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A New Approach of Polyvinylidene Fluoride (PVDF) Poling Method for Higher Electric Response

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Good response, in particular high sensitivity, of polyvinylidene fluoride (PVDF) attracts a lot of research works on its manufacturing process and applications. Polarization is an important factor that is influential to the performance of PVDF. In this article, a new polarization method using ITO (Indium Tin Oxide) glass is proposed. Different from the conventional poling method using metal eletrode coated onto PVDF film, ITO glass is used as electrode to transmit the electric field voltage into PVDF film for poling. With the advantage of applying higher electric field in the process of polarization and eliminating the occurrence of flashover and arcing efficiently, this approach can thus make PVDF film gain better electric output response. Also, thinner film is constructed would be another advantage of sensitivity improvement. Comparison with conventional method for regular polarization area is carried out by using both FTIR and DSC equipment. Piezoelectric charge constant d_{33} *and electricity output response as well as sensitivity of PVDF are measured in experiment, which indicate ITO glass poling method is superior to conventional technique.*

Keywords PVDF; ITO glass; poling.

I. Introduction

Piezoelectric PVDF film is a type of piezoelectric polymer that has been widely used for many applications such as sensor or energy harvesting due to their good sensitivity, flexibility, mechanical strength and displacement [1–9]. Generally, there are five main procedures in making piezoelectric PVDF film including: stretching PVDF films, depositing electrode onto PVDF surface, poling process, wiring in copper plate, and laminating with plastic. Fundamentally, stretching and poling processes determine the characteristic of PVDF. PVDF is a semi-crystalline polymer, commonly existing with α-phase in non-polar crystalline. Usually β-phase is the most useful phase for its piezoelectric and ferroelectric properties, which can be achieved from the α-phase by mechanical stretching or poling [10]. Including our previous studies [11], quite a few of researches have been investigated on the piezoelectric effect in the process of stretching the PVDF films [12–18]. Since stretching process implies skeleton chain being oriented particularly at stretching direction, transformation of α- into β-phase is largely occurred. However, the remaining α-phase may not be influenced by stretching, which can be reoriented in the later poling process.

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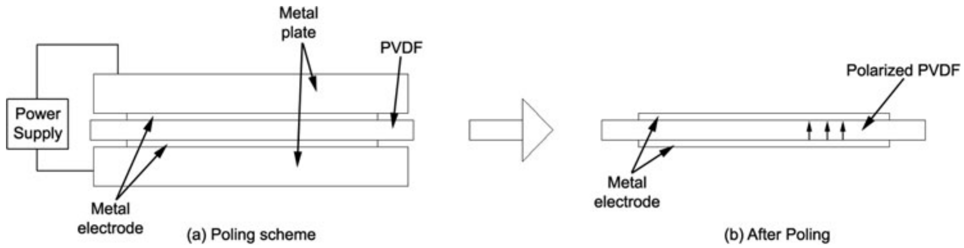

Figure 1. Conventional poling.

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Therefore, poling is very important and is the focus in this article. A number of conventional research works investigated poling on the stretched film, whereas temperature is gradually increased from room temperature to 80°C and then slowly cooled down [12, 17–21]. Conventional poling is illustrated in Figure 1. Poling process at room temperature is called cold poling in general. There are difficulties of poling at room temperature such as limitation of applied electric field and occurrence of arcing or flashover under high electric field [22]. To overcome this problem, paraffin oil [22] or silicone oil [23] or vacuum [24] were usually used as a medium of eliminating oxygen between films and plate in high electric field. Other approaches were proposed for the same purpose, for instance, dielectric plate [25] or silicone rubber O ring [26] was employed between polymer and electrode, or current-limiting resistor was added between the power supply and electrode [27]. Corona poling, electron beam poling, photo-thermal and photo-induced are other poling methods. For instance, in photo-thermal and photo-induced poling, ITO glass has been used as conductive electrode. Polymer PVDF is deposited onto ITO glass and metal electrode is deposited on top of PVDF. Light produced by laser beam would irradiate PVDF through ITO glass to generate positive and negative polarization in small area [28, 29]. Nonetheless, the equipment setup of these methods is complex and difficult, and the polarization area is very limited.

Applying high electric field is desired since the high electrostatic force will efficiently compress the film to increase chain separation that facilitates dipole orientation. As known that resistance of the metal electrode used in conventional poling is about zero so that almost all applied voltage and current are transmitted to PVDF. Therefore, applying high electric field would easily cause flashover or arcing and induce electrical breakdown so as to inhibit poling process. Avoiding from such phenomenon an index of allowable electric field is determined. For example, among the research works using conventional method, maximum electric field that could be applied is 500 kV/cm [18]. It is noted that the above conventional and photo poling methods basically utilize metal electrode coated onto surface of polymer that would induce flashover or arcing and cause electrical breakdown while applying high electric field [30]. As known, higher electric field is applied during poling process will induce the non-polar crystal to become polar crystal form, hence more β-phase content is gained, and the piezoelectric charge constant d_{33} is increased [31, 32]. Moreover, as seen in Figure 1, the electrode attached to the associated metal plate would reduce poling effect and influence output performance due to several defects: (1) large surface roughness, (2) gas trap in between the metal plate and electrode, (3) less flexibility. How to improve the above poling conditions to enhance applied voltage is therefore demanding. ITO is considered to use mainly because it is a high degenerate n-type semiconductor with low electrical resistivity of $2 \times 10^{-4} \sim 4 \times 10^{-4}$ Ωcm can be used as a conducting electrode, and it has wide band gap as well as small surface roughness [33]. While using ITO as a conducting electrode, it can deliver most of the electric field. Different from the conventional poling method makes use of metal electrode with nearly zero resistance that would produce large current and easily cause flashover or arcing, ITO with somewhat resistance itself would reduce current and allow applying higher electric field. With wide band gap, ITO is able to obtain high peak velocities, so that higher electric field can be transmitted through ITO and applied to the PVDF film [34]. Poling process at room temperature is preferable for the advantages of simplicity and convenience. Under such circumstances, gas trap likely exists in between the metal plate and electrode due to large surface roughness about 170 nm of the metal plate. With much smaller surface roughness below 18 nm, ITO glass can thus reduce gas trap in between the PVDF film and ITO glass. Also, flexibility of ITO glass can preserve good contact between ITO glass and PVDF film, thus gas trap would be eliminated efficiently. With the above good characteristics, thinner PVDF is therefore obtained and expected to have better response [35]. In our approach, distinct from photo-thermal and photo-induced poling using laser beam for polarization in small area, ITO glass is utilized as dielectric conductivity to transmit electric field from power source to PVDF film and complete polarization. Also, it is suitable for regular dimension of polarization employed with higher electric poling voltage up to 700 kV/cm, which cannot be achieved by the aforementioned methods.

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II. Experiment

A. *Poling*

The original sample PVDF film with 120μm thickness is used. Film is uniaxially stretched until the thickness becomes 10 μm at 80°C to change α-phase into β-phase and gradually cool down to room temperature. New poling technique using ITO glass is proposed in this article. Like cold poling, the poling temperature is kept at room temperature. To replace metal electrode and metal plate, ITO glass is used in our approach that consists of an ITO layer of 150 nm thickness coated onto a soda lime float glass of 1.1 mm to obtain electrical resistance of 12 ohm/sq [36]. When electric field is applied to the sandwich structure constructed with ITO glass-PVDF-ITO glass as shown in Figure 2, the current flow is reduced; thus, it can eliminate the occurrence of flashover or arcing [26]. Because of the smooth surface of ITO glass, complete contact with the PVDF film is achieved. The electric field in the range of 100$\sim$700 kV/cm is applied through the wire connected to ITO glass at room temperature for about 15 minutes. As shown in Figure 3, two pieces of Cu plates are attached to both sides of PVDF for transmitting the electricity of PVDF,

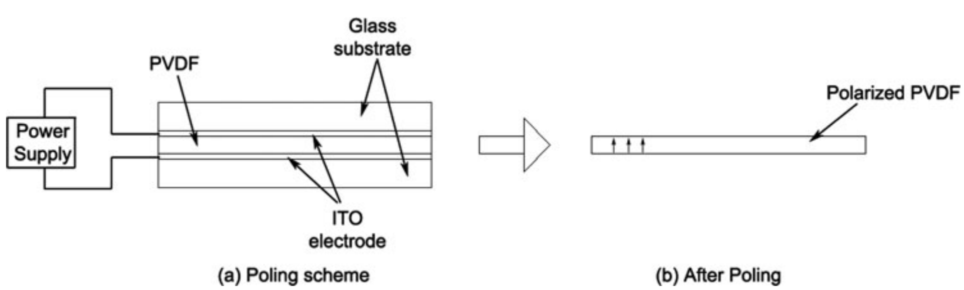

Figure 2. ITO poling.

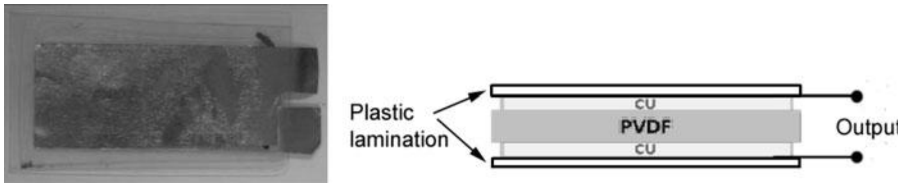

Figure 3. PVDF with Cu and plastic lamination (Color figure available online).

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and a plastic lamination used to embed and protect the Cu plates and the PVDF film. A same-size PVDF sample by using conventional poling method is selected as an example for comparison with the one by using ITO glass poling method.

B. Characterization

PVDF samples weight 4 mg are used to investigate the content of β-phase by means of Differential Scanning Calorimeter (DSC) equipment and Fourier Transformed Infrared Spectroscopy (FTIR). DSC model of Perkin-Elmer DSC 7 is operated at 10°C/min heating rate. The melting heat enthalpy is measured in the range of 50°C~200°C. Perkin-Elmer FTIR Spectrum One is set in the range of 450–1600 cm^{-1} wave number with average of 32 scans and a resolution of 4 cm^{-1}. By measuring the enthalpy using DSC, the degree of crystallinity is calculated by [37].

$$X = \frac{\Delta H_f}{\Delta H_f^*} \tag{1}$$

where ΔH_f is the fusion enthalpy of the sample PVDF and ΔH_f^* is fusion enthalpy of sample PVDF with 100% crystallinity, which is 102.5 J/g given in [38].

Fraction of β-phase crystals $F(\beta)$ is calculated by [39–41]

$$F(\beta) = \frac{A_\beta}{(X_\alpha / X_\beta) A_\beta + A_\beta} \tag{2}$$

where α and β subscripts refer to the crystalline phases; A is the absorption coefficient at the respective wave number read from FTIR; X is degree of crystallinity of each phase defined in eq.(1).

C. Piezoelectric Charge Constant and Sensitivity

Piezoelectric charge constant d_{33} of PVDF is characterized using D33 meter (Sinocera model YE2730). The output voltage signal and the input force are measured by SR785 Dynamic Signal Analyzer while using Hammer (PCB Piezotronics 086E80) to implement force onto the PVDF sample. Sensitivity of PVDF sample is defined as the output voltage of electric response divided by the input force [42].

III. Result and Discussion

According to the reference works [43, 44] and our experiment, stretching ratio over 3 is good enough for later poling. After stretching and/or poling process, α-phase will transform into β-phase. DSC profile of PVDF sample is shown in Figure 4. As seen, the crystallization

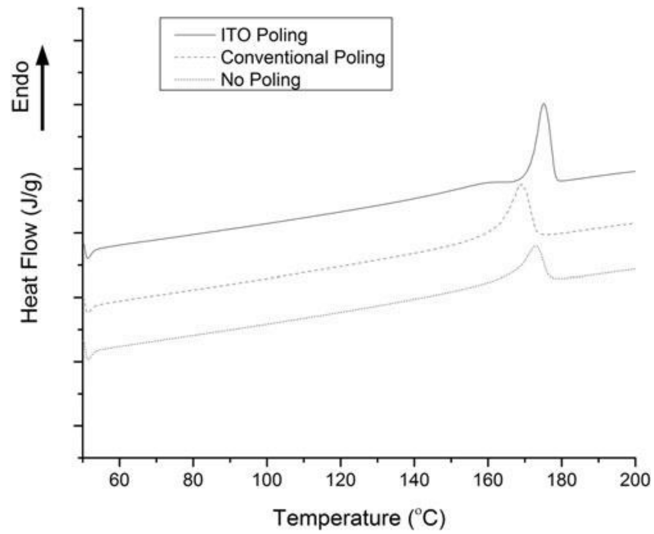

Figure 4. DSC profile with/without poling (Color figure available online).

endothermic peak becomes higher after poling. Enthalpy of PVDF sample is obtained by measuring the area of each peak in Figure 4 and listed in Table 1. With the measured fusion enthalpy, degree of crystallinity X of PVDF sample is calculated as 0.44 and 0.46 for conventional and ITO poling methods respectively by eq.(1) and listed in Table 1. It indicates that degree of crystallinity is higher while using ITO method because high electric field orients the dipole of PVDF film efficiently during poling process.

The β-phase crystallinity was also confirmed by FTIR spectra as shown in Figure 5. It is seen that the absorption peaks occur at 765 cm^{-1} for α-phase and 841 cm^{-1} for β-phase. Absorption coefficient of A_α and A_β are read *1.482* and *1.361* from the FTIR equipment. By using eq.(2), the β-phase fraction $F(\beta)$ after stretching process is calculated about 80.2%, 82.3% and 85.7% for the conditions of no-poling, conventional and ITO poling, respectively. As seen in Table 2, after poling process, the β-phase fraction is increased become 82.3% and 85.7%. This increasing fraction of β-phase results from more α-phase dipole orientation is changed when applying high electric field.

Figure 6 shows that the piezoelectric charge constant d_{33} of PVDF film produced by using ITO glass poling method and by conventional poling method. As seen in Figure 6, the piezoelectric charge constant of the proposed ITO glass poling method is greater than the conventional method. In the range of 100∼400 kV/cm higher applied electric field would have larger d_{33} for both conventional and ITO poling method. However, greater value is

Table 1
Enthalpy and degree of crystallinity

Parameter\Condition	ΔH (J/g)	X
No Poling	40.74	0.39
Conventional Poling	45.35	0.44
ITO Poling	47.29	0.46

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Table 2
β-phase fraction

Parameter\Condition	F(β)%
No Poling	80.2
Conventional Poling	82.3
ITO Poling	85.7

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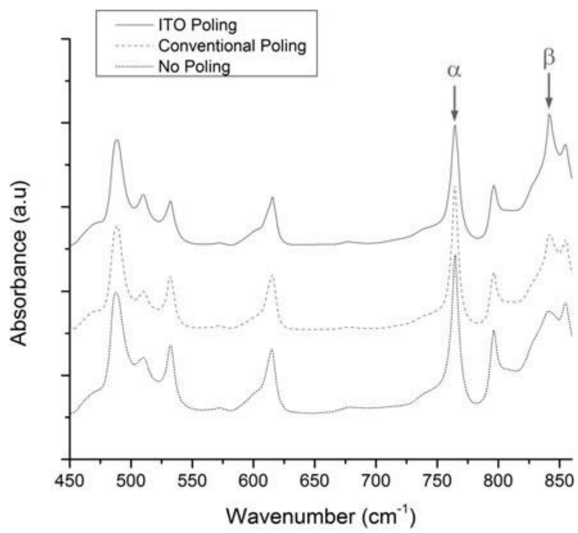

Figure 5. FTIR spectra with/without poling (Color figure available online).

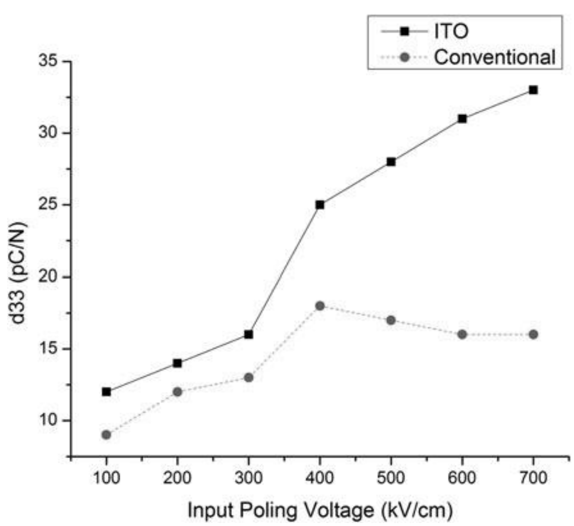

Figure 6. d_{33} for different poling voltage (Color figure available online).

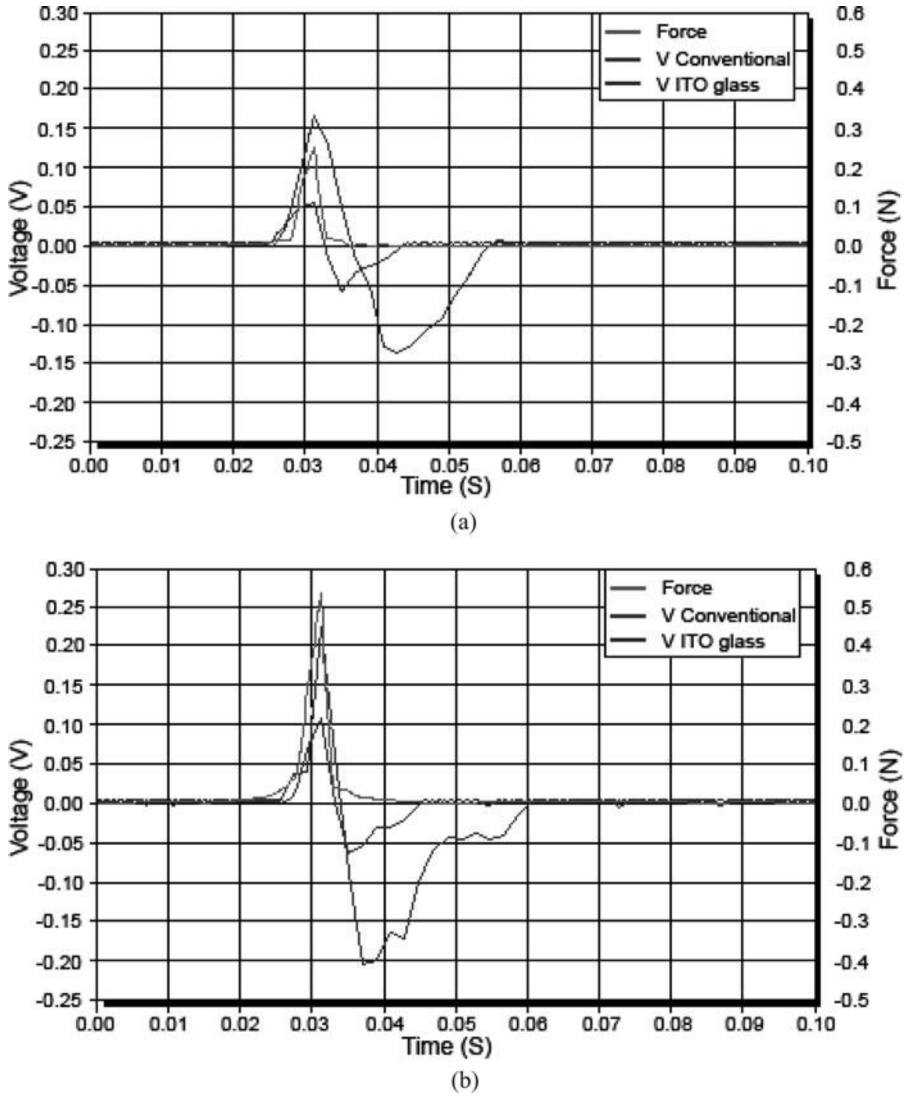

Figure 7. Input force and output electric voltage (a) Light input force F = 0.26N, (b) Hard input force F = 0.54N.

reached by using ITO poling method than conventional method. The lower d_{33} while using conventional method is probably due to poor conductivity between the metal electrode and the PVDF film so as to cause voltage drop during poling process [45]. On the contrary, ITO layer deposited onto glass provides good conductivity, so voltage drop can be minimized. As shown in Figure 6, since flashover and arcing happens when poling voltage higher than 400 kv/cm in conventional poling method, d_{33} is abruptly decreased. On the other hand, ITO with resistance itself would reduce current flow to avoid from flashover or arcing, so d_{33} continues increasing when applying higher electric field until the maximum allowable poling voltage of 700 kv/cm is reached.

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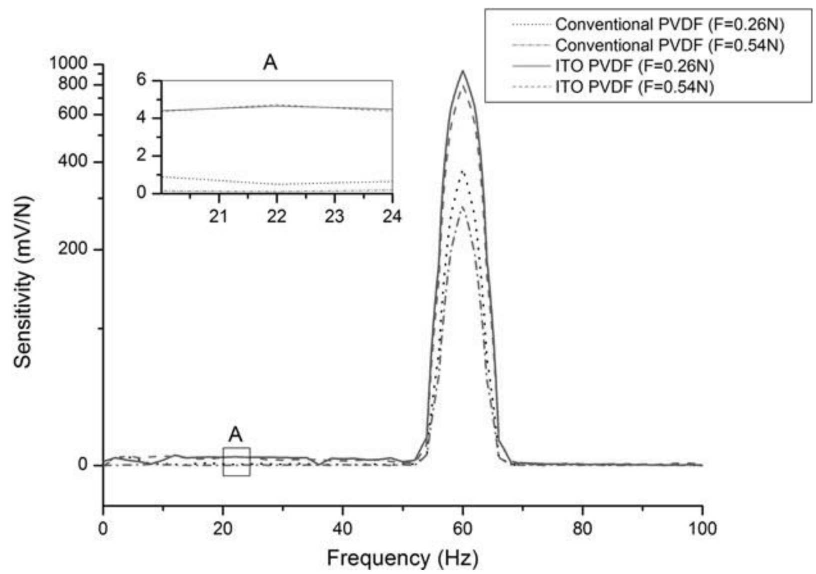

Figure 8. Sensitivity vs frequency (Color figure available online).

For the piezoelectric voltage coefficient is defined as [46]

$$g_{33} = d_{33}/(\varepsilon_r \varepsilon_0) \tag{3}$$

where ε_r and ε_0 are relative permittivity and free space dielectric permittivity, respectively. As known, the larger d_{33} is the larger g_{33} gains so that better output voltage response is achieved. Large surface roughness and stiffness appears while using metal plate in conventional poling process, therefore oxygen is easily existing in the gap that will cause flashover or arcing. Nonetheless, ITO glass has the advantages of smooth surface and high flexibility [47] to preserve good contact between the ITO glass and PVDF film, hence oxygen trap would be eliminated efficiently.

By using hammer, light and hard impulse force about 0.26N and 0.54N respectively is applied vertically to the PVDF film and the voltage response of PVDF film is measured by FFT analyzer and shown in Figure 7. Definition of sensitivity of sensor is the output electricity response divided by the input force [42]. Then, sensitivity against frequency is obtained and shown in Figure 8. In comparison with the outcome of conventional method, the proposed ITO glass poling method will have better sensitivity performance. Note that, sensitivity is drastically increased near the resonance frequency, but very small away from the resonance zone.

IV. Conclusion

As compared to the conventional poling method, much higher electric field up to 700 kV/cm can be applied to the PVDF film while using the proposed ITO glass poling method so that noticeably increasing d_{33} property of PVDF as well as obtaining better output electric response. Also, ITO glass with nonzero resistance would reduce certain amount of current flow. Therefore, it can effectively extend the poling voltage range and reduce the occurrence possibility of flashover or arcing. Moreover, using ITO glass as electrode

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will replace metal electrode and metal plate. Therefore, thinner PVDF is easily produced, which is instrumental to improve sensitivity. Due to smaller surface roughness and higher flexibility of ITO glass, good surface contact between ITO glass and PVDF film is thus preserved, that can eliminate gases trap in between ITO glass and PVDF film in the process of poling. To sum up, there are several noteworthy advantages in ITO glass poling method that cannot be reached by conventional poling method using metal electrode.

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