# Recent Trend on Application of Piezoactve Polymers to Acoustics E.Fukada\*, M.Date, and H.Kodama Kobayasi Institute of Physical Research, Tokyo, Japan

#### Abstract

Noble sound shielding system is developed by the combination of a curved piezoelectric polymer film and a negative capacitance circuit. Incident sound produces the in-plane strain and polarization in the piezoelectric film. The charge induced on the electrode by polarization is amplified and fed-back to the electrode. The resulting electric field produces the inverse piezoelectric strain. The strain of the film is thus the sum of the sound-induced strain and the field–induced strain and can be arbitrarily increased or decreased by changing the feed back voltage. Subsequently the reflection and transmission of sound can be controlled. The complete isolation of sound is attained at a single frequency. The agreement of the value of the capacitance and its frequency dependence between the film and the capacitor in the circuit is required to achieve the sound shielding over a broad frequency range. Combining two PVDF films, about 50 dB transmission loss is observed from 200 Hz to 1000 Hz. The efficiency of shielding is more effective at the lower frequencies in contrast to the ordinary sound absorbing materials.

### Introduction

A number of investigation have been published on the application of piezoelectric polymers to acoustics. Lerche et al. reported a microphone utilizing a curved film of polyvinylidene fluoride (PVDF) [1]. Tamura et al. succeeded to commercialize headphones and tweeters using curved PVDF films [2]. Ohigasi [3] and Sussner [4] succeeded to produce ultrasound transducers in MHz range using thin PVDF films. Toda [5] recently reported an ultrasound transducer at the 10-100 kHz range using a curved PVDF film.

Date discovered an original method to superpose the direct and inverse piezoelectric effects on the piezoelectric materials [6]. Let us consider an electroded piezoelectric sample film with a capacitance Cs. On application of mechanical stress the charge Q is generated in the electrode of the sample due to piezoelectric polarization. If the sample is connected in parallel to a capacitance C, a part of charge Q flows into the capacitance C from the sample Cs. However, if a capacitor with a negative capacitance C (< 0) is connected, a part of charge in the electrode flows into the sample from the sample form the sample induces the electric field in the material, which generates the strain by the inverse piezoelectric effect. The strain is the sum of the stress-induced strain and the

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field-induced strain and thus can be controlled by the amount of the charge Q, which flows from the negative capacitance C (< 0) into the sample Cs. If Q>0, the charge in the electrode increases and the strain decreases, leading to the increase of the apparent elastic constant. If Q<0, the charge in the electrode decreases and the strain increases, leading to the decrease of the apparent elastic constant.

A circuit with the negative capacitance has been constructed using an operational amplifier. The significant change of Young's modulus of a piezoelectric film of polyvinylidene fluoride (PVDF) connected to such circuit has been observed [6].

The reflection and transmission of airborne sound by a film is governed by the difference of acoustic impedance between the air and the film material. The latter is represented by  $(Y)^{1/2}$ , where is the density and Y is the Young's modulus of the material. It is anticipated, therefore, that if the Young's modulus of the film is arbitrarily controlled, the reflection and transmission of sound by a piezoelectric film is also arbitrarily controlled.

Based on this electrical control of elasticity of piezoelectric film, a new sound shielding system has been proposed [7,8]. The present paper will describe the recent developments on this investigation.

In the previous applications for the acoustic transducers such as microphone and headphone, a curved round film of piezoelectric PVDF is installed in the device, being clamped in its periphery. The incident sound pressure generates the in-plane strain in the curved film. The piezoelectric polarization is thus generated in the thickness direction. The piezoelectric constant  $d_{31}$  is effective in this type of electromechanical transduction. On the other hand, the ordinary ultrasonic transducers utilize the resonance vibration in the thickness direction of the piezoelectric film. The piezoelectric constant  $d_{33}$  is effective in such applications.

## Acoustic Tube Experiment

As a standard apparatus for measuring the transmission loss and absorption of sound through polymer films, the acoustic tube experiment has been employed. As shown in Fig. 1, a PVDF film with the thickness of  $28 \,\mu$ m is installed in the middle of an acrylic tube of 43 mm diameter and 1 m length. The film is backed by a thin urethane form layer and a steel wire mesh to maintain the curved form. The periphery of the film is clamped to the tube. The electrodes of both surfaces of the film are connected to the negative capacitance circuit.

Using the two microphone method the correlations between the pressure of incident sound  $p_i$ , the pressure of reflected sound  $p_r$ , and the pressure of transmitted sound  $p_t$  are determined [7]. The transmission loss (TL) and absorption coefficient (AC) are defined by the following equations.

$$TL = 10 \log_{10} (p_i^2/p_t^2)$$
(1)  

$$AC = 1 - (p_r^2/p_i^2)$$
(2)



Fig.1 Acoustic tube experiment Fig.2 Geometry of curved film

#### **Theoretical Background**

The theoretical equation for the transmission loss through a curved membrane has been derived by Mokry [9]. For simplicity we assume uniform vibration of the curved film as shown in Fig.2. The sinusoidal displacement of the film in the radial direction is expressed as  $w(t) = W \exp(i\omega t)$  and the acoustic pressure difference between the film is expressed as  $\delta p(t) = P \exp(i\omega t)$ . The relation between W and P is given by

$$W = \frac{R^2}{h} \frac{P}{Y - \rho \omega^2 R^2}$$
(3)

where R is the radius of curvature, h the thickness,  $\rho$  the density,  $\omega$  the angular frequency and Y the Young's modulus of film. It is seen that W approaches infinity at the critical angular frequency  $\omega_0 = (1/R)\sqrt{Y/\rho}$ . Since the acoustic impedance Z of the film is given by  $\delta p/v$ ,

where v is the velocity in the radial direction, we have,

$$Z = \frac{P}{i\omega W} = i\omega\rho h \left( 1 - \frac{Y}{\rho \omega^2 R^2} \right)$$
(4)

Transmission loss (TL) is given by

$$TL = 10\log_{10} \left( 1 + \frac{Z}{2\rho_0 c_0} \right)^2$$
(5)

where  $\rho_0$  is the density of the air and  $c_0$  the sound velocity of the air. From Eqs. (4) and (5) we have,

$$TL = 10\log_{10}\left[1 + \frac{Y''}{\omega\varsigma} + \frac{(Y'')^2 + (Y' - \rho\omega^2 R^2)^2}{(2\omega\zeta)^2}\right]$$
(6)

where  $\zeta = c_0 \rho_0 R^2 / h$  and Y = Y' + i Y'' is the complex Young's modulus.



Fig.3 Theoretical curves for the transmission loss vs. frequency for a curved film.

Figure 3 shows the examples of the frequency dependence of TL calculated by Eq. (6) for a curved film of PVDF. The following values are assumed:  $h = 50 \ \mu m$ , R = 4, 8 and 16 cm,  $\rho = 1770 \ \text{kgm}^{-3}$ , Y = 3.7 GPa, and Y"/Y' = 0.1. For air the normal values  $\rho_0 = 1.293 \ \text{Kgm}^{-3}$  and  $c_0 = 331 \ \text{ms}^{-1}$  are assumed.

It is noted from Eq. (6) and Fig.3 that the value of TL increases with increasing Y', Y" and h and with decreasing R and  $\omega$ . It is remarkable that TL is higher at the lower frequencies. The efficiency of most sound absorbing materials is very low at the frequency range near 100 Hz. However, the predicted value of TL in Fig.3 attains 40 dB at 100 Hz for R = 8cm which is the ordinary value for the modulus of curvature used in experiments. It is also important to note that the 10 times increase of Young's modulus Y' causes 20 dB increase of TL over all frequencies. That is the reason why the control of elasticity of piezoelectric polymer films is useful to control the transmission loss TL.

#### Negative capacitance circuit

Figure 4 shows the negative capacitance circuit to increase the effective Young's modulus of the piezoelectric film and is named as the circuit H. The sample film is connected to the non-inverted (+) input of an operational amplifier. The output voltage is divided by a variable resistor R2 and R1 and the divided voltage is led to the inverted (-) input, which is then fed back to the electrode of the sample, because the two inputs are nearly equal-potential or virtually connected.

The output voltage  $V_{out}$  is related to the input voltages  $V_{in}^+$ ,  $V_{in}^-$  by  $V_{out} = G (V_{in}^+ - V_{in}^-)$ , where the amplification factor G is about 10<sup>5</sup>. The value of  $V_{in}^+ - V_{in}^-$  is extremely small compared to  $V_{out}$  and we can put  $V_{in}^+ = V_{in}^- = V$ . Thus we have

$$V = \frac{R_1}{R_1 + R_2} V_{out} \tag{7}$$

The charge  $\delta Q$  in the capacitance  $C_0$  is given by

$$\delta Q = C_0 (V - V_{out}) \tag{8}.$$

It is seen that the sign of  $\delta Q$  is negative. From Eqs. (7) and (8), the effective capacitance C of the circuit is given by

$$C = \frac{\delta Q}{V} = -C_0 \frac{R_2}{R_1} \tag{9}.$$

The input voltage from the piezoelectric sample finally results in the storage of charge  $-\delta Q$  in the capacitance  $C_0$  and the flow of charge  $\delta Q$  into the sample capacitance Cs. It will then lead to the decrease of the strain in the piezoelectric film and the increase of the transmission loss.



Fig. 4. Negative capacitance circuit H

Fig.5 Negative capacitance circuit S

Figure 5 shows the negative capacitance circuit to decrease the effective Young's modulus of the piezoelectric film and named as the circuit S. Here, the sample film and the capacitance of the circuit  $C_0$  are connected to the inverted (-) input of an operational amplifier. Eqs. (7)-(9) also hold in this case. However, owing to the nature of the operational amplifier, the phase of  $V_{out}$  is  $180^0$  inversed with respect to  $V_{in}$ . Therefore the charge  $\delta Q$  stored in the capacitance  $C_0$  in Eq. (8) is positive and thus the negative charge  $-\delta Q$  flows into the sample. It will lead to the increase of the strain in the piezoelectric film and the decrease of the transmission loss.

#### **Elasticity Control of Piezoelectric Films**

Let us consider the case that a piezoelectric film with a capacitance Cs is connected in parallel to a capacitor with a capacitance C. The rectangular coordinates assigned to the piezoelectric film is as follows: the 1-axis is the direction of orientation of polymer molecules, the 2-axis is perpendicular to the 1-axis and lies on the plane of the film, the 3-axis vertical to the plane of the film. The electric displacement  $D_3$  is produced by the stress  $T_1$  and their coefficient is the piezoelectric constant  $d_{31}$ . The constitutive equations for the strain  $S_1$ , the stress  $T_1$  the electric displacement  $D_3$ , and the electric field

E<sub>3</sub> are:

$$S_{1} = s_{11}^{E} T_{1} + d_{31} E_{3}$$
(10),  

$$D_{3} = d_{31} T_{1} + \epsilon_{33}^{T} E_{3}$$
(11),

$$D_3 = d_{31}T_1 + \epsilon_{33}E_3$$

where  $s_{11}^{E}$  the elastic compliance and  $\varepsilon_{33}^{T}$  the dielectric constant.

When the stress  $dT_1$  is applied to the film with the area A and thickness h, the charge  $dQ_1 = -AdD_3$ appears in the electrode of the film due to the direct piezoelectric effect. Then the charge  $dQ_2 = -dQ_1$  is induced in the capacitor C and generates the voltage change  $dV = dQ_2/C$ . This dV is fed back to the film and induces the strain change  $dS_1 = -d_{31}dV / h$  by the inverse piezoelectric effect. From Eqs.(10) and (11)

$$dS_{1} = s_{11}^{E} dT_{1} - d_{31} dV / h$$
(12)  
$$dQ_{1} = -A d_{31} dT_{1} + Cs dV$$
(13)

$$dQ_1 = -A d_{31} dT_1 + Cs dV$$
(13)

From Eqs. (12) and (13) we get the equation for the effective elastic compliance  $s_{11}$ :

$$s_{11} = s_{11}^{E} \left( 1 - \frac{k_{31}^{2}}{1 + C/Cs} \right)$$
(14).

The effective Young's modulus  $Y = 1 / s_{11}$  is also obtained:

$$Y = Y_0 \left( 1 + \frac{k_{31}^2}{1 - k_{31}^2 + C/Cs} \right)$$
(15),

where  $Y_0 = 1/s_{11}^{E}$  is the ordinary value of Young's modulus. Since  $k_{31} = 0.1$  for PVDF, when C/Cs = -  $(1 - k_{31}^2) = -0.99$ , Y =  $\pm \infty$  occurs. Practically, when C/Cs approaches -1, the sharp increase or decrease of the Young's modulus Y is observed.

From Eq.(9) we get,

$$\frac{C}{Cs} = -\frac{C_0}{Cs} \frac{R_2}{R_1} \tag{16}$$

Thus the value of Y can be controlled by adjusting  $R_2/R_1$  in the circuit if  $C_0/C_5$  is constant. The experimental results for the dependence of Y on C/Cs for a PVDF film is shown in Figure 6. As the frequency is changed, the values of C and Cs vary with frequency. Both C and Cs should be expressed as the complex quantities including the loss factor.

To take into account the frequency dependence of Y, we must represent Cs as  $Cs^* = Cs' (1 - i \tan \delta s)$ , where  $tan\delta s$  is the dielectric loss of the film. Correspondingly we should connect a resistance  $R_0$  in series to the capacitance C<sub>0</sub> and represent C as  $C^* = C_0(1 - i \tan \delta) / (1 + \tan^2 \delta)$ , where  $\tan \delta =$  $2\pi fC_0R_0$ . The agreement of tands and tand occurs only at a frequency  $f_0$ , where tands = 2  $f_0C_0R_0$ . Thus we have the formula for  $C^* / Cs^*$  neglecting tan<sup>2</sup> $\delta$  compared to 1

$$\frac{C^*}{Cs^*} = -\frac{C_0}{Cs'} \frac{R_2}{R_1} \left\{ 1 + i \left( \tan \delta s - 2\pi f C_0 R_0 \right) \right\}$$
(17)

$$= -\frac{C_0}{Cs'} \frac{R_2}{R_1} \left\{ 1 + i \tan \delta s \left( 1 - \frac{f}{f_0} \right) \right\}$$
(18)

Eq.(18) indicates that the imaginary part of  $C^*/Cs^*$  changes its sign at a frequency  $f_0$ . In the frequency dependence of the transmission loss, which will be shown in Figure 10, a sharp peak followed by a dip appears at the frequency of this phase reversal.

### Control of Young's modulus

Dynamic Young's modulus of a rectangular film of PVDF with a size 25 x 5 x 0.05 mm was measured as a function of C/Cs and frequency by Rheolograph-Solid (Toyo-Seiki). Figure 8 shows the results measured at 9.8 Hz. C/Cs was varied by changing the ratio  $R_2/R_1$  in Eq.(17). If C/Cs is close to -1, the sharp increase and decrease of Y is observed in accord with Eq.(15).



Figure 9 shows the variation of the real part Y' and the imaginary part of Y" of the Young's modulus in the frequency range between 1 and 20 Hz as the circuit H is connected.. The value of C/Cs was fixed at -0.988 and  $f_0 = 9.8$  Hz was chosen. Y' shows a peak at  $f_0$  and Y" a peak below  $f_0$  and a dip above  $f_0$  suggesting the phase reversal of vibration at  $f_0$ . When the circuit S is connected, Y" showed a dip and Y" showed a minimum and a maximum with increasing frequency around  $f_0$ .

### **Transmission loss**

Figure 10 illustrates the frequency dependence of the transmission loss (TL) through a curved PVDF film. The circuit H was tuned at 1 kHz., TL increases with increasing the feed back voltage. Figure 11 indicates the output voltage of a microphone placed behind the film plotted against the feed back voltage. The output voltage decreases to zero and then increases with the reversed phase with



increasing the feed back voltage. The strain of the film is the sum of the strain induced by the incident Phase Reversal

Fig. 10 The increase of TL at a frequency 1 kHz

sound and the strain induced by the feed back voltage. The latter cancels the former with increasing feed back voltage, but above a certain value of the feed back voltage the latter exceeds the former resulting the phase reversal of vibration. Figure 12 shows the change of the transmission loss as the circuit S is connected. TL decreases at the tuned frequency 1 kHz.

## Sound shield over a broad frequency range

For practical applications the attenuation of sound over a broad range of frequency is necessary. The increase of transmission loss, which is the result of the increase of the Young's modulus of the film, is achieved by adjusting C/Cs as close as -1. Because C and Cs have different frequency character, the critical condition is obtained only at one frequency  $f_0$  shown in Eq.(18). The values of tan $\delta$  and tan $\delta$ s



Fig.12 The decrease of TL with the circuit S

Fig.13 The increase of TL at 200-2000 Hz.

are different at frequencies other than  $f_0$ . In other words, the magnitude and loss factor of C and Cs should have the same frequency dependence for all frequencies.

The attempt was made to construct a combined network of capacitors and resistors with the distribution of the relaxation time as same as Cs, but only partially successful. The solution to obtain the frequency independent C/Cs was to employ a PVDF film to replace  $C_0$  and  $R_0$ . Figure 13 illustrates one example of the transmission loss vs. frequency curves measured from 200 Hz to 2 kHz. About 7 dB increase of TL was observed for all frequencies.

In another attempt, two PVDF films were installed to obtain the further increase of TL. The output voltage of the circuit H connected to the first film was amplified again and led to the second film, so that the canceling effect is more increased. About 30 dB increase of TL was easily observed over a frequency range from 200 Hz to 2 kHz.

## Conclusions

Superposition of the direct and inverse piezoelectric effects is realized by means of simple circuits employing an operational amplifier. The remarkable change of the elastic constant of piezoelectric materials can be induced. The acoustic impedance of piezoelectric films can be also controlled by this technique. Employing a curved structure of piezoelectric PVDF film, controllable sound shielding systems have been demonstrated. The transmission loss can be increased or decreased under control at any frequency.

The remarkable feature of the proposed sound shielding system is that the transmission loss increases with decreasing frequency of sound. In most ordinary sound absorbing materials, the transmission loss decreases with decreasing frequency. Therefore, the shielding of low frequency noise has been a difficult task in engineering.

The present work describes some basic knowledge and experimental results on PVDF films with a small area  $4 \times 4$  cm. Future studies should be directed to the construction of sound shielding systems with a much larger area. The feature of such completed systems would be low weight, low cost and the effectiveness at low frequencies.

The elastic properties of piezoelectric ceramics are also controllable by connecting negative capacitance circuits to them. The considerable decrease of resonant frequency has been demonstrated for a system combining a piezoelectric PZT plate and a metal cylinder [8]. This technique should also be useful for the prevention of vibration through solid materials.

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