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# Structural, Dielectric and Piezoelectric Properties of PZT/PVDF Composites Prepared by Hot Press Method

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***Abstract* - The 0-3 PZT/PVDF piezoelectric composites are prepared separately by hot-press. The effects of the PZT content and the shaping-process on the composites are studied. The experimental results indicate that composites with 70% PZT nanopowders prepared by the hot-press method exhibit excellent piezoelectric and dielectric properties. This is mainly attributed to the favourable coupling of the two materials in the process of the hot press and the formation of the $\beta$-type PVDF, which possesses better electric properties.**

***Keywords* - *PVDF; PZT; composites; electrospinning; ferroelectric; piezoelectric; 0-3 composite.***

## I. INTRODUCTION

In recent years, the lead zirconate titanate (PZT) ceramicsmhas been widely used in sensors, actuators, hydrophones and ultrasonic transducers because of its excellent piezoelectric properties. As a result, the ferroelectric ceramic-polymer composites are the promising materials for applications in high-pressure sensors, hydrophones and shock accelerometers [1, 5, 8, 11, 12, 14, 16, 26, 28, 30, 31].

The simplest ceramic-polymer composite is that which consists of ceramic particles randomly dispersed in a polymer matrix, which is well known as 0-3 connectivity. Then, the PZT ceramic particles with excellent piezoelectric properties were evenly dispersed in a three-dimensional connected flexible polymer matrix. This type of composite has excellent comprehensive properties. Hot press technique that combines solution and melt processing was found to be the better method for the preparation of ceramic-polymer composites. Consequently, the 0-3 polymer-ceramic composites, especially of the ferroelectric ceramics such as PZT and PMN-PT and the ferroelectric polymer PVDF (polyvinylidene fluoride), have been the subject of a lot of research work**,** and detailed studies were reported on their piezoelectric, dielectric and pyroelectric properties [2, 3, 5, 9, 12, 14, 15, 18, 24, 27, 29, 34].

## II. EXPERIMENTAL SECTION

### A. *Materials*

PVDF of number-average molecular weight 534,000 supplied by Aldrich chemistry, France, The PZT ceramic ($d_{33}$ = 317 pC/N, $\varepsilon_r$ = 1800), which was prepared via the conventional solid-state reaction method, PZT powers are prepared with a mortar and pestle, the mean particle size of the PZT powders is about 200 – 500 nm (Figure 1). Dimethyl formamide (DMF) supplied by Merck and acetone (Merck, 99.7%), India, were used in this study.

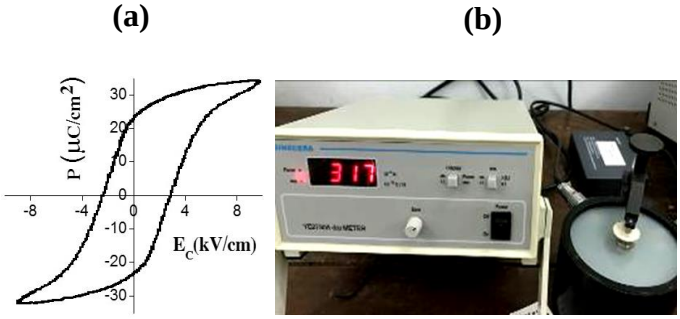


Figure 1. P-E hysteresis loop of PZT ceramic sample and the d33 values of PZT disc in the Model YE2730 meter

### B. *Devices*

The crystalline structure analysis was performed at room temperature using an X-ray diffraction system (XRD, Bruker D8 Advance, Germany) with Cu $K_\alpha$ radiation ($\lambda$= 0.154 nm). The surface morphology was examined using a scanning electron microscopy (SEM, Nova NanoSEM 450-FEI) operated at an accelerating voltage of 10–20 kV)

The polarization-electric field (P−E) hysteresis loops were measured with a HIOKI 3532 (Radiant Technologies) which testing unit connected with a high-voltage interface.

Dielectric properties of the materials were obtained together using an impedance analyzer (Agilent 4396B, Agilent Technologies, America, HIOKI3532) by measuring the capacitance and phase angle of the specimens from room temperature to 150$^o$C.

C. *Hot press method* combines solution and melt processing techniques. PVDF was dissolved in DMF/Acetone solution and the solution is concentrated to get an optimum viscosity for the loading of ceramic powder. PZT powder was added and stirred well to get a uniform distribution of the filler. The prepared slurry of PZT/PVDF was then coagulated by the addition of nonsolvent and dried, the obtained powders were dried and pressed into disks specimens with a diameter of 12 mm and a thickness of 1.2 mm only by uniaxial hot-pressing

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at 140°C with a pressure of 100 Mpa. The composite samples were prepared for 0.3 to 0.7 weight fractions, Table 1.
The silver pastes were fired at 120°C for 30 minutes on both sides of these sintered bulks as electrodes for electrical measurements.

**Table 1.** Samples of PZT/PVDF

| Samples | | Composite compound ratio | $\rho$ (g/cm$^3$) |
|---|---|---|---|
| *No.* | *Name of products* | | |
| *1* | F16 | PVDF 16 wt% + 0wt% PZT | 1.74 |
| *2* | F-P30 | PVDF 16 wt% + 30wt% PZT | 3.41 |
| *3* | F-P40 | PVDF 16 wt% + 40wt% PZT | 4.10 |
| *4* | F-P50 | PVDF 16 wt% + 50wt% PZT | 4.70 |
| *5* | F-P60 | PVDF 16 wt% + 60wt% PZT | 5.30 |
| *6* | F-P70 | PVDF 16 wt% + 70wt% PZT | 5.87 |
| *7* | PZT | PVDF 0 wt% + 100wt% PZT | 7.63 |

## III. RESULTS AND DISCUSSION

### *A. Density and porosity*

The experimental density values of the composite were measured and the theoretical density values were calculated using the formula (1), Fig 2a

$$\rho_c = \rho_1(1-x) + \rho_2 x \quad (1)$$

where $x$ represents the volume fraction of the ceramic phase and $\rho_c$, $\rho_1$ and $\rho_2$ are the densities of the composite, polymer and ceramic respectively [6, 18, 19, 21].

The densities of the composite samples were determined by weighing samples of known sizes accurately using a digital balance of accuracy 0.01 g resolution. Fig. 2 shows the variation in the experimental and calculated density values for the PVDF-PZT composites with the volume fraction of PZT. $\rho_{cal}$ increases linearly with increasing volume fraction of PZT, since the density of PZT (7630 kg/m$^3$) is higher than that of PVDF (1740 kg/m$^3$). Obviously the experimental density exhibits a linear increase with increasing volume fraction of PZT and especially for the volume fraction from 0.3 to 0.7, it stays unchanged with the theoretically calculated values.

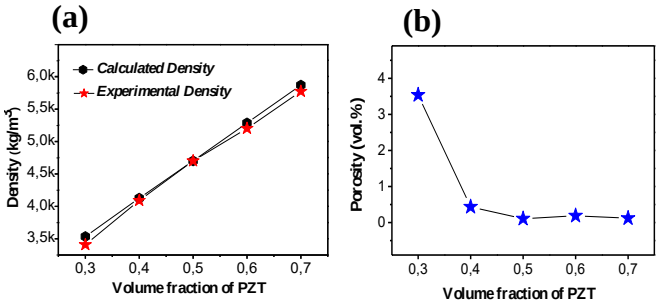


Figure 2. (a) Experimental and calculated density, (b) Porosity of composites at different volume fractions of PZT.

Furthermore the experimental density is smaller than calculated one at fixed volume fraction of the PZT, which may result from the presence of pores in composites. The porosity P, of composites can be calculated from the experimental and calculated density of composites using equation (2), Fig 2b.

$$P = 1 - \frac{\rho_{exp}}{\rho_{cal}} = 1 - \rho_{rel} \quad (2)$$

### *B. Crystalline phase analysis*

- *Surface morphology*

The microstructure of the 0-3 PZT/PVDF piezoelectric composite with 30 - 70 wt% PZT is shown in Fig. 3. It shows the typical characteristic of "particle connection". Quite a few PVDFs exist by the shaping of PZT grains. Furthermore, the SEM image also shows that the PZT grains are wrapped by the PVDF, which indicates the connection characteristic of "wrap and curl". The crevice is much less in the pellets of 0-3 PZT/PVDF piezoelectric composite material prepared by hot-press process, the connection between the grains of piezoelectric composite is much closer, and the grain size is also bigger [1-4, 9, 15, 16, 28-34, 37, 38].

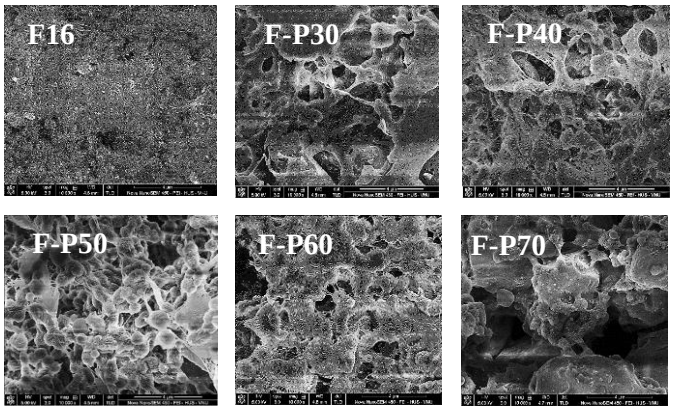


Figure 3. SEM images and grain size distribution for PZT/PVDF composites with different PZT content.

Figure 4 shows the schematic illustrations of the microstructures for the PZT/PVDF composites prepared by hot-press process. It explains the influence of the shaping-processes on the morphologies of the PZT/PVDF. For the low PZT content, the PVDF exists in the grain boundary of the PZT particles, which hold much more voids and porosities and are detrimental for polarization of the composites and the movement of ferroelectric domains. Accordingly, the electric properties are much lower.

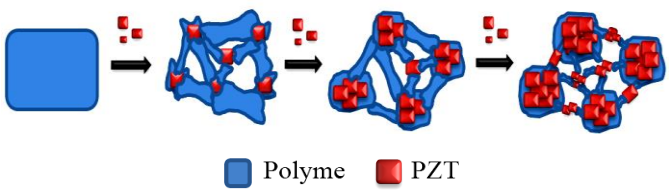


Figure 4. Schematic illustration of the microstructures for the PZT/PVDF composites prepared hot-press process with an increase in the PZT content

However, for the hot-press method with higher PZT content, the PZT grains are wrapped by the PVDF, indicating the connection characteristic of wrap and curl. The particles of PZT are much closer and the density of the composite is also higher, which are in favour of the polarization of the PZT/PVDF composites and will enhance the electric

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properties. Moreover, the formation of the $\beta$-type PVDF are also important to the improvement of the electric properties.

- *XRAY Analysis*

Figure 5b shows the XRD patterns of the PZT nanopowders, 50 wt% PZT/PVDF composites and PVDF prepared by the hot-press method, respectively. The single and sharp intensity peaks indicate the formation of the perovskite phase of the PZT/PVDF composite. Moreover, the composite is free from the pyrochlore phase, which is considered unwanted in the PZT system. Compared with the XRD pattern of the PVDF, it also can be observed the presence of the peak of the PVDF between 17.3 - 20.7°. It is therefore presumed the formation of the $\beta$-type crystalline, which is considered possesses of better electric properties [1, 3, 7, 14, 17, 19, 20, 23, 25, 28-30, 35, 39, 40].

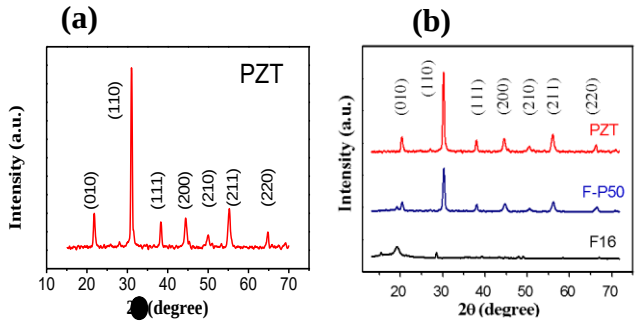


Figure 5. (a) SEM photograph and XRD pattern of the PZT nanopowders calcined at 1050°C for 2 h, (b) XRD patterns of PZT ceramic, PZT/PVDF composites and PVDF powders.

## C. The electrical properties of PZT/PVDF composites

- *Dielectric constant*

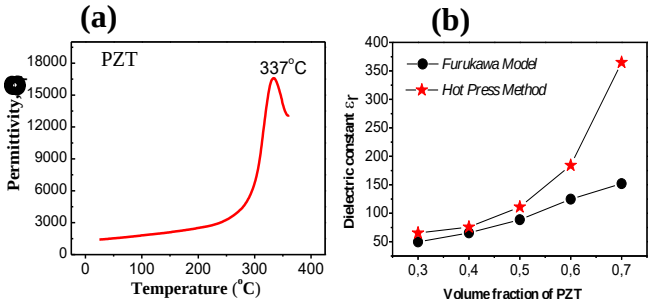


Figure 6. (a) The temperature dependence of dielectric constant measured at 1 kHz for the PZT ceramics.(b) Influence of PZT content on the dielectric $\varepsilon$ of the PZT/PVDF composites.

Variation of dielectric constant with temperature for various volume fractions of the composite poled at 30 kV/cm is shown in Fig. 6b. A steady dielectric constant values are observed in case of volume fraction from 0.3 to 0.7 till 140 °C and a steeper change is observed for volume fractions 0.6 and 0.7. Hence it is observed that the rate of variation of dielectric constant with temperature steadily increases with the volume fraction of PZT. This may be due to the fact that the poled PZT particles generate an internal field which favors the orientation of the PVDF molecules. At higher temperatures the molecules are more mobile and a higher concentration of PZT dipoles enables easier orientation of the molecules causing a steeper increase in the dielectric constant with temperature. A similar report has been presented by several authors [1, 2, 4, 6, 10, 13, 15-24, 28, 29, 33-38].

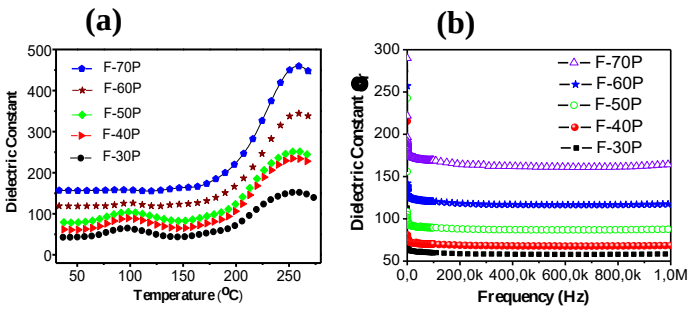


Figure 7. (a) Plot between dielectric constant with temperature, (b) variation in dielectric constant with frequency of PZT/PVDF composite

- *Ferroelectric properties*

A series of P–E hysteresis loops for PZT/PVDF composite measured at room temperature were shown in Fig. 8. The results showed that the composites with PZT volume fraction 0.3 to 0.7 exhibited non-saturated P–E hysteresis loop. It seems that the applied field of 30kV/cm was the main cause for the observed non-saturated hysteresis behavior in this study. It is also be noted that when increasing the PZT content to beyond 0.7 volume fraction, values of remnant polarization and coercive field increases as shown in table 2.

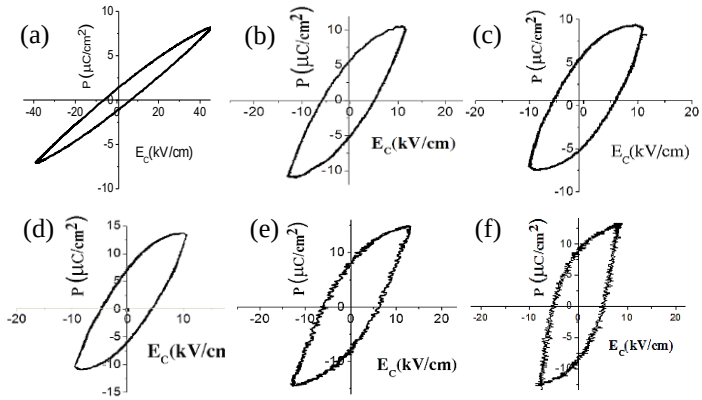


Figure 8. The P–E hysteresis loops for F16(a) and F16-x%PZT composites with different concentrations of PZT, x= 30 (b), 40 (c), 50 (d), 60 (e), 70 (f) wt%

Table 2. Ferroelectric properties of the PZT/PVDF composites

| *Samples* | | Composite compound ratio | Remnant Polariation ($P_r$) μC/cm² | Coercive field ($E_C$) kV/cm |
|---|---|---|---|---|
| *No.* | *Name of products* | | | |
| *1* | F16 | 0 | 2.1 | 9.3 |
| *2* | F-P30 | 30 | 5.1 | 5.2 |
| *3* | F-P40 | 40 | 5.4 | 5.1 |
| *4* | F-P50 | 50 | 6.3 | 4.9 |
| *5* | F-P60 | 60 | 7.7 | 5.0 |
| *6* | F-P70 | 70 | 8.2 | 5.0 |

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The results thus suggested that distribution of PZT granules in PVDF matrix played a significant role in controlling of ferroelectric behavior [5, 7, 15-20, 25, 35].

- *Piezoelectric measurements*

An attempt has been made to plot the predicted and experimentally observed variation in $d_{33}$ values with the volume fraction of PZT and it is illustrated in Fig. 9a. The $d_{33}$ values were calculated by using the equation (3) which is given by the Furukawa model [2, 9, 11-13, 18-23, 32-34, 36].

$$d = \frac{15x}{(2+3x)(1-x)} \frac{\varepsilon_p}{\varepsilon_c} d_c \qquad (3)$$

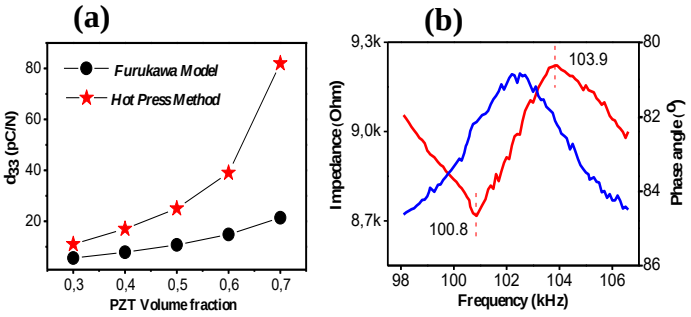


Figure 9 (a) Comparison of experimental and predicted values of $d_{33}$ with PZT volume fraction, (b) Impedance spectra of 0-3 composites (F-P70) with 1 mm thick pellet at room temperature.

Where $\varepsilon_p$, and $\varepsilon_C$ are the dielectric constant of polymer and ceramic phase respectively, and $d_C$ is the piezoelectric charge constant of ceramic.

The experimental as well as calculated $d_{33}$ values were found to be rising gradually with an increase in the ceramic phase of the composite for the PZT volume fraction up to 0.7. The experimental value of $d_{33}$ tends to change abruptly beyond 0.5 volume fraction due to the presence of higher ceramic content in the composite. This attributes to the orientation of the dipoles in the composite due to the more reinforcement of ceramic particles. Besides, an Agilent 4396B impedance analyzer is used to characterize the resonant frequency and quality factor behavior.

## IV. CONCLUSION

In this study, PZT-PVDF composites with 0–3 connectivity was successfully fabricated from a series of PZT volume fractions from 0.3 to 0.7 by hot press method. The heat treatment method can obviously enhance the content of tetragonal phase crystalline in piezoelectric ceramics granular, decrease the defect between ceramic phase and polymer phase and reduce the porosity. Experimental density values of the composites were found to be harmonized with calculated density values. SEM observations revealed a homogeneous mixture of PZT-PVDF phases.The dielectric constant values obtained in this work were found to be analogous to Furukawa model for the PZT volume fraction up to 0.7. The piezoelectric strain coefficient has been incremented with a raise in volume fractions of PZT but the Furukawa model fails to consider the experimental variations at higher volume fractions.

The values of piezoelectric strain coefficient, voltage coefficient and ferroelectric behavior obtained in this work revealed that it may be more suitable for piezoelectric devices, sensors, force transducer, and so on with technological applications [8, 22, 26, 27, 30, 32, 36, 39, 40].

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