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Properties and characteristics of P(VDF/TrFE) transducers manufactured by a solution casting method for use in the MHz-range ultrasound in air

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A B S T R A C T

Highly effective piezoelectric polymer transducers operating in air at high frequencies have been successfully made by casting a solution of ferroelectric poly(vinylidene fluoride-*co*-trifluoroethylene) P(VDF/TrFE) directly on a backing metal plate, and their performance has been evaluated. By utilizing this method, it has been possible to develop the three kinds of transducers that operate respectively at 4, 6 and 10 MHz in air. For precise evaluation of the performance of the P(VDF/TrFE) transducers, the absorption loss in air was measured up to 10 MHz. It was confirmed that the empirical formula obtained from the measured absorption values in air at high frequencies was in alignment with its theoretical value. In addition, a high lateral resolution acoustic image of a ROM-Chip (amplitude-image) at 6 MHz in air was successfully displayed using an air coupled concave type P(VDF/TrFE) transducer by bonding an epoxy adhesive.

1. Introduction

Air-coupled ultrasound has often been used for analysis and imaging in the evaluation of materials [1,2]. An acoustic image using a lead zirconate titanate (PZT) transducer was reported by Fox et al. in 1985 [3]. The air-coupled transducers at high frequency using micromachining technology were also developed [4,5]. In general, ceramics and composites such as PZT are utilized in these transducers. Although the piezoelectric ceramics have a large electromechanical coupling factor (k_t), their acoustic impedances are one order of magnitude larger than that of piezoelectric polymers. The silicon rubber of a quarter wavelength with PZT piezoelectric ceramics element is utilized as a matching layer because the acoustic impedance of air is smaller than that of PZT [3,6]. Piezoelectric polymer transducers, on the other hand, have lower acoustic impedances, and thus the matching loss between a transducer and the air is greatly reduced compare with piezoelectric ceramic transducers. In addition, the piezoelectric polymers can be excited with burst waves of much higher voltage, because they have higher ferroelectric coercive field strength. Although the efficiency of a piezoelectric polymer comparing to that of piezoelectric ceramics, it has the properties much advantageous over ceramics, such as flexibility and a concave type transducer can be fabricated easily. The copolymer P(VDF/TrFE) [poly(vinylidene fluoride-*co*-trifluoroethylene)] has the highest piezoelectric activity among known piezoelectric polymer materials [7,8]. We previously developed a concave type ultrasonic transducer operating in air at 2–3 MHz, using piezoelectric films of P(VDF/TrFE) with 75 mol% VDF, and have shown high resolution acoustic images [9,10]. The transducer was composed of two or three 95 μm-thick P(VDF/TrFE) films stacked together with epoxy adhesive on the metallic backing layer plate [11]. The parallel type transducer made up of three stacked P(VDF/TrFE) films excited electrically in parallel by a driving source, as shown in Fig. 1a. The transducer of this type has higher capacitance, which brings about improved electrical impedance matching between the transducer and the electric source. Thus it is possible for the efficiency of this parallel type transducer to increase more than that of a serial type transducer made by piling up P(VDF/TrFE) films with the same polarization direction in series, as shown in Fig. 1b. However, the transducer insertion loss is great due to the epoxy adhesive between the films and an electrode plate. The influence of the epoxy adhesive thickness is large in high frequency type transducers because the film thickness is relatively thin. There are three difficulties associated with the adhesive resulting in a decrease energy conversion efficiency: (1) it is technically difficult to bond the adhesive uniformly and thinly [12]; (2) small bubbles of air are trapped in the epoxy adhesive and (3) there are kissing bonds that reduce the strength of the bond between the adhesive and the adherent [13].

The objectives of the present study were to develop P(VDF/TrFE) transducers with higher sensitivity and higher working frequency, avoid adhesive problems by casting P(VDF/TrFE) solution directly on to the backing electrode, and to evaluate their performance as air-coupled transducers. Several kinds of transducers operating at high frequencies (MHz band) have been fabricated by the solution

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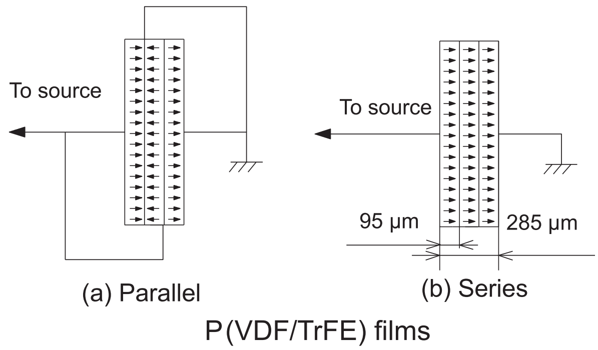

Fig. 1. Schematic configuration of P(VDF/TrFE) films and electrodes.

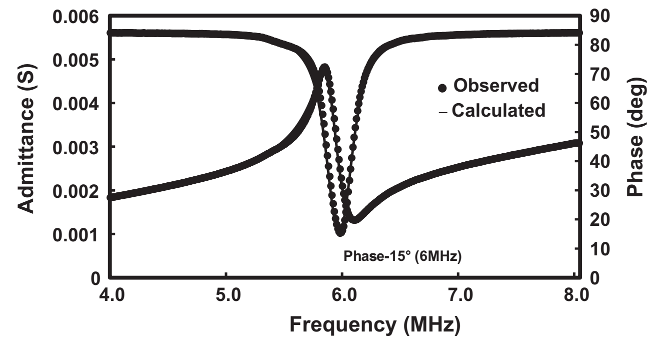

Fig. 2. Observed and simulated admittance-phase/frequency curves for the P(VDF/TrFE) film with Al-plate by casting method. The size of the film was 13×12 mm^2.

Table 1
Electromechanical properties of P(VDF/TrFE), epoxy and air for simulation.

Active area of transducer	156 (mm^2)
Density [P(VDF/TrFE)]	1.89 (g cm^{-3})
Sound speed [P(VDF/TrFE) longitudinal wave][a]	2400 (m s^{-1})
Thickness [P(VDF/TrFE)]	98 (μm)
Dielectric constant $\varepsilon/\varepsilon_0$ [P(VDF/TrFE)]	4.5
Coupling factor k_t [P(VDF/TrFE)][a]	0.27
Dielectric loss tangent, tanδ_e [P(VDF/TrFE)]	0.1
Mechanical loss tangent, tanδ_m [P(VDF/TrFE)]	0.05
Backing-layer (Al)	300 (μm)
Forward-layer (Al)	0.2 (μm)
Sound speed (Al)	6800 (m s^{-1})
Sound speed (air)	340 (m s^{-1})
Density (air)	0.0012 (g cm$^{-3)}$

[a] From the fitting curve shown in Fig. 2.

casting method. These air-coupled transducers were proven to work with much higher efficiency in the piezoelectric polymer transducers, as analyzed to the transducer fabricated by bonding method.

Application of concave polymer transducers to high frequency ultrasound imaging in the air is very important for the inspection of such objects as paper goods and electronic products. Fabricating a concave type P(VDF/TrFE) transducer is, however, still unsuccessful at present because of difficulty in making the P(VDF/TrFE) film uniformly over the concave electrode metal surface through casting. However, this difficulty will be overcome by refining the casting method. From such a perspective, we developed the concave type air-coupled transducer by bonding P(VDF/TrFE) film with epoxy adhesive onto the concave metal electrode plate of focal length of 6.3 mm, in order to obtain the usefulness of the polymer transducer for air-coupled imaging at high frequencies. Using this transducer, we have successfully displayed a 6 MHz acoustic amplitude air-coupled image of a ROM-Chip with sufficiently high lateral resolution.

2. Fabrication and evaluation of P(VDF/TrFE) piezoelectric film

P(VDF/TrFE) (with 75 mol% VDF, molecular weight 350,000 g/mol) was dissolved in dimethyl formamide (DMF) to give an approximately 20-wt.% P(VDF/TrFE) solution. A film was formed by casting the solution on a 0.3 mm thick aluminum plate substrate with area of 13×12 mm^2 that served as a backing electrode. The film was annealed at 145 °C for 2 h to enhance the crystallinity (β-form crystals), and an aluminum electrode was deposited by vacuum evaporation onto the surface of the film to form a front electrode. The P(VDF/TrFE) film, with a thickness of about 98 μm, was poled with an electric field Ep higher than the coercive field (Ep = ±75 MV m^{-1}). The complex admittance of the poled film was measured with a network analyzer (HP 4195A). Fig. 2 shows the piezoelectric resonance behavior of the P(VDF/TrFE) film [Air/Evaporated-Al (0.2 μm)/P(VDF/TrFE) (98 μm)/Al-plate (300 μm)]. The electromechanical coupling factor k_t was determined by fitting the resonance behavior using Mason's equivalent circuit [14,15]. The parameter values determined are listed in Table 1.

3. Influence of adhesive

To examine the loss induced by adhesive between the P(VDF/TrFE) film and the Al plate, at first the P(VDF/TrFE) film was directly deposited on the aluminum plate and measured with a network analyzer after poled. Then the Al-plate was dissolved in sodium hydroxide solution, and the film was bonded again to an aluminum plate using the epoxy adhesive [Araldite AR-R30 (2 liquid type)] at room temperature [curing conditions: 25 °C 24 h] with 128 kPa using a press machine (SSP-10A: Shimadzu GLC Ltd.). The thickness of the epoxy adhesive was measured by a scanning electron microscope (JSM-5800LV/JEOL) on the cross-sectional observation of the P(VDF/TrFE)/Epoxy-adhesive/Al-plate. The epoxy density (1.2 g cm^{-3}) was determined by forming a 10 mm cubic block made from the same epoxy adhesive, and the sound speed (2080 m/s) was measured by using the P(VDF/TrFE) transducer in water (25 °C). Fig. 3 compares the observed piezoelectric resonance behavior of the P(VDF/TrFE) film/epoxy (6 μm thickness)/Al plate and (from Fig. 2) the P(VDF/TrFE) film/Al plate. Fig. 4 shows the simulation of the transducer insertion loss (2TL) which was calculated from Mason's equivalent circuit. The transducer insertion loss (not including air loss) was higher by about 20 dB higher for a 6-μm epoxy layer. An irregular bonding layer, bubbles in the epoxy adhesive, and a kissing-bond are thought to be the cause of the increase in transducer insertion loss.

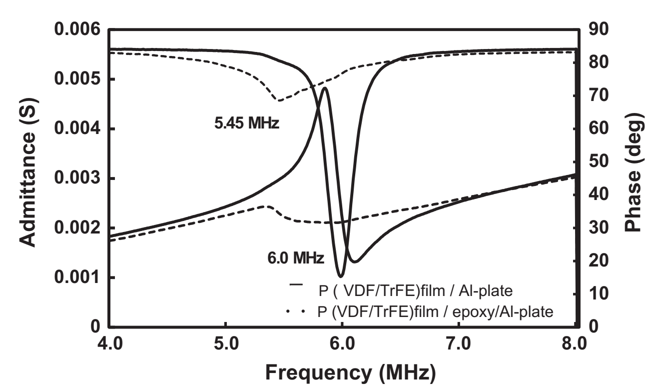

Fig. 3. Piezoelectric resonance behavior of P(VDF/TrFE) film/epoxy-adhesive/Al-plate and P(VDF/TrFE) film/Al-plate (present film).