signals were displayed on an oscilloscope (TDS2012, Tektronix Co.).

The pulser receiver had a high-pass filter with a cut off frequency of 1 kHz and a low pass filter with a cut off frequency of 35 MHz.

The distance between transmitting ultrasonic probe and receiving ultrasonic probe was 12 mm in each experiment.

III. EXPERIMENTAL RESULTS

A. Deposited condition and accuracy of dimension of PZT poly-crystalline film

One dimensional array pattern was observed before and after hydrothermal deposition of PZT poly-crystalline films with a scanning electron microscope (JSM-5500LV, JEOL).
The results are shown in Fig.3. The nucleation process of hydrothermal method for PZT poly-crystals starts from the elution of titanium ions from titanium substrate[3]. Therefore, we were apprehensive that the titanium electrode film was dissolved into the mineralizer of KOH solution and element width decreased. However, it was found from Fig.3 that the element widths did not change even after repeat of hydrothermal depositions 7 times. However, impurities were accumulated on the HAp substrate between elements after repeat of hydrothermal deposition process. It was confirmed from Fig.3 that the element pitches were about 75 µm and the changes of element pitches before and after hydrothermal deposition of PZT poly-crystals were very small. Furthermore, it was confirmed that the element widths increased from 35 µm to 40 µm by growth of PZT crystals on the titanium electrode film.

![Fig.3 SEM images of elements of 1D-array pattern on HAp after hydrothermal synthesis of PZT](image)

(a)

(b)

Fig.3 SEM images of elements of 1D-array pattern on HAp after hydrothermal synthesis of PZT (a) Before deposition of PZT (b) After deposition of PZT

The compositions of the hydrothermally deposited crystals on the titanium electrode films were analyzed with EDS (JSM5500-EDS, JEOL). The analyzed result is shown in Fig.4. The analyzed result of composition of HAp substrate is shown in Fig.5. It was found from Fig.4 that Pb, O, Ti, and Zr originating in PZT, were detected. Moreover, it was found from Fig. 5 that Ca and P were detected as components of HAp. However, Ca and P were not detected in the hydrothermally deposited PZT poly-crystalline films as shown in Fig.4. It was proved from these results that the compositions of hydrothermally deposited crystals on the titanium electrode films were similar as the conventional hydrothermally deposited PZT poly-crystal [4]. It can be thought that the hydrothermally deposited crystals did not affected by the compositions of Hap substrate. It was confirmed from Fig.4 that composition ratio of Zr : Ti ratio was 79 : 21. From these results, it can be thought that the PZT poly-crystalline films were deposited on the titanium film electrode array on the HAp substrate.

![Fig.4 poly-crystals on Ti film after hydrothermal synthesis by EDS](image)

![Fig.5 Compositions of hydroxyapatite (Ca_{10}(PO_4)_6(OH)_2) substrate](image)

B. The experiment of reception and transmission of ultrasonic wave with an element of trial fabricated probe

The transmitted ultrasonic wave with the 10 MHz commercial water immersion type ultrasonic probe was received by one element of 1D array probe fabricated by hydrothermal method. The received waveform is shown in
The PZT poly-crystalline film was deposited hydrothermally on the vapor deposited one dimensional titanium electrode film array on the HAp substrate by vapor deposition and photolithography method in this study. We were anxious about the dissolution and exfoliation of titanium from the vapor deposited electrodes, because of under the severe hydrothermally depositing condition of PZT poly-crystals with synthetic temperature about 160 °C and the strong alkaline solution of 4 M. However, it was confirmed that a PZT poly-crystalline films could be deposited precisely on the patterned titanium electrode films. We were apprehensive that the gaps between elements were filled with the grown PZT crystals by repeat of hydrothermal synthesizing process.

However, we could confirm that the width of elements with PZT poly-crystalline films was kept about 35 μm. We think from these results that our miniature one dimensional ultrasound probe with hydrothermally synthesized PZT poly-crystalline films can be applied to the array type miniature medical ultrasound probe.

Ultrasonic waveforms having 3 cycles were detected with good SN ratio at 10 MHz by an element in the fabricated 1D array probe by hydrothermal method. Furthermore, it was confirmed by experiment that an element of 1 D array probe fabricated by hydrothermal method could transmit ultrasonic wave pulse. The ultrasound was transmitted with an element of 1D array probe had the low SN ratio. We think that improvement of transmitting sensitivity is significant future work.

Moreover, eight elements in 1D array probe should be driven simultaneously and directivity is also measured.

### Reference


